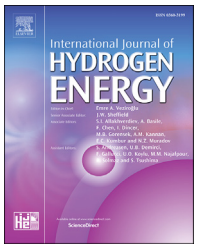
Available online at www.sciencedirect.com

ScienceDirect

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



Impacts of electrode coating irregularities on polymer electrolyte membrane fuel cell lifetime using quasi in-situ infrared thermography and accelerated stress testing

Adam Phillips [a,b], *Michael Ulsh* [a], *K.C. Neyerlin* [a], *Jason Porter* [b], *Guido Bender* [a,*]

[a] *National Renewable Energy Laboratory, 15013 Denver W Pkwy, Golden, CO, 80401, USA*
[b] *Colorado School of Mines, 1500 Illinois St, Golden, CO, 80401, USA*

ARTICLE INFO

Article history:
Received 6 December 2017
Received in revised form
8 February 2018
Accepted 9 February 2018
Available online 2 March 2018

Keywords:
Coating irregularities
PEMFC
Manufacturing
Infrared thermography
Accelerated stress test
Defects

ABSTRACT

In-line quality control diagnostics for roll-to-roll (R2R) manufacturing techniques will play a key role in the future commercialization of the polymer electrolyte membrane fuel cell (PEMFC) used in automotive applications. These diagnostics monitor the fabrication of the membrane electrode assembly (MEA), which detect and flag any non-uniformity that may potentially harm PEMFC performance and/or lifetime. This will require quantitative thresholds and a clear distinction between harmful defects and harmless coating irregularities. Thus, novel fuel cell hardware with quasi in-situ infrared (IR) thermography capabilities is utilized to understand how bare spots in the cathode electrode impact MEA lifetime. An accelerated stress test (AST) simulates chemical and mechanical degradation modes seen in vehicular operation. The actual open circuit voltage and rate of change of this voltage are used as in-situ indicators for MEA failure, enabling capture of the progression of failure point development. Bare spot coating irregularities located at the center of the electrode were found to have no impact on MEA lifetime when compared to a pristine MEA. However, MEA lifetime was found to be considerably shortened when these same irregularities are located at the cathode inlet and, especially, the anode inlet regions of the fuel cell.

Introduction

The polymer electrolyte membrane fuel cell (PEMFC), a zero-emission energy conversion device with hydrogen as its fuel, is a clean alternative to petroleum-based technologies such as the internal combustion engine (ICE). The PEMFC is capable of powering tomorrow's automobiles and portable electronics, as well as integrating into the electrical grid [23]. Durability improvement and cost reductions must be achieved for fuel cell technologies to reach full commercialization, and to meet transportation applications targets of less than 10% performance loss over 5000 h of operation and $14/kW [1,2]. Roll-to-roll (R2R) manufacturing techniques and in-line quality control diagnostics are critical enablers to alleviate cost and durability concerns, respectively. In-line quality control

* *Corresponding author.*
E-mail address: guido.bender@nrel.gov (G. Bender).
https://doi.org/10.1016/j.ijhydene.2018.02.050
0360-3199/

diagnostics are employed on R2R systems to monitor the manufacturing of fuel cell components. Their application ensures a desired quality standard by detecting and flagging non-uniformities that may harm fuel cell durability when present in a membrane electrode assembly (MEA). Unfortunately, in practice, non-uniformities do appear during the fabrication of PEMFC MEAs [3]. The MEA, a proton conducting polymer electrolyte membrane (PEM) with attached platinum-containing catalyst layers (CL) and gas diffusion layers (GDL), may possess non-uniformities in any of its constituent components. These non-uniformities, including pinholes and bubbles within the PEM, electrode coating irregularities within the CL, and cracks within the microporous layer of the GDL, may be created during the MEA fabrication process or subsequent handling [3,4]. Such irregularities may lead to a significant reduction in (i) initial cell performance, (ii) lifetime, and/or (iii) performance over time. All three effects need to be investigated to understand if such non-uniformities should be classified as defective material and flagged in the production process. Our previous work reported on the effects that 100% CL reduction cathode coating irregularities, herein referred to as bare spots, had on initial PEMFC performance using a segmented fuel cell [5]. Most of the electrode irregularities studied in that work did not impact initial total cell performance; however, they did produce local performance variations that may ultimately impact MEA lifetime by acting as seed locations for failures. The present work demonstrates the development of methods to understand if irregularities impact MEA lifetime, and, as an example, investigates the effect of bare spots in the cathode electrode on the development of failure points.

