Performance of Advanced Automotive Fuel Cell Systems with Heat Rejection Constraint

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Keywords

Polymer electrolyte fuel cells; Heat rejection; Mass transfer in cathode catalysts; Oxygen reduction reaction kinetics; Stability and durability

Submitted to the Journal of Power Sources

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Performance of Advanced Automotive Fuel Cell Systems with Heat Rejection Constraint

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Abstract

Although maintaining polymer electrolyte fuel cells (PEFC) at temperatures below 80°C is desirable for extended durability and enhanced performance, the automotive application also requires the PEFC stacks to operate at elevated temperatures and meet the heat rejection constraint, stated as $Q/\Delta T < 1.45 \text{ kW}/^\circ\text{C}$, where Q is the stack heat load for an 80-kW_e net power PEFC system and ΔT is the difference between the stack coolant temperature and 40°C ambient temperature. We have developed a method to determine the optimum design and operating conditions for an automotive stack subject to this $Q/\Delta T$ constraint, and illustrate it by applying it to a state-of-the-art stack with nano-structured thin film ternary catalysts in the membrane electrode assemblies. In the illustrative example, stack coolant temperatures >90°C, stack inlet pressures >2 atm, and cathode stoichiometries <2 are needed to satisfy the $Q/\Delta T$ constraint in a cost effective manner. The reference PEFC stack with 0.1 mg/cm² Pt loading in the cathode achieves 753 mW.cm⁻² power density at the optimum conditions for heat rejection, compared to 964 mW.cm⁻² in the laboratory cell at the same cell voltage (663 mV) and pressure (2.5 atm) but lower temperature (85°C), higher cathode stoichiometry (2), and 100% relative humidity.

Keywords: Polymer electrolyte fuel cells; automotive application; heat rejection; kinetic and mass transfer losses.

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1. Introduction

Durability and cost are regarded as the major barriers to mass commercialization of polymer electrolyte fuel cells (PEFC) for propulsion of light duty vehicles [1]. The current durability targets include 5,000 h of driving with less than 10% loss of performance [2]. Operating automotive fuel cells at temperatures below 70-80°C partially alleviates many of the durability concerns. For example, under rapidly varying potentials typical of automotive duty cycles, growth of Pt particles in the cathode catalyst layer, and the associated loss of electrochemical surface area (ECSA) and the catalyst activity for the oxygen reduction reaction (ORR), have been recognized as major degradation mechanisms [3]. The growth of the Pt particles is faster at higher temperature due to the enhanced solubility of Pt in the electrolyte and the accelerated rate of Pt particle coalescence/agglomeration [4, 5]. Catalyst durability is further exacerbated as Pt loading in the cathode is reduced to levels ($<0.1 \text{ mg.cm}^{-2}$) needed to approach the cost targets. Mass transfer issues at high current densities arise as the Pt loading in the cathode catalyst layers is reduced, and become even more prominent as the catalyst degrades and the ECSA decreases [6]. Similarly, even though the durability of the current generation of perfluorosulfonic acid (PFSA) membranes has greatly improved with chemical stabilization and mechanical reinforcement, the chemical stability of the membranes deteriorates under hot and dry conditions [7]. Finally, loss of hydrophobicity of the gas diffusion layer (GDL) is faster at higher temperatures and water transport can become an issue in the affected membrane electrode assemblies (MEAs) at high current densities [8].

To be competitive with the incumbent internal combustion engines (ICEs), fuel cells must operate over a wide range of ambient temperatures and relative humidities. The Fuel Cell Technical Team of the U.S. DRIVE Partnership that includes car companies conducted a study

analyzing the dependence of stack power density and heat rejection on stack operating temperature. They concluded that a viable automotive fuel cell system must have the ability to reject the stack waste heat (Q) at rated power and 40°C ambient temperature [2]. This requirement has been expressed as a constraint that a nominal 90-kW_e PEFC stack should have $Q/\Delta T$ less than 1.45 kW/°C, where ΔT is the initial difference between the stack coolant outlet temperature (T_c) and the ambient temperature (T_a). As will be shown in this paper, the $Q/\Delta T$ constraint implicitly requires that the PEFC stack be able to operate at coolant temperatures above 90°C. This constraint has been enforced to maintain the radiator size and frontal area in fuel cell systems to dimensions typical of passenger vehicles using ICEs for propulsion. The radiator size and frontal area are important parameters that affect packaging, cost, performance and drivability of light duty vehicles. Some of the first-generation fuel cell vehicles had oversized radiators and additional radiators in the front wheel housing. It is understood that in normal drive cycles, only for a limited time will the stack be challenged to $Q/\Delta T$ approaching 1.45 kW/°C. According to some studies, the stacks in light duty vehicles will be exposed to temperatures exceeding 90°C for less than ~1% of the 5,000 h required lifetime [9, 10].

The $Q/\Delta T$ constraint replaces the previous 55% stack efficiency target at rated power while avoiding the need to impose an additional target for stack temperature at rated power. The stack heat load is implicitly related to the cell voltage and, hence, the stack efficiency. As a reference, consider a state-of-the-art 90-kW_e stack that operates at 0.6 V cell voltage and 80°C for ~51% stack efficiency and $Q/\Delta T = 2.44$ kW/°C. The 1.45 kW/°C target can be met by decreasing Q , or increasing ΔT , or doing both. For lower Q , the stack efficiency can be raised by operating the stack at higher cell voltage. At 80°C, the required cell voltage for $Q/\Delta T = 1.45$ kW/°C is 0.76 V; the resulting power density at this high a cell voltage is likely too small (bulky and expensive

stack) for automotive application. For higher ΔT , the stack must be operated at higher temperature. At 0.6 V, the required operating temperature to meet the $Q/\Delta T$ target is 107°C; membrane stability, electrocatalyst durability and cell humidification are problematic at this high an operating temperature. The third option is to raise both the cell voltage and the operating temperature. For example, $Q/\Delta T = 1.45 \text{ kW}/^\circ\text{C}$ target can be met by raising the cell voltage to 0.663 V and the operating temperature to 95°C. Thus, even though the exact value of the appropriate $Q/\Delta T$ constraint may be debatable, the constraint does provide a useful metric for assessing the suitability of PEFC stacks for light-duty vehicles and offers a logical way of evaluating different options available for improving their automotive worthiness.

The purpose of this study is to discuss the impact of the $Q/\Delta T$ constraint on the performance and cost of automotive PEFC stacks and systems. The study uses state-of-the-art nanostructured thin film (NSTF) ternary catalyst electrodes for making MEAs. Section 2 describes the experimental characterization of the performance of 50-cm² cells fabricated using these MEAs under a wide range of operating conditions. Sections 3 and 4 describe the development of a cell model and its validation using the data collected in Section 2. In Section 5, the validated cell model is used in a reference PEFC system with all the relevant balance-of-plant (BOP) components to analyze the cost and performance of PEFC systems subject to the $Q/\Delta T$ constraint.

2. Experimental

Multiple 50-cm² active area single cells were assembled with a 24- μm membrane (850 equivalent weight), the Pt_{0.68}Co_{0.30}Mn_{0.02} ternary NSTF catalyst, and 3M gas diffusion layers (GDL) into a Fuel Cell Technologies test cell containing quad serpentine flow fields. The GDL consisted of a backing paper to which was applied a hydrophobic treatment and an MPL (micro-

porous layer). All cells had a Pt loading of 0.05 mg.cm^{-2} in the anode. Duplicate cells had nominal Pt loadings of 0.05, 0.1, 0.15 and 0.2 mg.cm^{-2} in the cathode; the actual Pt loadings in the cathode were 0.054, 0.103, 0.146 and 0.186 mg.cm^{-2} . The cells were conditioned using a “thermal cycling” process (TC), described in detail in Steinbach et al [11], which consisted of repeated temperature and voltage cycles over a period of 2-3 days until stable performance was reached.

