Journal of Power Sources 278 (2015) 652–659

Contents lists available at ScienceDirect

Journal of Power Sources

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

In-situ diagnostic tools for hydrogen transfer leak characterization in PEM fuel cell stacks part I: R&D applications

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HIGHLIGHTS

- In-situ diagnostic tool to quantify hydrogen transfer leaks in PEM fuel cell stacks.
- Closed form relationship for the rate of hydrogen transfer leak.
- Requires hydrogen/nitrogen supply to anode/cathode with anode overpressure.
- Requires pressure, flow, temperature, humidity, and OCV measurements.
- Accurately estimates leak in each cell of a stack, suitable for R&D applications.

ARTICLE INFO

Article history:
Received 15 October 2013
Received in revised form
31 October 2014
Accepted 20 December 2014
Available online 22 December 2014

Keywords:
Fuel cell
Diagnostic
Transfer leak
Pinhole

ABSTRACT

This paper describes a diagnostic tool for in-situ characterization of hydrogen transfer leak in individual cells of a Polymer Electrolyte Membrane (PEM) fuel cell stack, suitable for Research and Development (R&D) applications. The technique is based on supplying hydrogen and nitrogen to the anode and cathode of a PEM fuel cell stack while maintaining a prescribed anode overpressure. Under these conditions, hydrogen crosses over from the anode to the cathode, and the Open Circuit Voltage (OCV) represents the ratio of hydrogen partial pressure in the two electrodes. It is shown that by measuring temperature, pressure, flow, humidity, and individual OCVs, the proposed technique can accurately estimate the rate of hydrogen transfer leak in individual cells of a PEM fuel cell stack. This diagnostic tool is suitable for characterizing hydrogen transfer leaks during fuel cell R&D, as it only requires gasses and measurements that are readily available on fuel cell test stations, and does not need disassembling or modifying the fuel cell stack.

1. Introduction

PEM fuel cells are electrochemical cells that combine hydrogen and oxygen to produce electricity, water, and heat. The fuel cell output power is proportional to its electrochemical surface area and its output energy is proportional to the size of the hydrogen tank. This allows fuel cell to scale power and energy separately, making them suitable for a wide range of stationary and mobile applications. However, their commercialization involves satisfying stringent cost, performance, reliability, and lifetime requirements; in order to become competitive with mature rival technologies, i.e. batteries and Internal Combustion Engines (ICE).

One approach for satisfying the above constraints is by employing suitable diagnostic tools that can detect failures and characterize dynamics at various stages of the fuel cell lifecycle [1]. For R&D application, the diagnostic tools need to be repeatable, reproducible, and able to accurately detect and characterize the failures, in order to facilitate design improvements. For operational applications, diagnostic tools are required to detect and isolate failures before they fully develop, so that the control system can take compensatory actions. These tools also need to be inexpensive and compatible with product manufacturing practices. The diagnostic tool used for maintenance applications are required to identify the type of failures, so that appropriate repair decisions can be made. In this paper, we introduce a diagnostic tool that is

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http://dx.doi.org/10.1016/j.jpowsour.2014.12.093
0378-7753/

suitable for characterizing hydrogen transfer leak during fuel cell R&D. In the following papers of this series, we will introduce diagnostic tools that can be used to characterize hydrogen transfer leak during operation and maintenance.

The fuel cell membrane is responsible for conducting protons from anode to cathode while keeping the reactants separate. As the membrane ages, it develops pinholes, which results in transfer leak of reactants across the membrane [2]. Various chemical, mechanical, and thermal processes such as contamination, humidity cycles, and hydrogen crossover, respectively, contribute to the formation and growth of pinholes in PEM fuel cells [3,4]. In order to design membranes that are more durable and resilient to these processes, there is a need for in-situ diagnostic tools that can accurately identify the location and rate of transfer leaks. Such diagnostic tools have been proposed in a number of publications [4–13].

Authors in Refs. [4,5] used a gas tracer concept to measure the amount of transfer leak. They supplied hydrogen with 10% helium to the anode and measured helium cross over using a mass spectrometer at the cathode. This technique allowed them to monitor the development of transfer leaks under operating conditions. Furthermore, by adding tubes along the cell channel, they were able to measure the spatial distribution of the leaks, and by systematic change of anode pressure, they were able to separate cross over caused by convection vs. permeation [5]. While this technique is can accurately identify the rate and location of transfer leaks, it is not suitable for detecting leak rates in a typical fuel cell stack, as each individual cell requires a separate outlet for sampling the tracer gas.

