ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

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

# Model-based estimation of liquid saturation in cathode gas diffusion layer and current density difference under proton exchange membrane fuel cell flooding

CrossMark

*Junming Hu* [a], *Liangfei Xu* [a,b,c], *Jianqiu Li* [a,b], *Chuan Fang* [a], *Siliang Cheng* [a], *Minggao Ouyang* [a,*], *Po Hong* [a]

[a] *State Key Laboratory of Automotive Safety and Energy, Tsinghua University, Beijing 100084, PR China*
[b] *Collaborative Innovation Center of Electric Vehicles, Beijing 100081, PR China*
[c] *Institute of Energy and Climate Research, IEK-3: Electrochemical Process Engineering, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany*

ARTICLE INFO

*Article history:*
Received 25 May 2015
Received in revised form
20 August 2015
Accepted 1 September 2015
Available online 26 September 2015

*Keywords:*
PEM fuel cell
Cathode GDL flooding
Oxygen starvation
Current density difference
Simultaneous estimation
Online algorithm

ABSTRACT

Poor water management usually leads to various degrees of flooding and exacerbates oxygen starvation, both of which affect the performance and durability of a proton exchange membrane fuel cell (PEMFC). This paper proposes a model-based approach to estimating the liquid saturation and current density difference simultaneously. The current density difference is a parameter that indicates the oxygen starvation level. For estimation purposes, a fuel cell cathode model with separate inlet and outlet subsystems is developed to incorporate the effect of harmful phenomena such as flooding and oxygen starvation on the system dynamics and cell voltage. The cathode gas diffusion layer (GDL) flooding and oxygen starvation diagnoses are formulated as state estimation problems. The proposed approach is validated through an offline simulation using experimental data. The offline prediction suggests the effect of the anode purge and air steam conditions on the internal states of the PEMFC, which is helpful for system optimization and control design. The estimation problems are further decoupled, and a simplified algorithm is designed for onboard applications. Finally, the modified fuel cell model and optimized estimation algorithm are applied to an online test system as a demonstration.


## Introduction

The proton exchange membrane fuel cell (PEMFC) has proven to be a promising candidate for mobile and vehicle applications because of its high power density and low operating temperature. Nevertheless, several technical challenges and performance limitations impede the widespread commercialization of PEMFC technology. Previous publications [1–4] have reported that the degradation of PEMFC, either reversible or irreversible, is a primary issue that negatively affects its performance and durability. In particular, cathode flooding and oxygen starvation are two challenging technical problems for PEMFC operation.

* *Corresponding author*. Tel.: +86 10 62792797; fax: +86 10 62789699.
E-mail addresses: hu-jm12@mails.tsinghua.edu.cn (J. Hu), ouymg@tsinghua.edu.cn (M. Ouyang).
http://dx.doi.org/10.1016/j.ijhydene.2015.09.005

In a Hydrogen/air PEMFC system, external humidification [5] is often used to achieve full hydration of the membrane for high proton conductivity, especially in the cathode inlet region. However, this can easily lead to excessive water generated by the oxygen reduction reaction (ORR) rapidly condensing in the cathode electrode. Liquid water may fill open pores of the gas diffusion layer (GDL) and thus block the transport of oxygen into the catalyst layer (CL). The water may further cover the catalyst sites in the CL, rendering them electrochemically inactive. This process is known as GDL/CL flooding and results in a drop in PEMFC performance. If liquid water accumulation becomes excessive, a water lens or water band may form inside the gas channel, which will increase the parasitic power loss and even clog the oxidizer flow. This is referred to as channel flooding and clogging [6–8]. Oxygen starvation is the other adverse phenomenon on the cathode side and often occurs when insufficient air is distributed into the cathode compartment. In the extreme case, a cell voltage reversal [9] may occur, and hydrogen is produced on the cathode side to provide a compensatory current. As noted in the literature [10], the presence of hydrogen at the cathode can chemically generate heat on the platinum particles and result in local hot spots in the membrane electrode assembly (MEA), which subsequently degrades the PEMFC performance. Because of the delay of the compressor, oxygen starvation may also occur during transient state conditions [11].