The experimental campaign was organized as three classes of tests with reference H_2 and air inlet pressures of 1.5 (P15), 2.5 (P25) and 3 atm (P30). The reference H_2 and air stoichiometries (SR) were 2, i.e., H_2 and O_2 utilizations were 50%. P15 tests had 80°C reference cell temperature and 65°C inlet dew point temperature. P15 and P25 tests were conducted on cells with 0.1 mg.cm^{-2} nominal Pt loading in the cathode. P25 tests had 85°C reference cell temperature and 65°C inlet dew point temperature. P30 tests were conducted on cells with 0.15 mg.cm^{-2} nominal Pt loading in the cathode, 90°C reference cell temperature, and 64°C reference inlet dew point temperature.

Table 1 summarizes the test matrix for P15, P25 and P30. Eight series of tests were designed to investigate the effects of operating temperature, pressure, relative humidity, cathode stoichiometry (SR_c), anode stoichiometry (SR_a), start up from cold, and idling conditioning. An additional series of tests was run to investigate the effect of Pt loading in cathode. The H_2 and air streams had the same inlet dew point temperature in all tests. Unless explicitly stated otherwise, SR_c and SR_a were held at 2 in all tests.

Series 1 tests (T Series) varied the cell temperature from 75°C to 90°C at P15 and P25 and 75°C to 95°C at P30 reference conditions. The inlet dew points were pre-determined as a function of pressure, temperature and anode/cathode stoichiometry to maintain 100% relative humidity (RH) at cell exit. In all tests, humidification water was injected directly into the anode and cathode

feed streams to reach the set dew points. Also, the outlet RH was not measured but was estimated from the cell operating conditions.

Series 2 tests (P Series) varied the inlet pressure from 1 to 2.5 atm in P15 and P25 and from 1.5 to 3 atm in P30. The inlet pressure was higher than listed in Table 1 if the target pressure could not be reached even with the backpressure valve completely open to the ambient. As in Series 1 tests, the inlet dew points were adjusted to maintain 100% RH at cell exit. The P15 and P25 tests are listed together implying that the data are taken at the P15 reference temperature, 80°C, but the pressure extends to the P25 reference pressure, 2.5 atm.

Series 3 tests (RH Series) varied the inlet dew points from 50 to 80°C with the cell temperatures and pressures at reference conditions for P15, from 50 to 85°C in P25 and from 50 to 90°C in P30.

Series 4 tests (Pt Series) were conducted on cells with different Pt loadings in the cathode. These cells were only operated at P15 reference operating conditions.

Series 5 tests (SRc Series) varied the O₂ stoichiometry in P15, P25 and P30 tests from 1.5 to 5 with the inlet dew points adjusted to maintain 100% RH at cell exit ($SR_a = 2$).

Series 6 tests (SRa Series) varied the H₂ stoichiometry in P15, P25 and P30 tests from 1.2 to 5 with the dew points held at the reference values ($SR_c = 2$).

Series 7 tests (LT Series) were designed to investigate the cell operation during warm-up at 30, 45, and 60°C with dry feeds at P15 and P25 operating pressures and at 30, 45, 60 and 70°C at P30 operating pressures.

Series 8 tests (ID Series) were designed to investigate cell operation during idling at low pressures and high O₂ stoichiometries. The data were obtained only at P15 operating cell temperature and 100% inlet RH.

As in earlier work [12], the polarization curves for the eight series of tests were obtained using Galvanodynamic scans (GDS), starting near the open circuit at 0.02 A.cm⁻², increasing the current density in steps until either 2 A.cm⁻² was reached or the cell voltage decreased to 0.40 V, followed by an upscan back to 0.02 A.cm⁻². The cell voltages were measured at ten current steps per decade but the maximum step size was limited to 0.1 A.cm⁻². The cell was held for 120 s at each current step and the cell voltage was recorded every 5 s. Also, the cell resistance was determined every 5 s by on-line AC impedance measurements between 10 and 1 kHz, from which the high frequency resistance (HFR) was estimated by fitting to a Randle's cell model. The cell voltages reported in this are the measured values averaged over the last 30 s at the set current densities.

Prior to the start of the experiments, the electrochemical surface areas (ECSA) were determined by cyclic voltammetry (CV) between 0.085 and 0.65V (vs. RHE) at 100 mV.s⁻¹ scan rate, with H₂/N₂ at 70°C, 70°C dew points and ambient pressure. ECSA values were determined by averaging the oxidative and reductive H_{UPD} peak areas from 100 CVs and assuming a 210 μC.cm-Pt⁻² charge-to-area conversion factor.

Hydrogen crossover current density and cell short resistance were determined by measuring the plateau currents between 0.4 - 0.6 V cell potentials with saturated H₂/N₂ at 1.5 atm total pressure and 80°C cell temperature.

The mass activities of Pt in each cell were measured in H₂/O₂ at 80°C, 1-atm reactant H₂ and O₂ pressures and 100% RH. The measurement was made by first reducing the cathode with a 15-

min hold under H₂/N₂ at ~0.10 V, switching the cathode to O₂ and immediately holding the cathode potential at 0.90 V for 20 minutes. The mass activity values listed in Table 2 are the current densities, adjusted for H₂ crossover and shorting currents, at a cell voltage of 0.9 V after 17.5 minutes.

3. Theoretical

The starting point of the analysis is to represent the cell voltage (E) in terms of the Nernst potential (E_N), Ohmic drop due to the high frequency resistance (iR_Ω), cathode activation overpotential for the oxygen reduction reaction (η_a^c), mass transfer overpotential in cathode (η_m^c), anode overpotential for the hydrogen oxidation reaction (η_a^a), and mass transfer overpotential in anode (η_m^a) as

$$E = E_N - iR_\Omega - \eta_a^c - \eta_m^c - \eta_a^a - \eta_m^a \quad [1]$$

$$E_N = E_0 + \frac{RT}{nF} \ln\left(\frac{P_{H_2}^a (P_{O_2}^c)^{1/2}}{\phi_m^c}\right) \quad [1a]$$

where E_0 is the standard potential for the fuel cell reaction assuming that the product water is liquid, ϕ_m^c is the relative humidity (1 for 100% RH) in the cathode catalyst layer (CCL), and n equals 2.

$$E_0 = 1.23 - 0.000833(T - 298) \quad [1b]$$

In Eq. (1a), H₂ and O₂ partial pressures are evaluated in the anode and cathode gas channels

Following Ref. [12], the overpotential for the oxygen reduction reaction (ORR) on PtCoMn/NSTF catalyst is estimated from the following Tafel equation developed from the H₂/air polarization data at low current densities.

$$i + i_x = i_0^c A_{Pt}^c L_{Pt}^c P_{O_2}^\gamma (\phi_m^c)^\beta \exp\left(\frac{\alpha_c nF}{RT} \eta_a^c\right) \quad [2]$$

$$i_0^c = i_{0r}^c \exp\left[-\frac{E_{O_2}}{R} \left(\frac{1}{T} - \frac{1}{T_R}\right)\right] \quad [2a]$$

where i_x is the hydrogen cross-over current density. Table 2 lists the kinetic parameters for ORR kinetics on the ternary catalyst.

