



In-situ diagnostic tools for hydrogen transfer leak characterization in PEM fuel cell stacks part II: Operational applications



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HIGHLIGHTS

- Diagnostic tool for hydrogen leak characterization in operational PEM fuel cells.
- Requires only stack voltage and air flow measurements.
- Provides rate and distribution of hydrogen transfer leak in fuel cell stacks.

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ABSTRACT

This paper describes a diagnostic tool for in-situ characterization of the rate and distribution of hydrogen transfer leaks in Polymer Electrolyte Membrane (PEM) fuel cell stacks. The method is based on reducing the air flow rate from a high to low value at a fixed current, while maintaining an anode overpressure. At high air flow rates, the reduction in air flow results in lower oxygen concentration in the cathode and therefore reduction in cell voltages. Once the air flow rate in each cell reaches a low value at which the cell oxygen-starves, the voltage of the corresponding cell drops to zero. However, oxygen starvation results from two processes: 1) the electrochemical oxygen reduction reaction which produces current; and 2) the chemical reaction between oxygen and the crossed over hydrogen. In this work, a diagnostic technique has been developed that accounts for the effect of the electrochemical reaction on cell voltage to identify the hydrogen leak rate and number of leaky cells in a fuel cell stack. This technique is suitable for leak characterization during fuel cell operation, as it only requires stack air flow and voltage measurements, which are readily available in an operational fuel cell system.

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1. Introduction

Improvements in fuel cell cost, performance, reliability, and lifetime are key to their successful commercialization. One approach for such improvements is using diagnostic tools at various stages of the fuel cell lifecycle, from Research and Development (R&D), to operation, and maintenance, in order to detect and correct failures [1,2]. In the first part of this article series [1], we introduced a diagnostic tool for characterizing hydrogen transfer leaks in PEM fuel cells, which is suitable for R&D applications. In the present work, we introduce a diagnostic tool that is suitable for characterizing hydrogen transfer leaks in an operational fuel cell

stack. This tool uses available sensory information in an operational fuel cell system, therefore, it is inexpensive and compatible with mass manufacturing practices. In addition, it has the capability for early detection of membrane pinholes, which enables the control system and/or the operator to take compensatory actions.

PEM fuel cell membranes develop pinholes as they age, due to a multitude of chemical, mechanical, and thermal processes [3–7]. These pinholes result in reactant gasses to leak across the membrane and react with the species of the opposite electrode, which causes several drawbacks. First, the crossover of hydrogen from anode to cathode, results in reduced fuel utilization, which increases the operational cost of the fuel cell system. In addition, the reaction of crossover hydrogen with oxygen reduces the cathode oxygen concentration, which negatively affects cell voltage and system performance. Furthermore, as the size and number of the

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membrane pinholes grow, so does the flow of the crossover gasses, which can eventually cause anode or cathode starvation, depending on the direction of the overpressure. This affects the reliability of the fuel cell system. Therefore, it is essential to incorporate diagnostic tools that can quantify the rate and distribution of transfer leaks in operational PEM fuel cell systems, in order to improve their cost, performance, reliability, and lifetime.

Authors in Refs. [8–10] interrupted anode and cathode reactants flows and used the rate of cell voltage drop at Open Circuit Voltage (OCV) to identify leaky cells in a fuel cell stack. The basis of the flow interruption technique is that cells with membrane pinholes have higher transfer leak rates, which reduces reactant concentrations at the respective electrodes more rapidly. This can be observed as faster drop in cell voltage, which is used to identify the leak rate. However, one shortcoming of this technique is that other factors such as distribution of pinholes in the cell, flow sharing between cells through inlet and outlet manifolds, as well as pressure gradients in each cell can also affect the rate of voltage drop; therefore, it does not accurately represent the leak rate [8]. In addition, this technique requires individual electrical connection to each electrode, also known as Cell Voltage Monitoring (CVM), which is preferably avoided in commercial fuel cell systems due to cost and reliability considerations [11]. Therefore, this technique is not suitable for characterizing the rate and distribution of transfer leak in operational fuel cell systems.

