

Effective Technique for Improving Electrical Performance and Reliability of Fuel Cells

M. Alrweq, A. Albarbar

Advanced Industrial Diagnostics Research Centre, School of Engineering, Manchester Metropolitan University, Manchester, UK

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ABSTRACT

To optimise the electrical performance of proton exchange membrane (PEM) fuel cells, a number of factors have to be precisely monitored and controlled. Water content is one of those factors that has great impact on reliability, durability and performance of PEM fuel cells. The difficulty in controlling water content lies in the inability to determine correct level of water accumulated inside the fuel cell. In this paper, a model-based technique, implemented in COMSOL, is presented for monitoring water content in PEM fuel cells. The model predicts, in real time, water content taking account of other processes occurring in gas channels, across gas diffusion layers (GDL), electrodes, and catalyst layer (CL) and within the membrane to minimize voltage losses and performance degradation. The level of water generated is calculated as function of cell's voltage and current. Model's performance and accuracy are verified using a transparent 500 mW PEM fuel cell. Results show model predicted current and voltage curves are in good agreement with the experimental measurements. The unique feature of this model is that, no special requirements are needed as only current, and voltage of the PEM fuel cell were measured thus, is expected to pave the path for developing non-intrusive control and monitoring systems for fuel cells.

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Corresponding Author:

A. Albarbar,
School of Engineering,
Manchester Metropolitan University,
Manchester, M1 5GD, UK.
Email: a.albarbar@mmu.ac.uk

1. INTRODUCTION

Proton exchange membrane (PEM) fuel cells have strong potentials as they offer clean, noise free and relatively inexpensive sources of clear electrical energy. Unfortunately, they suffer some drawbacks including low reliability due to a number of common failure modes such as degradation, poisoning, hydration and dehydration. Hence, optimisation of water content is critical for smooth operation of PEM fuel cells and subject of on-going research works. To minimize voltage losses and enhance performance of PEM fuel cells, water inside them has to be properly managed in line with processes occurring in gas channels, across the GDL (gas diffusion layers), electrolyte, catalyst and membrane. Additionally, membrane needs to be hydrated well in order to enhance its conductivity and to ensure good performance while gas channel and GDL need to be protected from flooding.

In addition to water level in membrane and GDL, relative humidity (RH) and the formation of water droplets in the gas channels are identified as key parameters need to be managed [1]. Water is produced through the electrochemical reaction (see Figure 1 and Equations (1)), with the reactant gas needing to be humidified to assist in membrane humidification in order to enhance ionic conductivity. In-membrane water transport leads to back-diffusion and electro-osmotic drag [2]. Accordingly, the ideal water level is not easy

to practically determined, and is problematic to maintain, particularly without in situ water-level measurement.

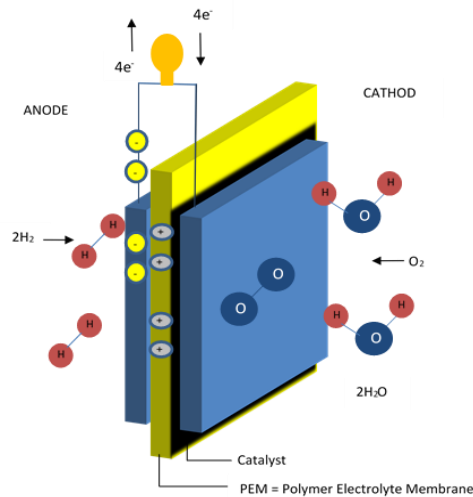
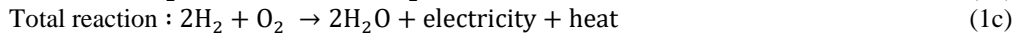
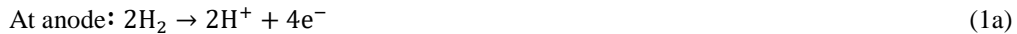