The overpotentials for the hydrogen oxidation reaction (HOR) on PtCoMn/NSTF catalyst are evaluated from the following Butler-Volmer equation [13] developed using data obtained in H_2 pump tests.

$$i = i_0^a A_{Pt}^a L_{Pt}^a P_{H_2} \left[\exp\left(\frac{\alpha_a nF}{RT} \eta_a^a\right) - \exp\left(-\frac{\alpha_c nF}{RT} \eta_a^a\right) \right] \quad [3]$$

$$i_0^a = i_{0r}^a \exp\left[-\frac{E_{H_2}}{R} \left(\frac{1}{T} - \frac{1}{T_R}\right)\right] \quad [3a]$$

Table 2 also includes the kinetic parameters for HOR on the ternary catalyst. The anode mass transfer overpotentials are small when the cell is operated with pure H_2 as fuel and are neglected in this work.

3.1 Cell Model

A simple one-dimensional model was adopted for estimating the local values of i , P_{O_2} , P_{H_2} and Φ in the counter-flowing anode (x direction) and cathode (-x direction) streams. The model solves the following set of equations for anode and cathode pressures and for molar flow rates ($\dot{N}$) of H_2 , O_2 and H_2O (subscript w) in the anode (superscript a) and cathode (superscript c) channels considering the source terms for the electrochemical reactions and water transport flux (J_w^m) across the membrane (superscript m) by electro-osmotic drag and diffusion (normal direction y).

$$\frac{dP^c}{dx} = \frac{\Delta P^c}{L} \quad [4]$$

$$\frac{dP^a}{dx} = -\frac{\Delta P^a}{L} \quad [4a]$$

$$\frac{d\dot{N}_{O_2}}{dx} = \frac{i + i_x}{4F} W_m \quad [5]$$

$$\frac{d\dot{N}_w^c}{dx} = -\left(\frac{i + i_x}{2F} + J_w^m\right) W_m \quad [5a]$$

$$\frac{d\dot{N}_{H_2}}{dx} = -\frac{i + i_x}{2F} W_m \quad [5b]$$

$$\frac{d\dot{N}_w^a}{dx} = -J_w^m W_m \quad [5c]$$

For expediency, the electro-osmotic drag coefficient (n_d) and the water diffusion coefficient (D_ϕ , $10^{-10}m^2/s$) were taken as function of average water activities across membrane ϕ_m , so that the following equation obtains for the water transport flux J_w^m across the membrane.

$$J_w^m = n_d \frac{i}{F} - \frac{D_\phi}{\delta_m} \frac{P_s(\phi_m^c - \phi_m^a)}{RT} \quad [6]$$

$$n_d = \begin{cases} 0.137 + 0.056e^{-1.12\phi_m^4 + 14.26\phi_m^3 + 7.95\phi_m^2 + 4.94\phi_m} & \phi_m < 1 \\ 1 & \phi_m \geq 1 \end{cases} \quad [6a]$$

$$D_\phi = \begin{cases} 0.28\phi_m^3 - 0.12\phi_m^2 + 0.33\phi_m & \phi_m < 1 \\ 0.5 & \phi_m \geq 1 \end{cases} \quad [6b]$$

where δ_m is the membrane thickness and λ is the water uptake in the membrane. The profiles of water vapor mole fraction across the anode and cathode GDLs were calculated in terms of the mass transfer coefficients (h_w^a and h_w^c) that are related to the diffusivities.

$$J_w^m = h_w^a \frac{P_s(\phi^a - \phi_m^a)}{RT} \quad [7]$$

$$h_w^a = \frac{D_w^a}{\delta_d^a} \left(\varepsilon_d^a \frac{T}{298} \right)^{1.5} \frac{1}{P^a} \quad [7a]$$

$$J_w^m + \frac{i + i_x}{2F} = h_w^c \frac{P_s(\phi_m^c - \phi^c)}{RT} \quad [7b]$$

$$h_w^c = \frac{D_w^c}{\delta_d^c} \left(\varepsilon_d^c \frac{T}{298} \right)^{1.5} \frac{1}{P^c} \quad [7c]$$

In Eq. (7a), the diffusion coefficient of water vapor in H_2 (D_w^a) at the reference conditions of 1 atm and 298 K is taken as $0.52 \times 10^{-4} \text{ m}^2 \cdot \text{s}^{-1}$. Similarly, the diffusion coefficient of water vapor in air (D_w^c) at the same reference pressure and temperature is taken as $0.256 \times 10^{-4} \text{ m}^2 \cdot \text{s}^{-1}$.

Reference [12] should be consulted for more details concerning the relationships between partial pressures and molar flow rates, between relative humidity and water vapor mole fraction, and between partial pressure of water vapor and the saturation pressure (P_s). Reference [12] also provides the functional dependence of λ on ϕ and the empirical correlations for pressure drops in anode and cathode gas channels.

3.2 Heat Transfer Model

We determine the MEA temperature by combining the energy equations for the anode and cathode flows and assuming that the anode electrode, cathode electrode, membrane, and anode/cathode gas flows are isothermal. The equation is written compactly in terms of enthalpy (H) that includes the heat of formation and the latent heat of condensation. The summation is performed over i species that make up the anode and cathode gases and implicitly accounts for the heat of reaction for $H_2 + 1/2 O_2 = H_2O$.

$$\frac{d}{dx} \left(\sum_i (\dot{N}_i^c M_i H_i^c) \right) - \frac{d}{dx} \left(\sum_i (\dot{N}_i^a M_i H_i^a) \right) = \dot{Q}_w W_m + i E W_m \quad [9]$$

We determine the coolant temperature profile by considering the heat transfer between the coolant and the MEA.

$$\dot{m}_w C_{pw} \frac{dT_w}{dx} = \dot{Q}_w W_m \quad [10a]$$

$$\dot{Q}_w = U(T - T_w) \quad [10b]$$

3.3 Cathode Mass Transfer Overpotential Model

For applicability to hotter and drier conditions, we analyzed the available polarization data to develop a rational model for mass transfer overpotentials in ternary PtCoMn/NSTF catalysts. The approach was to first define a limiting current density (i_L) at which the mass transfer overpotential equals a set value of 0.3 V. This limiting current density was determined from the cell polarization data and was correlated as a function of the operating pressure (P), temperature (T), oxygen mole fraction (X_{O_2}), relative humidity (ϕ) and catalyst surface area ($A_{Pt}^c L_{Pt}^c$). An elaborate procedure was formulated to extract the local value of i_L from the polarization data with finite stoichiometry and varying P, X_{O_2} , ϕ , L_{Pt}^c and current density.

$$i_L = i_L^r(P) f_X f_T f_\phi f_C \quad [11]$$

$$i_L^r(P) = 0.2767 + 0.5228P - 0.0165P^2 \quad [11a]$$

$$f_X = 20X_{O_2} - 40X_{O_2}^2 \quad [11b]$$

$$f_T = 1.224 \times 10^{-4} (T - T_r)^3 - 8.3102 \times 10^{-4} (T - T_r)^2 + 6.7383 \times 10^{-4} (T - T_r) + 1 \quad [11c]$$

$$T_r = \begin{cases} 353K & P = 1.5 atm \\ 358K & P = 2.5 atm \\ 363K & P = 3.0 atm \end{cases}$$

$$f_{\phi} = \begin{cases} -0.352(\phi_m^c)^3 + 0.427(\phi_m^c)^2 + 1.012\phi_m^c - 0.007 & P = 1.5 \text{ atm} \\ 1.366(\phi_m^c)^4 + 4.121(\phi_m^c)^3 - 3.713(\phi_m^c)^2 + 1.771\phi_m^c - 0.058 & P = 2.5 \text{ atm} \\ -0.053(\phi_m^c)^3 - 0.296(\phi_m^c)^2 + 1.162\phi_m^c + 0.053 & P = 3.0 \text{ atm} \end{cases} \quad [11d]$$

$$f_c = 0.0151A_{Pt}^c L_{Pt}^c + 0.8475 \quad [11e]$$

The second step was to correlate the mass transfer overpotential (η_m^c) in terms of the reduced current density (i/i_L), i.e., the current density (i) normalized by i_L . As above, an elaborate procedure was formulated to extract the local value of η_m^c from the polarization data with finite stoichiometry and varying P, X_{O_2} , T, ϕ and i/i_L . Higher order terms were included in the η_m^c correlation to correct for T, RH, and cathode stoichiometry (ψ).

