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Investigation of an Indicator for On-line Diagnosis of Polymer Electrolyte Membrane (PEM) Fuel Cell Flooding using Model Based Techniques

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ABSTRACT

The durability and reliability of producing high quality power for long periods of time have the potential to be the leading marketing factors for future hydrogen and fuel cell power sources. In the past few decades, several researchers have been devoted to investigating diagnostic techniques for fuel cell systems. However, in commercial fuel cell applications, on-line diagnosis is urgently required so that fuel cell degradation can be detected at its early stage, and mitigation strategies can be performed to recover fuel cell performance. In this paper, on-line diagnosis of fuel cell flooding is investigated. For this purpose, a generalised fuel cell stack model is developed, and water mass balance equations are used to study water balance inside the fuel cell stack. Moreover, with these equations, the flooding indicator is proposed and its relationship with liquid water inside the stack is evaluated. Results demonstrate that the proposed indicator is sensitive to the liquid water in the stack, thus can be used for flooding diagnosis. Furthermore, the expectation of the proposed indication in the cell flooding case is also presented. The advantage of this method is that parameters in the flooding indicator can be determined with measurements from tests, thus quick diagnosis can be made during the practical fuel cell operation.

1. INTRODUCTION

In the last few decades, hydrogen and fuel cells have emerged as potential initiatives that could serve as alternative energy sources, with characteristics of being zero-emission energy conversion and power generation devices. They are currently being engineered for a range of applications including automotive, stationary power, aerospace and customer electronics.

However, fuel cell reliability and durability is one of the barriers which block the wider application of fuel cell systems. A possible solution for this problem is effective diagnostic techniques. In the past few years, several studies Lei Mao et al. This is an open-access article distributed under the terms of the Creative Commons Attribution 3.0 United States License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited.

have been devoted to the diagnosis of fuel cell systems using both model and non-model based techniques (Fouquet et al. 2006, Giurgea et al. 2013, Hernandez et al. 2010, Onanena et al. 2013, Steiner et al. 2011, Zheng, 2013), and most of them have been verified with numerical or experimental studies. Meanwhile, there is only limited investigation about successful application of on-line diagnostic methods for commercial fuel cell systems (Ingimundarson et al. 2008, Narjiss et al. 2008, Li et al. 2013). Narjiss, et al. presented a method of using an isolated DC/DC power converter and digital signal processor to measure fuel cell harmonic impedance, thus fuel cell degradation related to gas feeding and membrane humidification could be detected by monitoring the measured impedance. Ingimundarson, et al. used hydrogen mass balance equations to detect the leak of hydrogen based on measurements from tests directly. Li, et al. employed a combination of Fisher discrimination analysis and a Gaussian mixture model to distinguish normal and faulty fuel cells using individual cell voltage measurements. With these techniques, fuel cell degradation can be detected and isolated during its operation, thus strategies can be taken to maximise fuel cell lifecycle performance. However, it should be noted that as processing of measurement data is required in these studies, with designed signal processors in the test configuration or signal processing techniques after collecting the measurements, these processings may be time-consuming in the real application, and can not give instant alert for the fuel cell degradation. Therefore, further studies are still required to investigate the on-line diagnostic techniques for fuel cell systems.

In this study, the on-line diagnosis of fuel cell flooding is investigated. The proposed indicator can give instant information about accumulated liquid water during the stack operation, and location of excess water can also be indicated. Therefore, the proposed indicator can not only detect the fuel cell stack flooding, but also show the amount of accumulate water inside the fuel cell stack, which can be used to quantify the level of flooding. As water management is one of the key points for fuel cell performance, enough water should be kept in the membrane for the passage of hydrogen ions, but too much water will

block the reaction sites, thus preventing reactant gases within the fuel cell (Knowles et al. 2010, Schmittinger et al. 2008, Wu et al. 2008). Moreover, flooding is an aging factor for fuel cell, that is, its performance will be reduced gradually. However, if flooding is not detected and mitigated at its early stage, it will lead to irreversible damage to the membrane, causing failure of the fuel cell (Ous and Arcoumanis, 2013, Rama et al. 2008).

In the paper, the water balance equations at the anode and cathode will be presented, these equations are commonly used in fuel cell system models (Khan and Lqbal, 2005, Pukrushpan, 2003, Mann et al. 2000), discussed in section 2. With these equations, water accumulation rates are calculated to study the water balance inside the fuel cell with a numerical study described in Section 3. Furthermore, section 4 proposes a flooding indicator, which can be used in practical fuel cell applications as it can be easily obtained using measurements during fuel cell operation. Numerical results show that the indicator can represent the liquid water condition inside the fuel cell, and sensitivity of the indicator is also studied. Section 5 predicts the performance of the proposed flooding indicator under the fuel cell flooding case, based on its performance under normal fuel cell operation. Finally, conclusions are given and further work is suggested.

