Journal of Power Sources 309 (2016) 178–191

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Performance of advanced automotive fuel cell systems with heat rejection constraint

R.K. Ahluwalia [a, *], X. Wang [a], A.J. Steinbach [b]

[a] Argonne National Laboratory, Argonne, IL, USA
[b] 3M Fuel Cell Components Program, St. Paul, MN, USA

HIGHLIGHTS

- Analyzed the impact of heat rejection requirement on system cost and performance.
- Determined desirable conditions for $Q/\Delta T = 1.45$ kW/°C in an 80-kWe fuel cell system.
- Showed that stack must operate at pressure above 2 atm and temperature above 90 °C.
- Best system performance obtained under drier conditions and cathode stoichiometry <2.

ARTICLE INFO

Article history:
Received 23 July 2015
Received in revised form
7 January 2016
Accepted 14 January 2016
Available online 5 February 2016

Keywords:
Polymer electrolyte fuel cells
Automotive application
Heat rejection
Kinetic and mass transfer losses
Mass transfer in cathode catalysts
Oxygen reduction reaction kinetics
Stability and durability

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 $Q/\Delta T < 1.45$ kW/°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/cm^2 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.

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 5000 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 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

* Corresponding author. Argonne National Laboratory, Argonne, IL 60439, USA.
E-mail address: walia@anl.gov (R.K. Ahluwalia).

http://dx.doi.org/10.1016/j.jpowsour.2016.01.060
0378-7753/

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/ΔT 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/ΔT 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/ΔT 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 5000 h required lifetime [9,10].

The Q/ΔT 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-kWe stack that operates at 0.6 V cell voltage and 80 °C for ~51% stack efficiency and Q/ΔT = 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/ΔT = 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/ΔT 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/ΔT = 1.45 kW/°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/ΔT 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/ΔT 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-cm^2 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/ΔT constraint.

2. Experimental

Multiple 50-cm^2 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–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 (SR_c Series) varied the O_2 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 (SR_a Series) varied the H_2 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