$$\eta_m^c = 0.3(i/i_L)^{1/2} e^{-g_P g_T g_{\phi} g_{\psi} (1-i/i_L)} \quad [12]$$

$$g_P = 4.116 \quad [12a]$$

$$g_T = 228.704(T/T_r)^3 - 623.6916(T/T_r)^2 + 567.5707(T/T_r) - 171.594 \quad [12b]$$

$$g_{\phi} = \begin{cases} 1.269(\phi_m^c)^3 - 4.799(\phi_m^c)^2 + 4.755\phi_m^c - 0.043 & P = 1.5 \text{ atm} \\ 0.799(\phi_m^c)^3 - 2.804(\phi_m^c)^2 + 2.482\phi_m^c + 0.671 & P = 2.5 \text{ atm} \\ 0.659(\phi_m^c)^3 - 2.655(\phi_m^c)^2 + 2.971\phi_m^c + 0.277 & P = 3.0 \text{ atm} \end{cases} \quad [12c]$$

$$g_{\psi} = -0.646\psi^2 + 2.804\psi - 2.027 \quad [12d]$$

The above correlations for i_L and η_m^c are valid over the following range of variables: $1 < P < 3$ atm, $65 < T < 95^\circ\text{C}$, $0.05 < X_{O_2} < 0.2$, $0.3 < \phi_m^c < 1.9$, $1.5 < \psi < 5$, and $0.05 < i < 0.15 \text{ mg-Pt.cm}^{-2}$.

4. Model Validation

Figures 1a shows the increase in local limiting current density (i_L) with increase in O_2 mole fraction at operating pressures as calculated from Eq. (11) at 80°C and 100% RH at membrane-

cathode interface (ϕ_m^c). As a reference, O₂ mole fraction is 21% in dry air, and it decreases to ~7% at exit of stack operating at SR_c = 2, 1.5 atm, 80°C and 100% RH. Figure 1b quantifies the corresponding decrease in local mass transfer overpotentials (η_m^c) with increase in operating pressure at different current densities; we used Eq. (12) to calculate η_m^c for a cathode stoichiometry of 2 and O₂ mole fraction averaged over the cell inlet and exit values. Figure 1c compares the modeled polarization curves with experimental data for tests at different operating pressures, 80°C, SR_a = SR_c = 2, and 100% RH at cell exit. For this comparison, the model results were obtained by integrating Eqs. (4), (5), (9) and (10) using 20 nodes along the flow direction, with proper accounting of counter-flowing anode and cathode streams. We also used Eq. (2) for ORR kinetics, Eq. (3) for HOR kinetics, and Eqs. (11) – (12) for mass transfer overpotentials.

As further validation, we similarly present the effect of temperature on local limiting current density in Fig. 1d, local mass transfer overpotential in Fig. 1e, and polarization curves in Fig. 1f at 100% exit RH, 2.5 atm operating pressure and SR_c = 2. Under these conditions, Fig. 1e indicates that the mass transfer overpotentials are small, <10 mV, at current densities below 1.5 A.cm⁻². The small differences in isothermal cell performance in Fig. 1f at different temperatures are due to other direct and indirect effects of temperature on membrane resistance (R_Ω), ORR kinetic overpotentials (η_a^c) and P(O₂). Relative to the polarization curve at 85°C, the high current density performance is worse at 75°C because of greater mass transfer overpotentials and is slightly worse at 90°C because of smaller P(O₂). Overall, the modeled effect of temperature on polarization curves at 2.5 atm is in good agreement with the experimental data.

For brevity, we have not shown the equally good comparisons between the modeled and measured polarization curves for other series of tests with RH, anode/cathode stoichiometries, cathode Pt loadings as variables and different combinations of P, T, RH and stoichiometries.

5. Results and Discussion

The cell model was used to analyze the performance of an 80-kW_{net} reference fuel cell system (see the figure included in Table 3) with ternary PtCoMn/NSTF catalysts in the PEFC stack. The details of the system configuration are discussed in Refs. [14, 15] and will not be repeated here. Table 3 lists the important performance parameters of the components comprising the stack, fuel management system, air management system with a compressor-expander module (CEM), heat rejection system and water management system. In applying the model developed from a single 50-cm² cell to a large stack with >250 cm² active membrane area and >300 cells, we assumed a 10 mV cell-to-stack voltage degradation (ΔV_{cs}) at 1 A.cm⁻² current density. This allowance, amounting to 10 mΩ.cm² additional resistance, accounts for possible electrical losses due to channel-to-channel and cell-to-cell flow non-uniformities in large stacks. Also, whereas, the model has been developed by using data for isothermal cells, it can be applied to non-isothermal cells since it is based on local temperature, taken as the bipolar plate temperature which can be calculated from the local coolant temperature, coolant flow rate and heat transfer to the coolant.

Most of the results presented below are based on an optimization study that minimizes the fuel cell system cost subject to multiple constraints including $Q/\Delta T$ of 1.45 kW/°C. In this study, we use the cost correlations developed by James et al [16] for the subsystems in Table 3 at high volume manufacturing, 500,000 units/year, and 1500 \$/tr-oz. price of Pt. Table 4 lists the correlations for the cost of the various subsystems as a function of the important parameters: active membrane area, Pt loading and Pt price for stack with metal bipolar plates and NSTF

electrocatalyst; pressure and air flow rate for the air management system with high-speed centrifugal compressor and expander; heat loads and temperature difference for the thermal management system; humidifier membrane area and pre-cooler heat load and temperature differential for the thermal management system; and the operating pressure for the fuel management system. The constants and coefficients account for the costs of all components (e.g., GDLs, bipolar plates, end plates, demister, ejector, balance-of-plant, etc.) for which the costs do not explicitly appear in the correlations.

Figure 2 presents illustrative results from an initial parametric study on the dependence of stack and water management system costs on the coolant exit temperature. The results are for specified Pt loading in anode and cathode ($0.05/0.1 \text{ mg.cm}^{-2}$), stack inlet pressure (2.5 atm), rise in coolant temperature (10°C), ambient temperature (40°C), and anode/cathode stoichiometries (2/2). Two sets of results are included in Fig. 2: one in which the exit cathode RH is restricted to 100%, and the other in which it is optimized for minimum FCS cost. For fixed $Q/\Delta T$, raising the coolant temperature results in higher ΔT , higher allowable Q , and, therefore lower cell voltage. The stack cost decreases sharply because of higher stack power density at lower cell voltages. However, raising the exit coolant temperature from 85 to 95°C for 100% exit RH leads to more than 10-fold increase in humidifier membrane area (<0.4 to 4.7 m^2), so that there is an optimum coolant temperature ($\sim 91^\circ\text{C}$) for which the system cost is the lowest. Without the 100% exit RH restriction, the stack cost decreases even faster upon raising T_c while the increase in humidifier cost is slower [17], so that the minimum system cost is at the highest coolant temperature considered in this study. For the parameters of this study, the optimum RH at stack outlet decreases from 97.5% to $\sim 80\%$ as T_c is raised from 85°C to 95°C .