Durability and degradation studies for PEMFCs have gathered interest within the last decade as experimental efforts have shown that current fuel cells fall short of their lifetime targets [6,7]. The effects of operating conditions on degradation, degradation mechanisms of fuel cell components, mitigation strategies for degradation, and other experimental efforts with respect to PEMFC durability have been reported in the literature [8–14]. One primary reason much is still unknown about PEMFC durability is that experiments are both difficult to conduct and time intensive [15,16]. In order to decrease experiment time while still simulating degradation modes present in automotive operation, researchers have developed accelerated stress tests (ASTs) for fuel cells [17,18]. These testing protocols aim to stress individual fuel cell components or materials such as the ionomer, the catalyst layer, or the membrane, by activating one or multiple degradation mechanisms. Zhang et al. [19] and Petrone et al. [17] provide detailed reviews on the main stressors for degradation of the primary MEA components and discuss several AST protocols employed to study PEMFC lifetime. The U.S. Department of Energy (DOE) suggests employing specific AST protocols for MEA chemical stability and membrane mechanical stability under automotive conditions [20]. Several research groups have utilized such protocols by cycling cell relative humidity and operating at its open circuit voltage (OCV) [21–23]. In this work a combination of these AST protocols was employed to age MEAs with electrode coating irregularities under similar conditions as they would experience in an automotive setting at only a fraction of the total testing time [24,25]. Note that our use of ASTs is not for the purpose of a classical study of degradation mechanisms, but rather for the exploration of the impact that coating irregularities in catalyst layers may have on the lifetime of the cell.

The primary goals of this work are to (i) develop methods to capture and localize the onset of failure, (ii) determine if electrode coating irregularities impact the failure location, and (iii) investigate if electrode coating irregularities impact MEA lifetime. If the MEA experiences a significantly shorter lifetime as a result of a coating irregularity, then this irregularity should be classified as a defect. Classification of irregularities as defects will ultimately help develop threshold values for in-line quality control diagnostics, leading to improved MEA production yield.

Experimental

Accelerated stress tests

Chemical degradation occurs most severely at OCV conditions as OH radical attacks are maximized [26]. As a result, the membrane is expected to thin and eventually to fail, causing increased hydrogen crossover. Mechanical degradation can result from changing humidification conditions. As the membrane material swells and shrinks due to repeated humidity cycling, the probability for the development of stress fractures and hot spots which can shorten lifetime is increased [27–30]. In an operating fuel cell both degradation mechanisms occur simultaneously to an extent that depends on the operating conditions. Therefore, DOE AST protocols for chemical and mechanical degradation were combined (and slightly altered) and utilized to assess the lifetime of MEAs with and without coating irregularities. The combined AST protocol entailed operating at OCV while cycling the incoming gas humidification between dry and humidified levels. This protocol is expected to result in impacts to the cell's performance and degradation that are dependent on flow-field design and gas flow configuration. Two flow configurations, i.e. co-flow and counter flow, were investigated to understand their potential impact on the effect of the coating irregularity on failure development. During the AST the cell temperature was 80 °C. Anode/cathode operating conditions were ambient pressures, i.e. 90/90 kPa in Golden, Colorado, 0.5/0.5 standard L/min H_2/air gas flow rates, and 30/30 s duty cycle for dry and wet humidification, respectively. Tests were conducted in both co-flow and counter-flow configuration. For counter-flow operation, the hydrogen flow through the cell was reversed from that of co-flow operation, i.e. the hydrogen inlet became the hydrogen outlet and vice versa.

Fig. 1 displays the relative dew point temperatures measured with a humidity transmitter (Vaisala, HMT330) at the inlet and outlet of the anode electrode for both cases: co-flow is shown in Fig. 1A and counter flow is shown in Fig. 1B. The fluctuation of the dew point at the anode outlet varies over a range of 54.1–68.1 °C for co-flow and 55–66.2 °C for counter-flow, while the frequency and the amplitude of the fluctuation remained virtually identical. The humidity cycling resulted in virtually constant amplitude of the H_2/N_2