2. FUEL CELL STACK MODEL

The Proton Exchange Membrane (PEM) fuel cell typically includes two porous electrodes separated by a proton conducting membrane, which is impermeable to gases but can allow proton to pass through it. Catalyst is commonly used to separate electrodes from the membrane.

During fuel cell operation, hydrogen enters the cell on anode side while air enters on the cathode side. A catalyst on the anode side splits hydrogen atoms into electrons and positive charge hydrogen protons. The protons can pass through the membrane while electrons pass the electrical circuit to reach the other side. Voltage is created across the cell by passed protons. A catalyst on the cathode side will react passed protons, electrons, and oxygen to form water and also product heat.

According to previous studies (Khan and Lqbal, 2005, Pukrushpan, 2003), a fuel cell model can be developed to express the behaviour of the fuel cell. In the fuel cell model, the water condition inside the cell should be considered, and the water balance equations are usually used for this purpose. Based on these investigations, the water balance equations at the cathode and anode sides should be expressed separately (Eqs. 1 and 2 respectively):

$$\frac{dm_{w,ca}}{dt} = W_{v,in,ca} + W_{l,in,ca} + W_{v,gen} + W_{v,membr} - W_{v,out,ca} - W_{l,out,ca} \quad (1)$$

$$\frac{dm_{w,an}}{dt} = W_{v,in,an} + W_{l,in,an} - W_{v,membr} - W_{v,out,an} - W_{l,out,an} \quad (2)$$

Where m is the gas mass (kg), W is the gas mass flow rate (kg/s). Subscripts used in the equations have different meanings, 'ca' and 'an' mean cathode and anode, respectively, 'in' represents inlet flow terms, 'out' represents outlet flows, 'reacted' means reacted gas, 'l' represents liquid water, while 'v' means water vapour, and 'membr' represents water vapour across the membrane.

From Eqs.(1) and (2), the water accumulation rates at the cathode and anode can be calculated. It should be noted that inlet water vapour, inlet liquid water, generated water product can be measured directly, outlet water vapour and liquid water can be obtained using measurements during fuel cell operation, and water across the membrane should be determined with the model.

3. INVESTIGATION OF WATER ACCUMULATION INSIDE THE FUEL CELL USING THE WATER BALANCE EQUATIONS PUBLICATION BY THE ANNUAL SOCIETY OF THE PROGNOSTICS AND HEALTH MANAGEMENT SOCIETY

3.1. Development of the Fuel Cell Model

With the water balance equations in section 2, water balance inside the fuel cell can be investigated. In this study, a generalised fuel cell model is developed, which includes modules determining anode and cathode flows, a module which evaluates membrane condition, and a module calculating fuel cell voltage. Fig. 1 shows the block diagram of the developed fuel cell model, details of the model, such as detailed differential equations, and determination of model parameters, can be found in other studies (Ous and Arcoumanis, 2013, Rama et al. 2008, Khan and Lqbal, 2005).

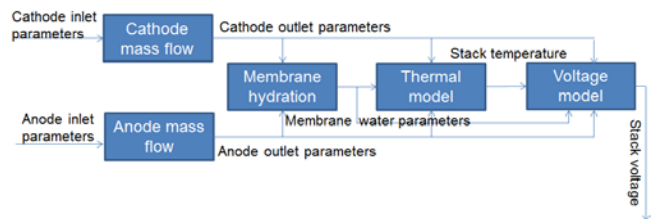


Figure 1 Fuel cell model block diagram

3.2. Verification of Developed Fuel Cell Model

Before using the developed model for analysis, its performance is verified with polarisation curves from previous studies (Khan and Lqbal, 2005). Table 1 lists the input parameters from tests in the reference paper, and Fig. 2 depicts the comparison of the polarisation curves from test data in the reference and the data from the developed model. It can be observed that the polarisation curves match well,

which indicates the developed model can express the fuel cell stack behaviour with good quality.

Table 1 Input parameters for fuel cell model from Khan and Lqbal, (2005)

Parameter	Value
Number of fuel cells	54
Active electrode area of single cell	46.5cm ²
Hydrogen flow rate	1.15 stoich
Air flow rate	2.0 stoich
Hydrogen pressure	3.5 bar
Air pressure	3.5 bar