In Refs. [6–8], authors use the rate of drop at OCV caused by interruption in hydrogen/air flow to identify the leaky cells in a fuel cell stack. Once anode and cathode compartments are sealed, reactants will crossover the membrane and recombine with the reactants of the opposite electrode. This recombination results in a drop in hydrogen and oxygen concentrations in the anode and cathode, which is observed as a drop in OCV. As the rate of reactant crossover is higher for membranes with pinholes compare to those without, leaky membranes result in a faster OCV drop. While this technique is a simple method that only requires individual cell voltages to detect the leaky cells in a stack, it has limitation in terms of reliable estimation of the rate and location of the leak. The main shortcoming is due to the fact that other factors such as the location of the pinholes in the membrane, as well as pressure gradients across the flow fields and between individual cells, can also affect the rate of voltage drop, resulting in inaccurate detection [6].

In Refs. [9,10], authors applied a small anode over pressure and measured the change in OCV in order to detect the formation and growth of a pinhole in the membrane. Unser these conditions, hydrogen would leak from anode to cathode, where it combines with oxygen and results in an overpotential, as explained by the Tafel equation. This technique is very simple and can detect pinhole formation and growth in individual cells of a PEM fuel cell stack. However, the shortcoming of this method is that it can only provide qualitative information regarding the membrane pinholes [10]. This can be explained by the localized overpotential around the pinhole, which makes the measured OCV sensitive to parameters other than the leak rate, such as the location of the pinhole in each cell.

Authors in Refs. [11–13] used local voltage and current density measurements to identify the location and rate of hydrogen transfer leak in individual cells. When hydrogen leaks from anode to cathode, it results in a mixed potential due to the superposition of hydrogen oxidation and oxygen reduction, lowering the local cathode potential [14]. In addition, the remaining hydrogen that has not reacted at the cathode diffuses back to the anode in the form of hydrogen ions, which can be observed as local negative currents around the pinhole. Authors show that by measuring spatial voltage and current profiles, it is possible to accurately identify the location of the leak in the MEA. Yet, the leak size estimation of this technique has less accuracy due to the many competing parameters that contribute to local current and voltage profiles, such as variations in local humidity and temperature profiles caused by the leak. Furthermore, this technique requires a specific fuel cell hardware design, which limits its usage for leak detection in typical PEM fuel cell stacks. Also note that while authors in Ref. [11] observe a negative current during OCV around the leak, authors in Ref. [12] report a positive current around the leak at OCV, and negative current around the leak at higher loads.

In order to estimate hydrogen crossover, authors in Refs. [15,16] supplied hydrogen and nitrogen/argon to the anode and cathode of a PEM fuel cell, and measured the cell current at a cell potential of ~400 mV. Under these conditions, the current is consumed to oxidize the hydrogen that crosses over to the cathode. Therefore, the current can be used to accurately quantify the amount of leak using Faraday's law. Furthermore, when combined with a segmented cell, the authors where able to identify the location of the pinholes in the cell [15]. This technique can accurately characterize the rate and location of transfer leaks in a single cell, however, it has limitations for stack applications, as it requires modifying the stack design to allow drawing separate currents from each individual cell.

This paper introduces a novel diagnostic tool that can accurately quantify the size of hydrogen transfer leak in individual cells of a fuel cell stack. The technique is based on supplying hydrogen and nitrogen to the stack anode and cathode, respectively, while maintaining an over pressure between the two electrodes. Under these conditions, hydrogen crosses over from anode to cathode through the membrane pinholes, affecting hydrogen concentration in the cathode. It is shown that by measuring individual cell OCVs, along with the anode and cathode pressure, flow, humidity, as well as the stack temperature, the rate of hydrogen transfer leak in each cell can be accurately estimated. Since this technique only requires measurements that are readily available on standard fuel cell test stations, without the need of any modifications to the stack architecture, it is suitable for characterizing hydrogen transfer leaks during the R&D phase.

2. Experimental

2.1. Single-cell setup

Ballard Power Systems heavy duty bus stack architecture and Membrane Electrode Assemblies (MEA) were used for single cell experiments. The test station used to carry out the experiments is shown schematically in Fig. 1. Anode and cathode input stream pressures were measured using PT-Ain and PT-Cin with a max pressure of 100 psi and accuracy of 1% Full Scale (FS), and controlled using manual backpressure control valves to 13 and 10 psig, respectively. Anode and cathode input Mass Flow Controllers (MFC), with a max flow rate of 3 and 5 slpm and accuracy of 1% FS were used for the supply gases, respectively. A nitrogen MFC with a maximum flow rate of 30 slpm was used to supply the cathode stream with pure nitrogen through the three-way valve, V1. In this case, the nitrogen flow rate was set above the requirement for the cathode stream, and the excess nitrogen flow rate was vented to the outlet manifold, in order to ensure that sufficient flow was available at the cathode. The vent backpressure controller maintained a high nitrogen pressure to ensure nitrogen flow through the stack. Three mass flow controllers, MFC1, MFC2, and MFC3, with a respective maximum flow rate of 20, 200, and 2000 sccm, were used to inject hydrogen to the cathode stream in order to simulate transfer leak at a known rate. Ball valves V2–V5 where used to isolate the cathode