5.1 Optimization Study with $Q/\Delta T$ Constraint

A multi-variable optimization study was conducted to determine the combination of coolant temperature, relative humidity at stack exit and the cell voltage for minimum system cost, subject to $1.45 \text{ kW}/^\circ\text{C}$ $Q/\Delta T$ constraint for specified Pt loading in anode/cathode catalysts and 10°C rise in coolant temperature. The cathode stoichiometry and stack inlet pressure were treated as study parameters. The results from the study are summarized in Figs. 3-6.

Figure 3 summarizes the dependence of the optimum coolant exit temperature and the cell voltage on cathode stoichiometry and the stack inlet pressure for 100% exit RH. For fixed RH at stack exit, raising the cathode stoichiometry or lowering the inlet pressure increases the required water transport in the cathode humidifier. Since the humidification load is also a strong function of temperature, Fig. 3a shows that the optimum coolant exit temperature for minimum system cost must decrease under these conditions. The results in Fig. 3b indicate that with the $Q/\Delta T$ constraint, the cell voltage at the optimum coolant exit temperature has to increase to compensate for the additional CEM power at higher cathode stoichiometry. We conclude that low stack inlet pressures (1.5 atm) may not be acceptable because the optimum coolant exit temperature is restricted to $<81^\circ\text{C}$, ΔT to $<41^\circ\text{C}$ and Q to $<60 \text{ kW}$, so that the required cell voltage has to be $>760 \text{ mV}$. Also, the cathode stoichiometry needs to be <2 to keep the cell voltage below 700 mV at 2.5 atm stack inlet pressure.

The results indicate that, over the range of cathode stoichiometries (SR_c) and stack inlet pressures investigated in this study, the required cell voltage and the system cost ($\$/\text{kW}_e$) are lowest at $SR_c = 1.5$. Therefore, only the results for $SR_c = 1.5$ are included in Figs. 4-6.

Consistent with the results in Fig. 2, Figs. 4a and 4b show that the differences in system cost and Pt content (g/kW_e) between saturated ($RH_c = 100\%$) and un-saturated ($RH_c \leq 100\%$) cathode

exits are large at low operating pressures and diminish at high operating pressures. The optimum cathode exit RH is $>86\%$ if the stack inlet pressure is higher than 2.5 atm and it is $<73\%$ if the inlet pressure is 1.5 atm. At the optimum conditions, both the exit anode and cathode streams are slightly supersaturated ($RH > 100\%$) at 3 atm stack inlet pressure.

The relative humidity affects the system cost and performance at many levels. It determines the size and cost of the humidifier because the required water vapor transport (WVT) is directly proportional to RH for given pressure and temperature. For specified RH and temperature, WVT in the humidifier is inversely proportional to pressure. Whereas, raising the RH improves the membrane conductivity (smaller Ohmic resistance), it decreases the Nernst potential because of lower $P(H_2)$ and $P(O_2)$ and higher $P(H_2O)$. More importantly, the resultant drop in oxygen concentration at higher RH leads to much larger kinetic and mass transfer overpotentials, especially at low pressures.

The system cost in Fig. 4a is lowest at 3-atm stack inlet pressure, but the cost saving is small compared to 2.5-atm inlet pressure. Although the stack has to generate additional power to compensate for the greater CEM parasitic power as the inlet pressure is raised, the system cost and Pt content are lower. Figure 4a clearly indicates that, for 100% RH at cathode exit, stack inlet pressures below 2 atm are unacceptable from the standpoints of system cost (>90 $\$/kW_e$) and system performance (>0.6 g-Pt/ kW_e).

Figure 4c shows that the cell voltage required to meet the $Q/\Delta T$ constraint at 1.5 atm stack inlet pressure is 94 mV lower if the stack is operated hotter ($92^\circ C$ vs. $82^\circ C$) and drier (73% RH vs. 100% RH). The required cell voltage with exit RH restricted to 100% decreases sharply as the stack inlet pressure is raised from 1.5 atm to 3 atm. Without the 100% RH restriction, the required cell voltage decreases by ~ 20 mV as the stack inlet pressure is raised from 1.5 to 2 atm,

and then increases as the inlet pressure is raised to 3 atm because of the increase in power consumed by the CEM. The stack power density in Fig. 4d is $<500 \text{ mW.cm}^{-2}$ for stack operating pressures below 2.2 atm for 100% exit RH and 1.7 atm for unrestricted RH. The difference in power densities for restricted and unrestricted RH diminishes as the stack inlet pressure is raised above 2.5 atm since, even without the RH restriction, the optimum cathode exit RH approaches and exceeds 100% at 3 atm stack inlet pressure.

Figure 4e presents the system efficiency on a lower heating value (LHV) basis that the FCS must have in order to meet the $Q/\Delta T$ constraint at rated power. Imposing the heat rejection constraint makes the system efficiency at rated power a function of the operating pressure. The required efficiency is lower (desired result) at higher stack inlet pressures or if the cathode exit RH is less than 100% (although there are durability implications). Note that the anode outlet may contain condensed water; the stack heat load in Fig. 4f includes this latent heat and the sensible heat loads due to rise in gas temperatures in addition to the waste heat generated (related to cell voltage) by the thermodynamic irreversibilities in the cell electrochemical reactions (i.e., cell overpotentials). With the maximum coolant temperature limited to 95°C (study assumption), the allowable stack heat load corresponding to $1.45 \text{ kW}/^{\circ}\text{C}$ $Q/\Delta T$ in Fig. 4f is restricted to 79.8 kW.

5.2 Optimum Pt Loading

A parametric study was conducted to determine the optimum Pt loading in the cathode catalyst for $SR_c = 1.5$. Figure 5a shows that the cathode with the smallest Pt loading considered in this study (0.054 mg.cm^{-2}) has the lowest Pt content but also the lowest power density (see Table 5 for 2.5 atm stack inlet pressure). Table 5 indicates that the cost of the balance-of-stack (BOS) components (membrane, bipolar plate, GDL) decreases from 65% to 46% as the cathode Pt loading is increased from 0.054 to 0.146 mg/cm^2 and the power density improves from 565 to

690 mW/cm². Increasing the Pt loading above 0.15 mg/cm² does not result in higher power density, suggesting that there may be a room to further optimize the electrode structure (e.g., adjusting whisker density, diameter and aspect ratio) for cathodes with such high Pt loadings. Figure 5b shows that the lower BOS cost nearly offsets the higher Pt cost so that there is less than 2% change in system cost as the Pt loading is increased from 0.054 to 0.103 mg/cm². Since low-Pt cathodes are known to have issues with durability and tolerance to contaminants, considering cost and durability, 0.103 mg/cm² is likely the optimum Pt loading for the ternary NSTF catalyst.

It is worth mentioning that the market price of Pt has fluctuated between \$1100 and \$1500 per tr-oz. The optimum Pt loading in cathode would be higher than 0.103 mg/cm² if the Pt price is \$1100/tr-oz rather than \$1500/tr-oz assumed in this study.