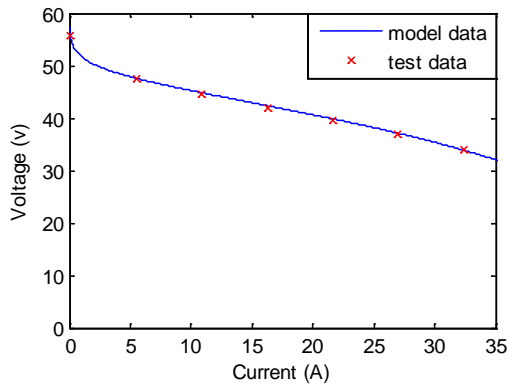


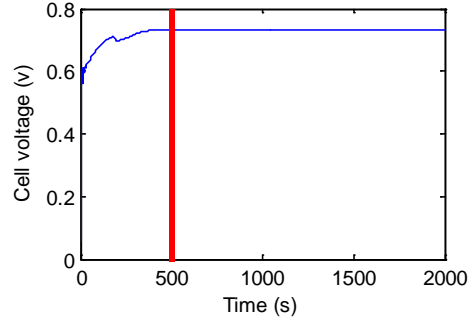
Figure 2 Comparison of polarisation curves from the model and test in Khan and Lqbal, (2005)

3.3. Investigation of Water Accumulation Inside the Fuel Cell

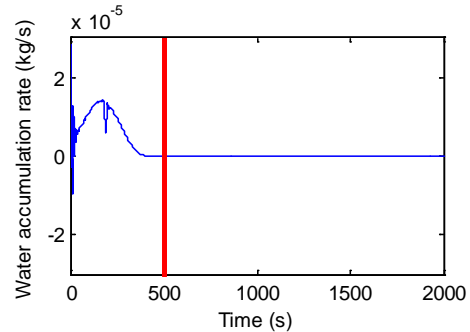
With the developed fuel cell stack model, the water balance at the cathode and anode can be evaluated with Eqs. (1) and (2). It should be mentioned that as the performance of the developed model has only been verified under normal operations, its performance under degradation conditions needs further verification, thus in this study, only the normal operation condition is used for analysis. Moreover, a constant current value of 20A is employed in the analysis, this value can give high cell voltage and low parasitic power demand, leading to about 50% system efficiency at full load, which is commonly used in practical fuel cell systems.

Fig. 3 depicts the cell voltage, and water accumulation rates at the anode and cathode with a cathode relative humidity (ratio of the partial pressure of water vapour in the mixture to the saturated water vapour pressure at same temperature and pressure) of 1, which can give optimal fuel cell performance. It can be observed that without fuel cell degradation, the individual cell voltage will reach a stable value, and the water accumulation rate approaches zero. However, unbalanced water content and increasing trend of cell voltage can be found at the beginning, which is

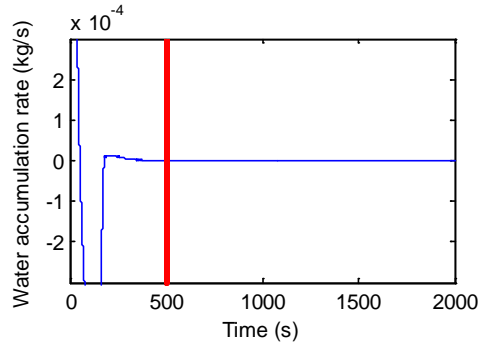
indicated in the figure reflecting the warm up stage and the stable state of the fuel cell stack (shown by the bold vertical line). The reason for this is that the fuel cell stack requires some time to reach the balanced state.



(a) Cell voltage



(b) Anode side



(c) Cathode side

Figure 3 Cell voltage and water accumulation rates under normal cell condition

By increasing the cathode relative humidity to 2 (under this condition, water will be accumulated inside the fuel cell stack, but the stack voltage will not decrease, this can be seen from Fig. 4a), more water may be kept at the cathode side, the performance of the cell model is analysed and results are shown in Fig. 4. From the results, the same value of cell voltage at the steady state stage can be seen, which indicates no degradation exists in the fuel cell. In this case, the water accumulation rates are still zero, meaning balanced water at the anode and cathode, even with liquid

water inside the cell. However, it should be noted that in this case, the fuel cell system requires a longer time to reach the stable state, which means the fuel cell system does not work under the optimum operation condition due to the increased cathode relative humidity.

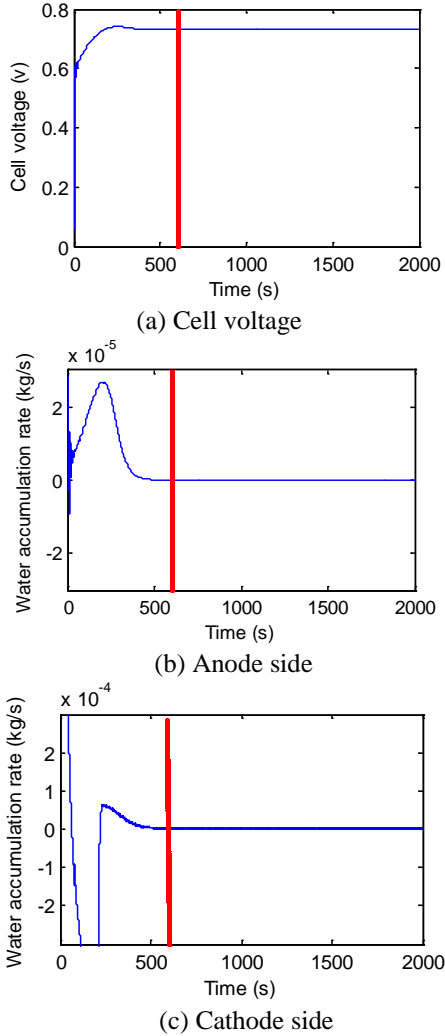


Figure 4 Cell voltage and water accumulation rates under normal cell condition with increased current