Figure 1. Functionality of the proton exchange membrane fuel cell

Working under flood condition for long time leads to mechanical degradation of the membrane electrode assembly (MEA) and starvation [3]. Alrwashdeh et al [4] used synchrotron X-ray imaging to study the transportation of liquid water inside gas diffusion medium (GDM) in proton exchange membrane fuel cells. He et al [5] proposed a method for monitoring the flow of water in porous electrodes by measuring the pressure drop between the inlet and outlet of electrodes. Legros et al used electrochemical impedance spectroscopy (EIS) and acoustic emission (AE) measurements techniques [6]. An electrical equivalent circuit was designed and flooding was found to considerably affects the 'resistance and the cathodic impedance. AE measurements show the possibility of tracing the water content within a PEM fuel cell. Le Canut et al. [7] used EIS to detect the level of water in the membrane. Li et al. An EIS was used to detect and isolate the cause of the fault, and a simple boost-buck cascaded dc-dc converter was designed to mitigate the fault. Zhang et al. [8] used the water management technique at the cathode of the fuel cell as a key approach for reducing the degradation of reliability and improving performance. Other studies have focused on developing control strategies to optimise the electrical performance of hybrid power systems that include PEM fuel cells [9-13].

This paper presents an accurate model for quantifying liquid and water vapour accumulated in PEM fuel cells. Effects of water flooding on gas channel and how water level develops inside the cell are studied in detail. Section II, discusses water formulation in PEM fuel cells and the derivation of the proposed model is presented in Section III. An experimental set up, to verify model's outcomes, is shown in Section IV, results and comments are in Section V. Finally a brief conclusions are given at the end of the paper.

2. FORMATION OF WATER IN PEM FUEL CELL

During the normal operation of PEM fuel cells, water is produced at cathode side due to electrochemical reactions. That water is used to humidify membrane, to enhance its ionic conductivity, and the rest of water is removed. If water production rates exceeded removal rates, water clog membrane pores and gas diffusion layers (GDLs). This leads to cell degradation due to starvation of gas at both electrodes. Mass transport losses is tightly related to flooding, especially at the cathode side because of the transport rates of the reactants to the electro-catalyst layers is significantly reduced. Output voltage could be recovered, relatively fast, as soon as both sides of cathode and anode are purged. Main function of a membrane is to prevent the electrons passing from side to another while permit only positive negative ions and water molecules to pass through. Therefore, it has to be adequately humidified for good conductivity and low

resistance losses. It is a usual practice to assume that gas diffusion layers (GDL), catalyst layers (CL) and membrane are at same temperatures. Membrane conductivity and water content can be described by Equation (2).

$$\lambda = 0.043 + 17.18a_{water_vap} - 39.85(a_{water_vap})^2 + 36(a_{water_vap})^3 \quad (2)$$

The value of the a_{water_vap} can be computed by Equation (3).

$$a_{water_vap} = \frac{P_w}{P_{sat}} \quad (3)$$

Where the P_w denotes the partial pressure of water vapour and P_{sat} the saturation water vapour pressure of the system. Water accumulation of the cathode with respect to the anode can be approximated by a linear difference of a single step as shown in Equation (4).

$$\alpha = n_{drag} - \frac{F}{I_a} D_w \frac{c_{w,c} - c_{w,a}}{t_m} \quad (4)$$

Where t_m is the thickness of the membrane. The parameter n_{drag} is the electro-osmotic drag coefficient, which is the same as the number of water molecules carried by a proton. This quantity is highly dependent on the content of the water in the given membrane, which is a function of the water activity in gas phase adjacent to that membrane. The partial dehydration which, exists along the anode, as well as the saturation which exists along the cathode, is very likely to occur especially at current densities of higher magnitudes. This is due to the higher transport rate of water because of the electro-osmosis drag from the anode to cathode compared to the back-diffusion rate of water from the cathode to the anode. Physically, this means that water content at the anode side is lower. For the same reason, the water activity on the anode side may be used to calculate the electro-osmotic coefficient in the membrane. The electro-osmotic coefficient is a function of water activity in the low channel of the anode used and can be expressed by Equation 5. The water content can be limited by water activity described in Equation (5).