To prolong the PEMFC lifetime and assure its performance, water management to prevent the cathode GDL from water flooding and air supply optimal control are critical. To increase the effectiveness of these mitigation strategies, fuel cell diagnostic methods for water flooding and oxygen starvation have been investigated independently in many previous works. The main objective of diagnosing cathode flooding, including GDL/CL flooding and channel flooding, is to predict the mass of liquid water at different locations. Visualization techniques such like neutron imaging [12] and transparent fuel cell [13] have already been applied in the research on fuel cells. Although the visualization-based method makes it easier to study the formation or distribution of liquid water, its implementation on vehicles is still far away. As a non-intrusive method, the pressure drop is widely used to monitor PEMFC flooding because the pressure information can be easily acquired with sensors [14,15]. However, the measured pressure drop is only a good indicator for channel flooding and contains little useful information on the liquid water content in the GDL [16]. In recent years, the electrochemical impedance spectroscopy (EIS) technique has become commonly used in PEMFC diagnosis [17]. In 2010, Toyota Motor Corporation tried to detect the water content of a membrane based on the AC impedance [18]. However, there is still a controversy over the application of EIS to a PEMFC. Yuan et al. [19] pointed out that the AC spectrometer can only give a qualitative prediction and that the correlation between the AC impedance spectra and a specific parameter is still not fully understood. If taking the cost and the integrity into account, model-based approaches are usually regarded as a more viable solution for fault diagnosis, especially in real-time applications [20]. In several numerical studies on water transport in PEMFC, the liquid saturation is often used to precisely describe the flooding level in a GDL, and its spatial and temporal distribution is helpful for understanding the flooding mechanisms [21–25]. Generally, the distribution of liquid saturation will cost a lot of computational effort, which makes it inapplicable for real-time monitoring, but this concept is accepted in some control-oriented models of PEMFC. McKay et al. [26] developed a lumped parameter model and liquid saturation in discretized GDLs was predicted during anode purge event. Hussaini and Wang [27] estimated the liquid saturation based on the Tafel equation to design an intermittent RH control for dynamic water management of a PEMFC. Their results suggest that the mean liquid saturation in GDL is a useful parameter for predicting the degree of water flooding through appropriate modeling and simplification. And recently, Zhang and Pisu [28] proposed a diagnostic approach for fuel cell flooding based on a diagnostic-oriented fuel cell model and unscented Kalman filter to simultaneously predict the channel and GDL liquid saturation. However, they only validated their approach by a simulation, and the online performance has not yet been proven. In terms of oxygen starvation, previous studies mostly focused on air supply control during transient conditions [29–32]; the air stoichiometry $\lambda$ [33,34], which is defined as the ratio between oxygen entering the cathode and oxygen reacting in the fuel cell stack, is commonly used to indicate the degree of air starvation. Dou et al. [9] detected the local interfacial potential, current, and temperature distribution under different air stoichiometric conditions to further reveal the internal connection between the air stoichiometry and oxidant starvation phenomena.

These studies always focused on cathode flooding and oxygen starvation separately, even though these two failures always occur on the cathode simultaneously, especially under high load conditions. Some papers [35–37] have also pointed out that oxygen starvation can be exacerbated by the presence of liquid water in channels or other blockages. This means that, when the PEMFC is under severe flooding conditions, the normal air stoichiometry (usually 2–3) cannot prevent local oxygen starvation owing to the excessive liquid water inside the GDL. In this situation, the air stoichiometry is inadequate as an indicator of oxygen starvation. Previous studies discovered more uneven current distribution characteristics during general air starvation. Gerard et al. [36] investigated the degradation mechanism during oxygen starvation and concluded that the transient current density becomes very high at the air inlet if there is a time delay between the power demand and increase in air flow. Liu et al. [37] analyzed the current distribution during oxidant starvation and observed the decreased local current density in the starvation region. These results imply that the current density distribution in the cathode may provide more direct information for monitoring the state of oxygen starvation than the air stoichiometry. Nowadays the current density real-time measurements have been realized in PEMFC by segmented current collectors [38] or some other advanced measurement techniques [36]. However these measurements may have an impact on fuel cell structures and lead to more cost once implemented. To avoid this consequent effect, the current density distribution along the flow channel is simplified to the current density difference, which is defined as the difference between the mean current densities in inlet and outlet regions, according to experimental results and conclusions shown in previous literatures. And the current density