5.3 Q/ΔT Constraint

In our study, Q is the actual stack heat load (AQ) as calculated in the model considering variable P(O₂), P(H₂), P(H₂O), T, and current density along the flow directions. The model includes latent heat released by water condensation (if any) in the stack and the sensible heat transfer to the anode and cathode gases. Besides the cell voltage, AQ should depend on operating conditions such as the operating pressure, temperature, and anode/cathode stoichiometry, and rise in coolant temperature. The heat load in the Q/ΔT target (SN) is written in terms of voltage efficiency ($\eta_V = E / E_N$), defined as the ratio of cell voltage to the Nernst potential (E_N).

$$Q = P_s \left(\frac{1}{\eta_V} - 1 \right) = P_s \frac{(E_N - E)}{E}$$

where P_s is the stack gross power required for a fuel cell system that generates 80 kW_e net power; the suggested value of P_s is 90 kW. The target also suggests that, for simplicity, E_N be approximated as 1.25 V, independent of the stack operating conditions.

Figure 6 summarizes results from a study to investigate the effect of stack heat load estimates (AQ vs. SN) on the cost and performance of the reference system at different operating pressures. Both sets of results, labeled as $Q/\Delta T$ (AQ) and $Q/\Delta T$ (SN), are for optimized T_c and RH with specified stack inlet pressure (1.5-3 atm), cathode stoichiometry (1.5), rise in coolant temperature (10°C), and Pt loading in anode and cathode. The results show that, under most conditions, Q (SN) is an acceptable approximation to the actual stack heat load. Q (SN) grossly underestimates the heat load for conditions under which liquid water forms in the stack and significant amount of the latent heat of condensation is released, as at 3-atm stack inlet pressure.

5.4 Data Variability

As discussed in Section 2, an extensive data base was developed by running more than 130 polarization and electrochemical characterization tests on multiple cells. The reference conditions at 1.5 (P15), 2.5 (P25) and 3 atm (P30) were visited many times during different test series. Figure 7a shows the variability in polarization curves at 2.5-atm reference pressure in two of the eight cells (23102 and 23272); similar variations were also observed at 1.5 atm and 3 atm reference pressures. The inset in Fig. 7a identifies the test series in which the data were taken and the order in which the tests were conducted: Series 5 (SRc), 6 (SRa), 1 (T) and 3 (RH) for cell 23102, and Series 1, 5, 6 and 3 for cell 23272. Figure 7b indicates that the variability in cell voltage (ΔV) is a function of the current density and is higher at higher current density. We may regard ΔV as a measure of the recoverable losses since the test campaign is not long enough for manifestation of significant irrecoverable losses. In a different study, we confirmed that the

voltage losses in Fig. 7a can be recovered by reconditioning the cells by subjecting them to multiple thermal conditioning (TC) cycles. Nevertheless, the variability in cell voltage is representative of the performance variation that may be expected over drive cycles before the onset of permanent degradation mechanisms. The variability may be attributed to the accumulation of impurities and the initial state of the membrane at the start of a test series. The test data are likely to be more reproducible if, prior to taking polarization data, the cell is subjected to a standardized break-in and oxide-reduction protocol such as the one recommended in Ref. [2].

The model discussed in this paper is representative (REP) of the average performance in that the kinetic and transport parameters were determined by using the average of all available polarization data. We also developed a model for the best-of-class (BOC) performance in which we determined the kinetic and transport parameters using the only best polarization data under different conditions. Figures 7c and 7d compare the FCS cost and performance using the REP and BOC models. Table 6 shows ~10% improvement in power density (753 mW/cm²), Pt cost (10.8 kW_e) and stack cost (25.7 \$/kW_e) if the results for 2.5 atm stack inlet pressure are based on BOC rather than REP polarization data.

5.4 Impact of Q/ΔT Constraint

We conducted a systematic study to evaluate the impact of ΔV_{sc} and Q/ΔT constraint on cost and performance projections. We used the BOC model for this study because many of the cell polarization data reported in literature are equivalent to the BOC rather REP data.

Figure 8a presents stack power density at three different cell voltages needed to satisfy the Q/ΔT (SN) constraint at cathode stoichiometries of 2.5, 2 and 1.5, 2.5 atm stack inlet pressure, and 95°C coolant exit temperature. The Pt loadings, ΔT_c and anode stoichiometries are the same as in

Fig. 4. The required cell voltage is progressively higher as SR_c is raised because, for fixed 80- kW_e net system power, the stack has to produce additional power to compensate for the higher CEM parasitic losses at higher air flow rates. So to maintain the same Q ($Q/\Delta T$ fixed at 1.45 $kW/^\circ C$), the cell voltage has to increase for higher efficiency. For this reason, in spite of higher $P(O_2)$, the power density in Fig. 8a is smaller at $SR_c = 2.5$ than at $SR_c = 2$.

The leftmost cluster in Fig. 8a indicates that the modeled BOC power densities can reach 880-964 mW/cm^2 at 85°C and 100% exit RH at the cell level ($\Delta V_{sc} = 0$). These values are comparable to the BOC power densities measured in isothermal cell tests under similar conditions. Introducing 10 mV cell-to-stack voltage loss (cluster 2) results in 9.0-9.2% reduction in the stack power density. Raising the exit coolant temperature to 90°C (cluster 3) causes 10-26 mW/cm^2 (1.2-3.1%) reduction in power density. Further raising the coolant temperature to 95°C (cluster 4) to meet the $Q/\Delta T$ constraint results in an additional ~40 mW/cm^2 (4.5-5.0%) reduction in power density. Finally, relaxing the 100% exit RH constraint for minimum system cost further reduces the power density by 7-30 mW/cm^2 (0.9-3.9%).

Figure 8b presents the variation in FCS cost on introducing the 10 mV cell-to-stack voltage loss and as the coolant exit temperature is raised from 85 to 95°C and the 100% exit RH constraint is relaxed. Introducing 10 mV cell-to-stack voltage loss (cluster 2) results in 1.5-2 $$/kW_e$ increase in FCS cost. Raising the exit coolant temperature to 90°C (cluster 3) increases the costs of PEFC stack (lower power density) and water management system (higher WVT requirement) but reduces the costs of the thermal management system; the overall FCS cost increases by 2.1 $$/kW_e$ for $SR_c = 2.5$ and by 0.7 $$/kW_e$ for $SR_c = 2$, but slightly decreases by 0.3 $$/kW_e$ for $SR_c = 1.5$. Further raising the coolant temperature to 95°C (cluster 4) results in an additional 1.5-5.9 $$/kW_e$ increase in FCS cost. Finally, relaxing the 100% exit RH constraint reduces the FCS cost

by 0.5-5.5 $\$/kW_e$ because of saving in the costs of the water management system. The last cluster shows that the FCS cost is lowest for $SR_c = 1.5$ although the stack power density is highest in Fig. 8a for $SR_c = 2$.

Figure 8c presents the breakdown of the costs for conditions needed to satisfy the $Q/\Delta T$ constraint at minimum cost (cluster 5 in Figs. 8a and 8b). The stack cost is lowest for $SR_c = 2$, but the air, thermal and water management subsystems have the lowest cost for $SR_c = 1.5$. The air management system contributes significantly to the overall system cost; the projected cost of the air system is 45-55% of the stack cost.

6. Summary and Conclusions

We have investigated the impact of the heat rejection ($Q/\Delta T$) constraint on the design, performance and cost of automotive fuel cell systems for propulsion of light duty vehicles. The combined experimental and theoretical work was conducted on MEAs with nanostructured thin film (NSTF), ternary PtCoMn electrocatalysts and low Pt loadings in the cathode ($<0.20 \text{ mg-Pt}/\text{cm}^2$). The validated cell model was used in a reference PEFC system with all the relevant balance-of-plant (BOP) components to analyze the cost and performance of automotive systems subject to the $Q/\Delta T$ constraint. The main conclusions from this investigation are highlighted below.