$$\begin{aligned} \lambda &= A_1 + A_2 a - A_3 a^2 + A_4 a^3 \quad \text{for } 0 < a \leq 1 \\ \lambda &= 14 + 1.4(1 - a) \quad \text{for } 1 < a \leq 3 \end{aligned} \quad (5)$$

Where: $A_1 = 0.043, A_2 = 17.18, A_3 = 39.85, A_4 = 36$
Equation (5) can be rewriting as shown in Equation (6).

$$\begin{aligned} \lambda &= 14 a \quad \text{for } 0 < a \leq 1 \\ \lambda &= 12.6 + 1.4a \quad \text{for } 1 < a \leq 3 \end{aligned} \quad (6)$$

The flooding of the cathode is associated with three mechanisms:

- The influence of applied electric that occurs at the membrane causing electro-osmosis drag, causing the protons move from anode to cathode carrying water molecules with them particularly at high current load. The current density in the cathode increases alongside the relative humidity of membrane increases because of electron conductivity.
- Water forms due to the reaction between oxygen molecules and hydrogen molecules at the cathode. The level water increases when the load and/or current density increases.
- Liquid water injection and saturated reactant gases contribute to causing flooding.

3. MODELLING AND SIMULATION

In order to gain thorough understanding of the flooding phenomenon in PEM fuel cells, a model was implemented in a COMSOL software environment as shown in Figure 2. The model is based on the finite element method (FEM) in which the modelling domain is discretized to finite elements and the equations are solved in each element using piecewise continuous polynomials. COMSOL software is found suitable for this task as it has the ability to create the mesh for complex geometries and offers various solver algorithms for different problems. Oxygen, nitrogen and water are considered to exist at cathode while just hydrogen and water at the anode. To simulate flooding, low temperature and velocity gases are applied. The Maxwell-Stefan equation is utilized to compute the chemical species that diffuse in gas phase (Equation (7)).

$$\frac{\partial}{\partial t} \rho w_j + \nabla \cdot \left[-\rho w_i \sum_{j=0}^N j + (\nabla w_j + w_j) * \frac{\nabla m}{m} + (x_j - w_j) \frac{\nabla \rho}{\rho} \right] + w_j \rho u + D_j^T * \frac{\nabla T}{T} = R_i \quad (7)$$

Brinkman's law, expressed in Equation (8), was used in this model to describes flow velocities in porous media namely GDL and gas channel (GH).

$$\frac{\rho}{\varepsilon_p} \left[(u \cdot \nabla) \frac{u}{\varepsilon_p} \right] = \nabla \cdot \left(-pI + \frac{\tau}{\varepsilon_p} \right) - \left(\frac{u}{k} + \frac{Qbr}{\varepsilon_p^2} \right) u \quad (8)$$

The main negative effect of the flooding phenomena is the water content will hinder the reactions reach to the active areas such as catalyst layer also the porous media may blocking that prevent the protons to emigrating from anode to cathode.