In Refs. [5,12], authors employed a gas tracer concept in order to measure the amount of transfer leak. They supplied a mixture of hydrogen and helium to the anode and measured helium crossover using a mass spectrometer at the cathode. In Refs. [13,14] authors supplied hydrogen and nitrogen/argon to the anode and cathode of a PEM fuel cell when the cell voltage was fixed voltage of ~350 mV, and used Faraday's law to quantify the hydrogen transfer leak from the cell current. In Refs. [1], we also supplied anode and cathode with hydrogen and nitrogen, and quantified the hydrogen transfer leak from OCV using Nernst equation. However, none of these techniques are suitable for quantifying transfer leaks in an operational fuel cell, as they require supplying gasses that are not readily available in these systems. Furthermore, assuming that such gasses became available in specific intervals, e.g., during preventive maintenance practices, these methods can still not be used to quantify the rate and distribution of transfer leaks in a PEM fuel cell stack. This is because the gas tracer technique of [5,12] requires sampling the cathode of each individual cell, which is not feasible in an operational fuel cell. The Faradaic current [13,14] and Nernst potential [1] based techniques also require CVM to electrically connect with each individual cell in the stack, which is not the preferred hardware in fuel cell systems as explained above. Therefore, these diagnostic techniques are also not suitable for characterizing the rate and distribution of transfer leak in operational applications.

Authors in Refs. [15–17] used a segmented cell to quantify the rate and location of transfer leaks by measuring local voltage and current densities. While this technique can accurately identify transfer leaks, it is also not suitable for operational applications, as it requires segmented cells as well as specialized hardware to measure local voltage and currents.

In this work, we introduce a novel diagnostic tool that enables characterizing the rate and distribution of transfer leaks in an operational fuel cell stack. We first characterize the effects of oxygen concentration and hydrogen transfer leak on a single cell voltage. We show that once the air flow reaches a value close to oxygen starvation, reduction in air flow drops the cell voltage sharply to zero volts. Furthermore, we show that at a fixed current, hydrogen transfer leak increases the air flow rate at which cathode starves, as the crossover hydrogen consumes the oxygen in the

cathode. Therefore, we propose using the air flow rate at which the cell voltage drops to zero to estimate the rate of hydrogen transfer leak in each cell. Next, we expand on this methodology to characterize the rate and distribution of leaky cells, given the stack voltage. The formulation is derived by calculating the residual between the measured stack voltage and the cell voltage drop due to reduction in oxygen concentration as the air flow rate is reduced from a high to low value. We show that this residual represents the number of leaky cells that have oxygen starved at the lower air flow rate, and use that to estimate the rate and distribution of hydrogen transfer leak in the fuel cell stack. Since the technique requires only the measurement of air flow and stack voltage, it is suitable for integration in an operational fuel cell system, as these measurements are readily available in these systems.

2. Experimental

A commercial stack architecture and Membrane Electrode Assemblies (MEA) were used for the single-cell and stack experiments. Single-cell test hardware was used to develop the leak characterization methodology, and a full-size stack with significant transfer leaks was used to verify the developed methodology.

2.1. Single-cell setup

The test station used to carry out the single-cell experiment is similar to that explained in Ref. [1]. The modification made to the station for the purpose of this experiment was the addition of an oxygen Mass Flow Controller (MFC) in parallel to the nitrogen MFC on the cathode side, so that different oxygen/nitrogen mixture concentrations could be supplied to the fuel cell, as shown in Fig. 1. The operating conditions used for the fuel cell in this experiment are shown in Table 1, unless mentioned otherwise.

2.2. Stack setup

For the stack-level experiments, a 408 cell stack was used. The module had been used at Ballard Power Systems for many years and had developed pinholes. The anode inlet pressure was controlled using a pressure regulator, and a Hydrogen Recirculation Blower (HRB) was used to circulate hydrogen and avoid anode flooding and starvation. A solenoid valve was used at the outlet of the anode for purging purposes. The cathode outlet pressure was controlled using a backpressure controller, the cathode air flow rate was controlled using an MFC and humidified with a water-spray humidifier. The streams were heated using electric heating elements and the stack temperature was controlled using deionized water as coolant. The stack load was controlled using an electronic switching load and individual cell voltages were measured using CVM hardware. The temperature and humidity set points used for the experiment are also as indicated in Table 1, unless mentioned otherwise.

3. Results and discussions

When pinholes are formed in the membrane, they promote crossover of gasses across the membrane through diffusion and convection [12,15,18]. As the pinhole size grows, so does the gas crossover flow rate, to the extent that it could cause anode or cathode starvation. In the case of anode starvation, the starved cell could experience reverse potential, which results in significant damage to the MEA [19–22]. Oxygen starvation can also result in loss of performance [20–23]; however, its effects are less damaging and critical than hydrogen starvation. Therefore, in order to mitigate from anode starvation in the case of formation of large

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