- Meeting the $Q/\Delta T$ constraint requires that the stack be operated hotter (coolant exit temperature $> 90^\circ\text{C}$), drier (stack exit RH $<100\%$) and at elevated pressures (inlet pressures $>2 \text{ atm}$).
- Low stack inlet pressures (1.5 atm) and 100% cathode exit RH may not be acceptable because the coolant exit temperature is restricted to $<82^\circ\text{C}$, and the required cell voltage has to be $>760 \text{ mV}$, resulting in a power density $<160 \text{ mW}/\text{cm}^2$.

- The cathode stoichiometry needs to be less than 2 to keep the cell voltage below 700 mV at 2.5 atm stack inlet pressure. Within the range of stoichiometries investigated in this study, $SR_c = 1.5$ yields acceptable performance (i.e., low Pt content) at least cost.
- The optimum cathode exit RH is $>86\%$ if the stack inlet pressure is higher than 2.5 atm and it is $<73\%$ if the inlet pressure is 1.5 atm.
- The optimum Pt loading in the cathode ternary catalyst is between 0.1 and 0.125 mg.cm^{-2} although the Pt content is reduced at lower Pt loading (0.054 mg.cm^{-2}) and the power density is higher at higher Pt loading (0.146 mg.cm^{-2}).
- For the conditions of this study, the projected Pt content and cost of an optimized 80-kWe fuel cell system that meets the 1.45 kW°C constraint are 0.25 g/kW_e and 56.2 $\text{\$/kW}_e$. The performance and cost improve by $\sim 10\%$ if the projections are based on the best-of-class data rather than the representative data.
- Figure 8 illustrates the cascading manner in which imposing the heat rejection constraint and introducing 10 mV cell-to-stack voltage degradation results in the BOC power density decreasing from 964 mW.cm^{-2} at 673 mV, 85°C, 100% exit RH and $SR_c = 2$ ($Q/\Delta T = 1.8 \text{ kW}^\circ\text{C}$) to 753 mW.cm^{-2} at 663 mV, 95°C, 91.4% RH, and $SR_c = 1.5$ ($Q/\Delta T = 1.45 \text{ kW}^\circ\text{C}$).

Nomenclature

A	specific electrochemical surface area	D	water diffusion coefficient
E	cell voltage, potential or activation energy	F	Faraday constant
H	enthalpy	h	mass transfer coefficient
i	current density	J	water transport flux
L	length or Pt loading	M	molecular weight

$\dot{N}$	molar flow rate	n	number of electrons
P	pressure or power	Q	stack heat load
$\dot{Q}$	heat transfer flux	R	gas constant or resistance
T	temperature	U	heat transfer coefficient
W	width	ϕ	relative humidity
η	overpotential or voltage efficiency		

Subscripts and Superscripts

0	standard	a	activation, ambient or anode
c	cathode	d	electro-osmotic drag, diffusion layer
L	limiting	m	mass transfer or membrane
N	Nernst	R	reference
r	reference	s	saturation or stack
sc	cell to stack	w	water or coolant
X	mole fraction	x	cross-over or anode flow direction
α	symmetric factor	β	relative humidity dependence
δ	thickness	ε	porosity
ϕ	relative humidity	γ	O ₂ partial pressure dependence
Ω	Ohmic	ψ	cathode stoichiometry

Acknowledgments

This work was supported by the Fuel Cell Technologies Office of the U.S. Department of Energy's (DOE) Office of Energy Efficiency and Renewable Energy. Dr. Nancy Garland is the DOE Technology Development Manager for this work. Argonne is a DOE, Office of Science

Laboratory operated under Contract No. DE-AC02-06CH11357 by the UChicago Argonne, LLC.

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Tables

Table 1 Test matrix

Table 2 ORR and HOR kinetic parameters

Table 3 Summary of performance parameters of reference FCS components

Table 4 Parameters of cost correlations

Table 5 Summary of optimum Pt loading study for 2.5 atm stack inlet pressure

Table 6 Effect of variability in supporting data on stack cost and performance

Figures

Figure 1 Development and validation of correlations for limiting current density and mass transfer overpotentials a. Limiting current density (80°C) b. Mass transfer overpotential (80°C) c. Model validation (80°C) d. Limiting current density (2.5 atm) e. Mass transfer overpotential (2.5 atm) f. Model validation (2.5 atm)

Figure 2 Parametric study to illustrate the effect of the stack operating temperature on the costs of the fuel cell system and FCS components a. Dependence of stack and water management system (WMS) cost on coolant exit temperature b. Dependence of FCS cost on coolant exit temperature

Figure 3 Optimization study to determine operating conditions for minimum system cost subject to $Q/\Delta T$ constraint, exit RH restricted to 100% a. Dependence of optimum coolant temperature on cathode stoichiometry and stack inlet pressure b. Variation of cell voltage with cathode stoichiometry and stack inlet pressure

Figure 4 Optimized system cost and performance subject to $Q/\Delta T$ constraint, with and without 100% exit RH restriction a. System cost b. System performance c. Cell voltage d. Stack power density e. System efficiency f. Stack heat load

Figure 5 Study to determine the optimum Pt loading in the cathode ternary catalyst a. Pt content b. System cost

Figure 6 Impact of stack heat load estimate on system cost and performance a. System cost b. Pt content

Figure 7 Comparison of system cost and performance for representative (REP) and best-of-class (BOC) data a. Variability in polarization curves b. Variability in cell voltage from the mean c. System cost d. Pt content

Figure 8 Waterfall charts showing changes in power density and FCS cost due to cell-to-stack voltage degradation, rise in coolant temperature, and relaxation of 100% exit RH constraint a. BOC power density b. Systematic increase in FCS cost c. Cost breakdown for cluster 5 conditions

Table 1 Test matrix

P15 Series		$L_{Pi}(c)$ mg/cm ²	$L_{Pi}(a)$ mg/cm ²	T C	$T_{DP}(c)$ C	$T_{DP}(a)$ C	P atm	SR(c)	SR(a)
Reference	FC19478	0.1	0.05	80	65	65	1.5	2	2
Conditions	FC19453	0.1	0.05	80	65	65	1.5	2	2
Test Series	Tests	1	2	3	4	5	6	7	8
T Series	T(C)/ T_{DP} (C)	75/55	80/65	85/74	90/82				
P Series	P(atm)/ T_{DP} (C)	1/73	1.25/70	1.5/65	2/56	2.5/40			
RH Series	T_{DP} (C)	0	50	55	60	65	70	75	80
L_{Pi} Series	$L_{Pi}(c)$ (mg/cm ²)	0.05	0.1	0.15	0.2				
SR _c Series	SR(c)/ T_{DP} (C)	1.5/60	2/65	2.5/68	5/75				
SR _a Series	SR ₀ (a)	1.2	1.5	2	2.5	5			
Cold T Series	T(C)/ T_{DP} (C)	30/0	45/0	60/0					
Idle Operation	T(C)/P(atm)/SR(c)	80/1.05/10	80/1.1/5	80/1.2/2.5	80/1.3/1.75				
P25 Series		$L_{Pi}(c)$ mg/cm ²	$L_{Pi}(a)$ mg/cm ²	T C	$T_{DP}(c)$ C	$T_{DP}(a)$ C	P atm	SR(c)	SR(a)
Reference	FC23102	0.1	0.05	85	65	65	2.5	2	2
Conditions	FC23272	0.15	0.05	85	65	65	2.5	2	2
Test Series	Tests	1	2	3	4	5	6	7	8
T Series	T(C)/ T_{DP} (C)	75/25	80/50	85/65	90/75				
RH Series	T_{DP} (C)	50	55	60	65	70	75	80	85
SR _c Series	SR(c)/ T_{DP} (C)	1.5/52	2/65	2.5/70	5/79				
SR _a Series	SR(a)	1.2	1.5	2	2.5	5			
Cold T Series	T(C)/ T_{DP} (C)	30/0	45/0	60/0					
P30 Series		$L_{Pi}(c)$ mg/cm ²	$L_{Pi}(a)$ mg/cm ²	T C	$T_{DP}(c)$ C	$T_{DP}(a)$ C	P atm	SR(c)	SR(a)
Reference	FC26115	0.15	0.05	90	64	64	3	2	2
Conditions	FC25979	0.15	0.05	90	64	64	3	2	2
Test Series	Tests	1	2	3	4	5	6		
T Series	T(C)/ T_{DP} (C)	80/0	85/46	90/64	95/76				
P Series	P(atm)/T(C)/ T_{DP} (C)	1.5/80/65	2.5/85/65	3/90/64					
RH Series	T_{DP} (C)	50	60	64	70	80	90		
SR _c Series	SR(c)/ T_{DP} (C)	1.5/0	2/64	2.5/70	5/81				
Cold T Series	T(C)/ T_{DP} (C)	30/0	45/0	60/0	70/0				