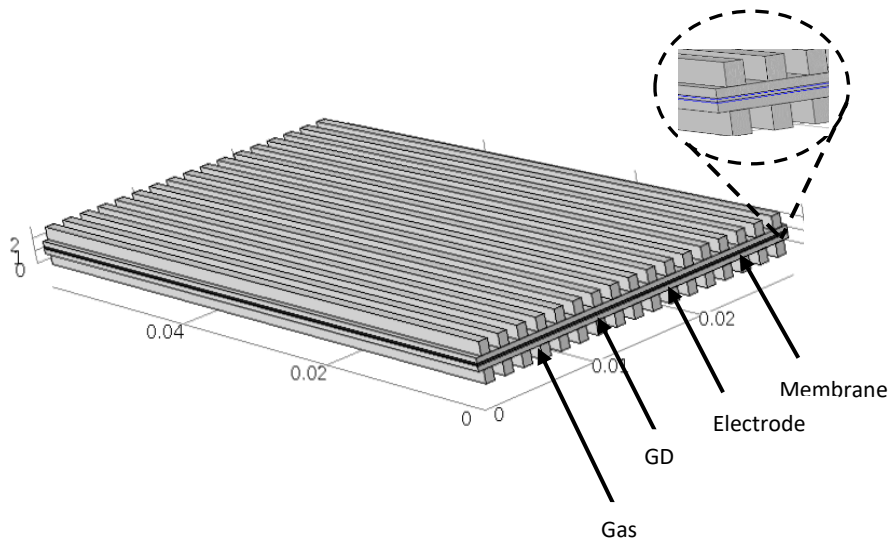


Figure 2. Model of the PEMFC considered in this work

4. EXPERIMENTAL SETUP AND DATA COLLECTION

A 500mW, single PEM fuel cell with the following specifications was tested, Active Area: 16cm², Membrane Electrolyte Assembly: Nafion 115, 1 mg. Pt/cm² (platinum load at both sides) and Toray carbon fibre paper with 0.19mm thickness as GDL.

In this experiment, operating temperature and pressure have been precisely controlled due to their significant impact on water management issue. Electrolysis is used to ensure both hydrogen and oxygen have humidity and purity of 100% and 99.9% respectively. Gas pressures and temperatures are controlled automatically while the rate of water drained is controlled manually. The cell temperature ranged from 20 to 800°C. There are two approaches to simulate flooding conditions. The first method relies on the difference between the temperature of the gases and the cell i.e, if the cell's temperature is higher than the temperature of gasses, the cell will be under a dry condition. In contrast, if the gas's temperatures are higher than the cell's temperature, the cell will be flooded. The second method controls the water drainage. Owing to its simplicity, the second method was chosen to mimic a flooding condition. Water accumulated inside the fuel cell was released using air stream.

This method should be used with caution as it may damage the delicate components such as membrane. To ensure repeatability, independent flooding experiments were carried out seven times. For experimental simplicity, the flow channel of the PEM fuel cell was signal-phase flow, so that the flooding condition can be declared if the measured values of current and voltage drop deviate significantly from its predicted values due to presence of liquid water in the system. External variable DC load was connected to the cell to vary the current drawn from it. The output voltage and current is then acquired and recorded using a data acquisition card interfaced to a PC runs a LabVIEW software. A block diagram representing the experimental setup is shown in Figure 3.

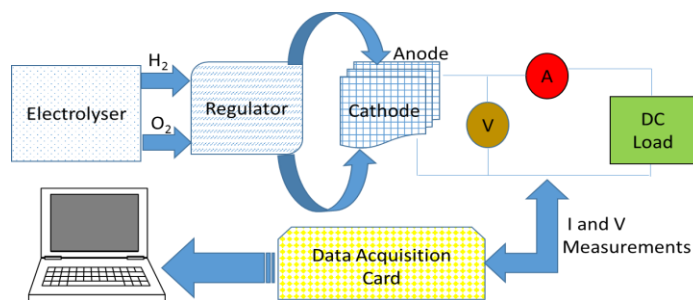


Figure 3. Block diagram of the experimental setup

5. RESULTS AND DISCUSSION

At low current density, no liquid water is accumulated in the channels, just a small amount of water generated at the cathode side due to oxygen reduction reaction (ORR). Figure 4 shows the flooding of PEM fuel cell at constant pressure, fully humidified reactance and a slowly increasing current. The output voltage and current curves were as same as normal operating condition.

