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Identifying in operando changes in membrane hydration in polymer electrolyte membrane fuel cells using synchrotron X-ray radiography

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ABSTRACT

The cost of the polymer electrolyte membrane (PEM) fuel cell must undergo significant reductions before the widespread adoption of PEM fuel cell powered automotive drivetrains can be achieved. Eliminating the need for active anode humidification is one strategy for reducing the cost and system size of the PEM fuel cell. In this study, we investigated the impact of anode gas inlet relative humidity (RH) on membrane hydration and the associated electrochemical performance of the PEM fuel cell. The anode gas inlet RH was varied to study the impact on fuel cell potential, during which simultaneous in operando visualizations were performed using synchrotron X-ray radiography, and electrochemical impedance spectroscopy was used to gain an understanding of the membrane hydration and water dynamics. The thickness of a Nafion® N115 membrane expanded by up to 26 μm (20% of nominal thickness) compared to the manufacturer specification, as a result of changes in membrane hydration. Through this work, we present the utility of synchrotron X-ray radiography for tracking changes in membrane hydration of an operating PEM fuel cell.

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Introduction

Achieving effective water management has been shown to be a crucial element in obtaining the maximum performance and efficiency from polymer electrolyte membrane (PEM) fuel cells [1–5]. Excess water in the fuel cell accumulates within the gas diffusion layer (GDL), resulting in increased resistance to oxygen transport [6–9]. On the other hand, insufficient water in the fuel cell leads to a loss in membrane hydration and an increase in ionic resistance [3]. Both these scenarios can lead to losses in cell performance [10].

For the incorporation of PEM fuel cells in automotive drivetrains, system durability must be enhanced, and the system cost must be reduced [11]. Eliminating active anode humidification is one strategy to attain this goal [12]. In this introduction, the following four main areas of research that have emerged regarding the effect of humidification on PEM

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fuel cell operation will be discussed: (i) effect of humidification on cell performance, (ii) electrochemical impedance spectroscopy, (iii) membrane swelling, and (iv) membrane water content.

Büchi and Srinivasan [13] demonstrated that PEM fuel cells can be operated in the absence of external humidification if the cell temperature and gas stoichiometry are closely controlled; however, a performance reduction of up to 40% below the conventional mode of operation would be the result. While this strategy shows promise, the impact of reduced anode humidification on the cell performance, membrane hydration, and overall water balance must be thoroughly understood. Although, the impact of inlet gas humidification on fuel cell performance has been studied [10,13–17], there has been limited research on asymmetric humidification [18–20].

It has been well established that the ionic conductivity of the Nafion® membrane is directly proportional to its hydration and proton conduction increases under high hydration conditions [21–24]. Saleh et al. [18] showed that increases in inlet gas relative humidity (RH) lead to rapid decreases in membrane resistance; however, this linear improvement in performance only occurs at low current densities. As they increased current densities (with higher rates of water production), a distinct reduction in performance was observed with fully humidified gases. Saleh et al. [18] proposed that a mid-range RH should be selected for optimal performance, and they concluded that while implementing a dry anode, the cathode inlet RH can be modified to achieve various levels of performance. However, with a dry cathode, the cell was only functional with a fully humidified anode. Amirinejad et al. [19] demonstrated that the removal of anode humidification resulted in a significantly stronger impact on cell performance compared to the removal of cathode humidification. Zhang et al. [15] provided an analysis of operating fuel cell resistances as a function of RH and current densities.

Reducing membrane hydration results in increases in ionic transport resistance, which is a major contributor to the overall ohmic resistance of the PEM fuel cell. High frequency resistance (HFR) measurements are useful for measuring ohmic resistances [25]. Friede et al. [26] reported that a significant increase in cell resistance was observed 50 s after the introduction of dry reactant gases, pointing to the time required to impact membrane hydration. Yu and Ziegler [27] also demonstrated that changes in membrane hydration were predominantly observed when current density was increased, and they attributed this relationship to increasing electro-osmotic drag [28].

Membrane water balance and water transport through the membrane have been studied extensively under steady state operation [3,4,8,29–32]. However, for the dynamic load expected in automotive applications, it is important to investigate the dynamic changes in water content in the individual layers of the PEM fuel cell during transient operation. Banerjee and Kandlikar [33] presented a comprehensive review of transient processes affecting fuel cell performance, and they identified changes in membrane hydration as a process that undergoes transient changes over time periods of up to several minutes.

Niya and Hoorfar [34] provided a thorough review of the use of electrochemical impedance spectroscopic techniques for modeling fuel cell behavior. The Randles circuit is a commonly used equivalent circuit model used to evaluate fuel cell performance (Fig. 1). Antonacci et al. [8,35] correlated the mass transport resistances obtained from the Randles circuit model to the liquid water distributions in a PEM fuel cell, quantified via X-ray radiography. In the Randles equivalent circuit model, R_Ω ($\Omega.cm^2$) represents the ohmic resistance of the membrane, C (F/cm^2) is the double layer capacitance of the catalyst layer, and R ($\Omega.cm^2$) is the ohmic resistance of the catalyst layer. The mass transport resistance in the cathode is represented by the Warburg element.

Niya and Hoorfar [36] identified changes in membrane water transport with electrochemical impedance spectroscopy that occurred at low frequencies (~0.1 Hz). Chevalier et al. [37] also used electrochemical impedance spectroscopy to investigate changes in oxygen mass transport behavior in an operating fuel cell. The authors observed a loss in membrane hydration and implemented a membrane water transport component into their equivalent circuit model to account for changes in membrane hydration. The changes in membrane hydration levels occur at slow rates [33] and therefore are observed at the low frequency region of impedance spectra.

Membrane hydration and membrane water transport have also been studied through the use of high frequency resistance measurements. High frequency resistance is a component of the electrochemical impedance spectra, where a frequency is selected to represent the high frequency intercept of the real component of impedance on a Nyquist plot. Several authors have pointed to the use of 1000 Hz as a typical frequency for measuring high frequency resistance [38,39].

PEM fuel cell flooding [40] and visualization-based studies of liquid water transport through the GDL [41–44] have been the focus of several studies. Researchers have also investigated the impact of membrane hydration loss on cell performance [14,18,29,30,45]. However, the changes in membrane hydration have been investigated mainly through indirect methods such as high frequency resistance (HFR) measurements [25,38] and electrochemical impedance spectroscopy [8,25,38,46,47]. A direct measurement of membrane hydration using high resolution visualization techniques would facilitate a further understanding of membrane hydration and the distribution of water content within the membrane.

Lai et al. [29] developed a novel technique for measuring the hydration of the membrane by measuring the swelling

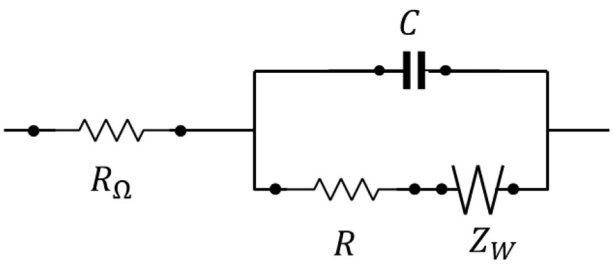

Fig. 1 – Randles equivalent circuit model.

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