Table 2 ORR and HOR kinetic parameters

a. ORR Kinetics ($T_R = 353\text{ K}$)							
L_{Pt}	A_{Pt}^0	i_{0r}	E_{ORR}	n	α_c^c	β	γ
mg.cm^{-2}	$\text{m}_{Pt}^2.\text{g}_{Pt}^{-1}$	mA.cm_{Pt}^{-2}	kJ.mol^{-1}				
0.1	9.8	0.1×10^{-3}	56	2	0.51	0.9	0.45
b. HOR Kinetics ($T_R = 353\text{ K}$)							
L_{Pt}	A_{Pt}^0	i_{0r}	E_{HOR}	n	α_a^a	α_c^a	
mg.cm^{-2}	$\text{m}_{Pt}^2.\text{g}_{Pt}^{-1}$	mA.cm_{Pt}^{-2}	kJ.mol^{-1}				
0.05	11.2	489	38.9	2	0.5	0.5	

Table 3 Summary of performance parameters of reference FCS components

PEFC Stack

- 1.5-3 atm at rated power
- 40-67% O₂ utilization (SR_c: 1.5-2.5)
- 50% H₂ consumption per pass
- Cell voltage at rated power: TBD
- 24- μ m 3M membrane at TBD temperature
- 3M ternary alloy: 0.1/0.05 mg-Pt/cm² on cathode/anode
- GDL: 235- μ m non-woven carbon fiber with MPL
- 1.1-mm metal bipolar plates, each with cooling channels
- 17 cells/inch

Fuel Management System

- Hybrid ejector-recirculation pump
- 35% pump efficiency
- 3 psi pressure drop at rated power

Air Management System

- Compressor-expander module
- Liquid-cooled motor
- Efficiencies at rated power: 71% compressor, 73% expander, 89.5% motor, 89.5% controller
- Turn-down: 20
- 5 psi pressure drop at rated power

Heat Rejection System

- Two circuits: 75-95°C HT, 10°C Δ T
65°C LT coolant, 5°C Δ T
- 55% pump + 92% motor efficiency
- 45% blower + 92% motor efficiency
- 10 psi pressure drop each in stack and radiator

Water Management System

- Membrane humidifier, TBD dew-point temperature at rated power

Table 4 Parameters of cost correlations

Component	Variable	Validation Range	Cost Model
Stack (300V)	A, cm ²	[63655, 124938]	$3.71 \times 10^{-5} (0.87A - 2.57) L_{Pt} C$ $+ 6.70 \times 10^{-3} A + 399.2$
	L _{Pt} , mg·cm ⁻²	[0.1, 0.8]	
	C, \$/tr-oz	[800, 2000]	
Air Management	P, atm	[1.5, 3.15]	$515.9 - 2.14P + 0.62P\dot{m}_{air}$
	$\dot{m}_{air}$, kg/h	[200, 400]	
Thermal Management	Q _{HT} , kW	[56, 109]	$(100.25Q_{HT} / \Delta T_{HT} + 190.9)$ $+ (302.17(Q_{LT} / \Delta T_{LT})^2 + 313.06Q_{LT} / \Delta T_{LT})$ $+ (8.36P^2 - 25.47P - 99.48PQ_{LT} / \Delta T_{LT})$ $+ 17.77$
	ΔT_{HT} , K	[38, 70]	
	Q _{LT} , kW	[17, 27]	
	ΔT_{LT} , K	[28, 70]	
Water Management	A, m ²	[0.32, 4.25]	$(-1.04A^2 + 48.01A + 16.6)$ $+ (588.69Q / \Delta T - 2.7)$
	Q, kW	[5.5, 10.9]	
	ΔT , K	[97, 100]	
Fuel Management	P, kW	[0.1, 0.25]	$(3801.97P^3 - 2967.73P^2 - 1573.1P - 87.8)$ $+ 241.3$
BOP			551.9

Cost variables

Stack: A: active membrane area; C: Pt price

Air management system: P: compressor discharge pressure; $\dot{m}_{air}$: air flow rate

Thermal management system: HT: high temperature radiator; LT: low temperature radiator;

Q: heat load; ΔT : T-T_a

Water management system: A: humidifier membrane area; Q: pre-cooler heat load

Fuel management system: P: blower power

Table 5 Summary of optimum Pt loading study for 2.5 atm stack inlet pressure

Pt Loading	mg/cm ²	0.054	0.103	0.146	0.186
Power Density	mW/cm ²	565	672	703	690
Pt Cost	\$/kW _e	9.8	12.1	14.8	18.2
Stack Cost	\$/kW _e	28.0	28.2	30.4	34.0

Table 6 Effect of variability in supporting data on stack cost and performance

		2.5 atm, 95°C		1.5 atm, 87°C	
		REP	BOC	REP	BOC
Cell Voltage	mV	662	662	697	697
Current Density	A/cm ²	1.02	1.14	0.57	0.64
Power Density	mW/cm ²	674	753	394	447
Pt Cost	\$/kW _e	12.1	10.8	19.9	17.5
Stack Cost	\$/kW _e	28.1	25.7	43.1	38.5

Figure 1 Development and validation of correlations for limiting current density and mass transfer overpotentials

(a) Dependence of stack and water management system (WMS) cost on coolant exit temperature

(b) Dependence of FCS cost on coolant exit temperature

Figure 2 Parametric study to illustrate the effect of the stack operating temperature on the costs of the fuel cell system and FCS components

(a) Dependence of optimum coolant temperature on cathode stoichiometry and stack inlet pressure

(b) Variation of cell voltage with cathode stoichiometry and stack inlet pressure

Figure 3 Optimization study to determine operating conditions for minimum system cost subject to $Q/\Delta T$ constraint, exit RH restricted to 100%

Figure 4 Optimized system cost and performance subject to $Q/\Delta T$ constraint, with and without 100% exit RH restriction

(a) Pt content

(a) System cost

Figure 5 Study to determine the optimum Pt loading in the cathode ternary catalyst

(a) System cost

(b) Pt content

Figure 6 Impact of stack heat load estimate on system cost and performance

(a) Variability in polarization curves

(b) Variability in cell voltage from the mean

(c) System cost

(d) Pt content

Figure 7 Comparison of system cost and performance for representative (REP) and best-of-class (BOC) data

Figure 8 Waterfall charts showing changes in power density and FCS cost due to cell-to-stack voltage degradation, rise in coolant temperature, and relaxation of 100% exit RH constraint