First water droplet emerged, is subject to three forces: gravitation (G), viscous force between droplet and solid surface (F_s), and shear drag force induced by the gas flow (FD). The droplet was small at the beginning, $FD + G \leq F_s$, and held on the GDL surface or/and the channel sidewall. With continued operation of the fuel cell, more droplets emerge and grow larger, which started to block the porous membrane and GDL, hindering gas flow. This is in a good agreement with the simulation results for water accumulated in the cathode side shown in Figure 5(a). Clearly, the level of water at the outlet is higher than the inlet, owing to water production because of ORR and electro-osmotic drag all exiting through the outlet point. Formation and accumulation of water in anode side is shown in Figure 5(b).

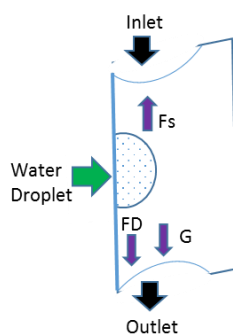
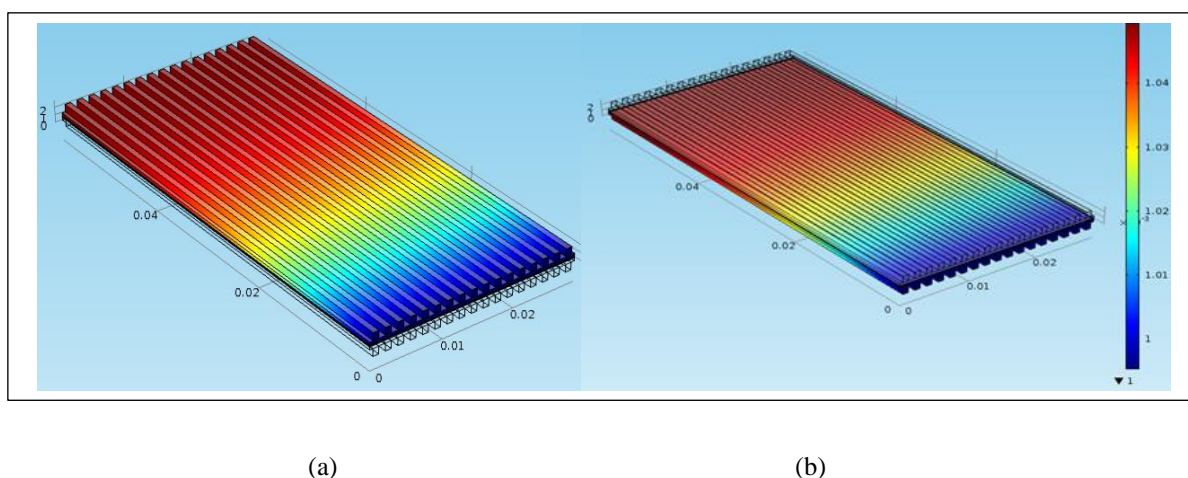


Figure 4. Water droplets induced forces in the channel



(a)

(b)

Figure 5. Water concentration at the (a) cathode side (b) anode side

Due to the lack of water generation at the anode, the water accumulation is as result of the gas humidifying and back diffusion phenomenon, so the amount of water is less than that at the cathode. The membrane is one of many components with the PEM fuel cell affected by water management issues. The membrane requires adequate water to insure high ions conductivity and reducing the Ohmic losses. The water accumulates in the membrane is due to electro osmotic drag and the back diffusion phenomenon. Water concentration, for 10000 seconds of operation, at anode, cathode and membrane are shown in Figure 6. The pressures of hydrogen, oxygen and water during cell in flooding condition are shown in Figure 7. Water pressure increases with flooding while H_2 and O_2 pressures decrease. This is because of water droplets clog the GDL and gas channels. Output voltage from the PEM fuel cell, for the both normal and during flooding cases, are shown in Figure 8. The voltage curve in flooding condition can be classified into three category: the first section describe the water produced and accumulated at cathode side (cathode side is main source of the water because of the electrochemical reaction as describe in Equation 1c). Water takes time to accumulate inside the cathode side depends on a number of factors such as current density, pressure and temperature. When water clogs the GDL, voltage amplitude declines sharply. Last part of curve illustrates the voltage recovery after water purging. Figure 9 shows output voltage curves that obtained from both simulation and experimental results. There is good agreement and reasonable correlation between the experiment and simulation results.

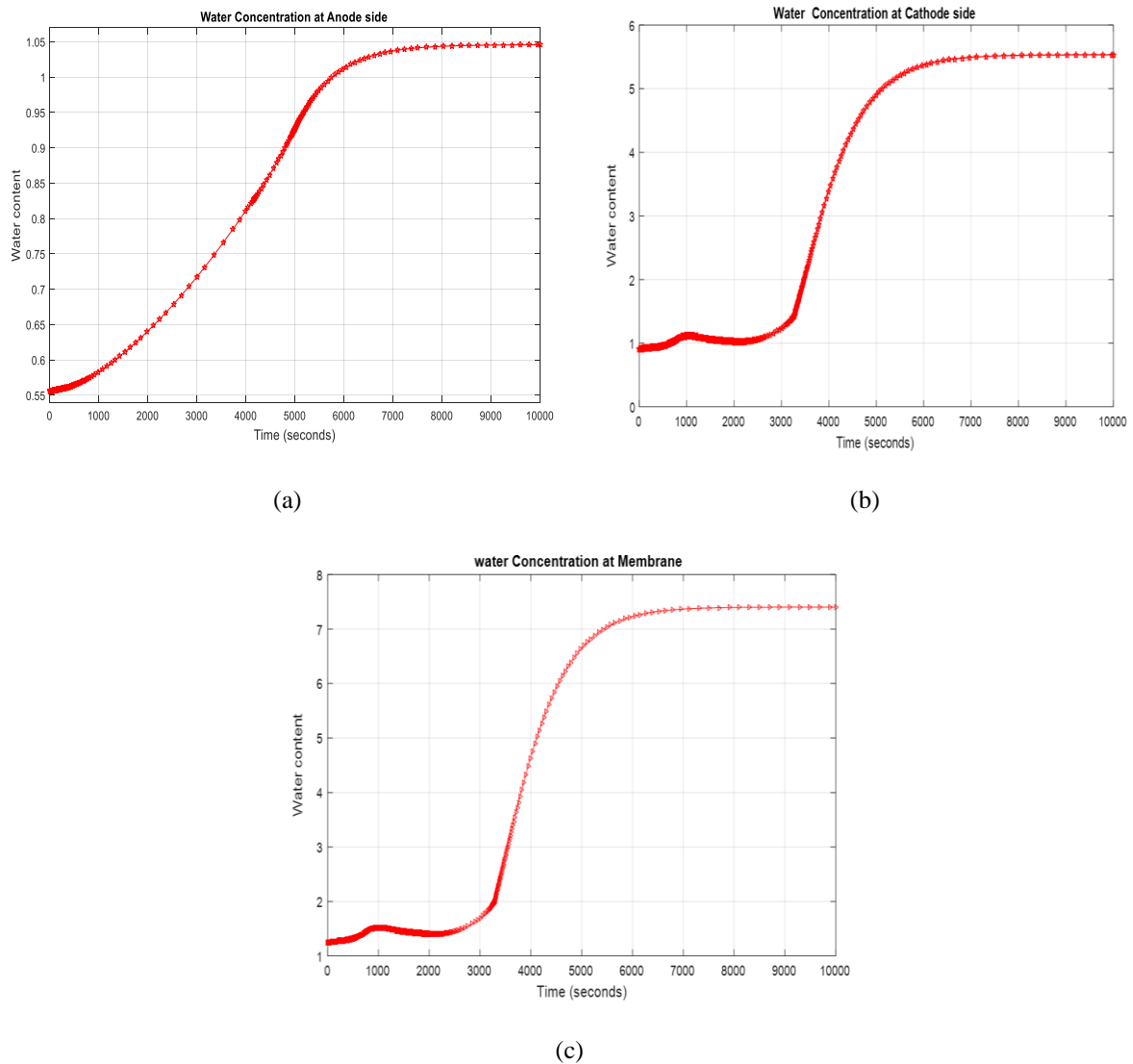


Figure 6. Water concentration at (a) anode, (b) cathode (c) membrane

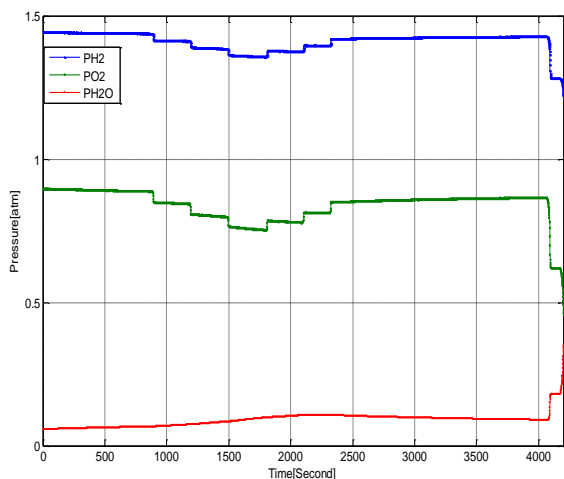


Figure 7. Pressures of the PEM fuel cell during flooding

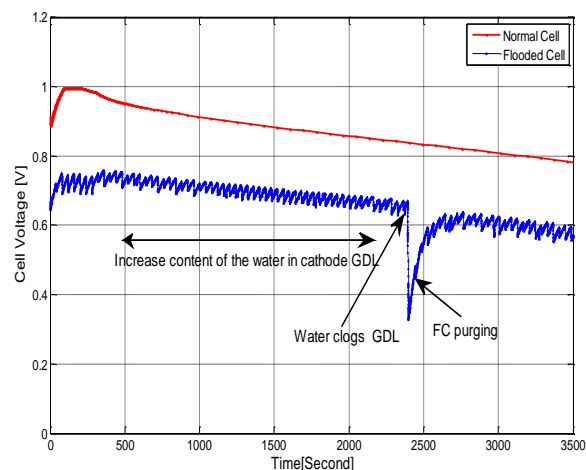


Figure 8. Voltage of the PEM fuel cell during flooding state

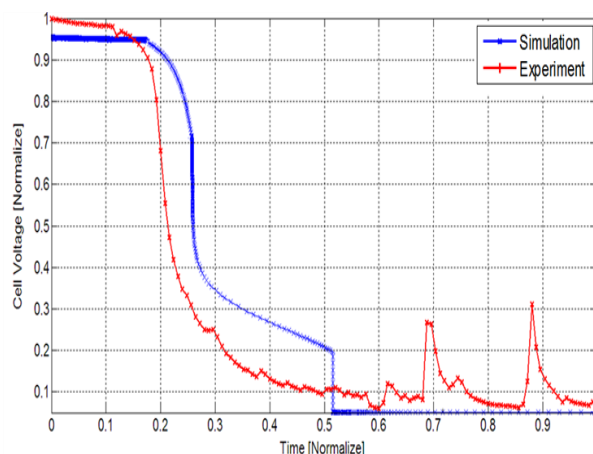


Figure 9. Experiment and simulation results

6. CONCLUSION

In this study, a model-based technique for determining flooding in proton exchange membrane (PEM) fuel cell was presented. The performance and outcomes of the model were verified through experimental setup using 500mW PEM single fuel cell. The proposed technique can offer real-time instant information about the level of accumulated water in the cell while in operation. Location where water is exceeding the normal levels can also be identified. Presented results demonstrate the effectiveness of the derived technique especially for deployment in real-time monitoring system for PEM fuel cell. It is anticipated that the developed technique combined with other work on fuel cell control and monitoring systems, especially, those presented in references 9, 10 and 11, will pave the path for the development of accurate and powerful control and condition monitoring systems for PEM fuel cells.

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