According to the results, it can be concluded that under fuel cell normal operation, water balance at the anode and cathode can be observed and water accumulation rates are zero. From these results, the flooding indicator is proposed for on-line diagnosis (as all parameters can be acquired directly and indirectly during operation, which is described in section 2), and its sensitivity to liquid water inside the fuel cell stack will be studied in following sections.

4. PROPOSED FLOODING INDICATOR AND ITS RELATIONSHIP WITH LIQUID WATER INSIDE THE FUEL CELL

As described in section 2, all terms in Eqs. (1) and (2) can be determined with measurements during fuel cell operation except water across the membrane, thus in order to use the water balance equations for on-line diagnosis, water across the membrane should not be used.

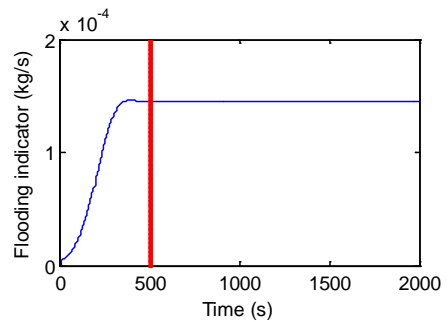
According to results in section 3, under normal operation conditions, Eqs. (1) and (2) should be zero, meaning zero water accumulation rates at anode and cathode. Therefore, the water accumulation rates can be used in this study to determine excess water inside the fuel cell stack, and the flooding indicator is proposed as the difference between the inlet and outlet water amount, which can be expressed by modifying Eqs. (1) and (2).

$$FL_{ca} = W_{v,in,ca} + W_{l,in,ca} + W_{v,gen} - W_{v,out,ca} - W_{l,out,ca} \quad (3)$$

$$FL_{an} = W_{v,in,an} + W_{l,in,an} - W_{v,out,an} - W_{l,out,an} \quad (4)$$

Where FL_{ca} , and FL_{an} are the flooding indicators for the cathode and anode sides, respectively. All other variables are as described in Eqs. (1) and (2).

Fig. 5 depicts the flooding indicator using Eqs. (3) and (4), and liquid water inside the anode and cathode with cathode relative humidity of 1. Results demonstrate that under normal conditions, flooding indicators from Eqs. (3) and (4) are the same, meaning balanced water condition inside fuel cell stack. Moreover, in this case, liquid water can not be found inside the cell, although at stack warm up stage, liquid water inside fuel cell stack can be observed.



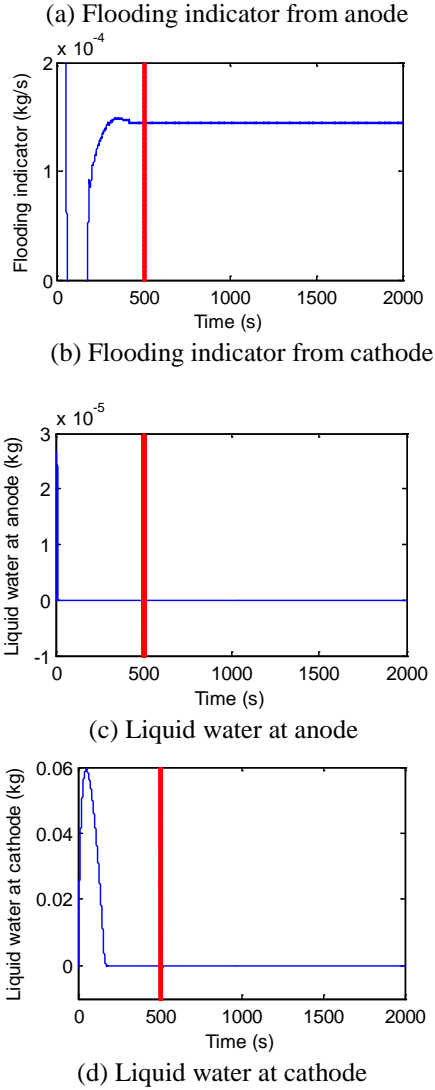


Figure 5 Flooding indicators and liquid water at anode and cathode

The sensitivity of the flooding indicator with liquid water inside fuel cell stack is investigated by increasing the cathode relative humidity to 2, flooding indicators are calculated and shown in Fig. 6, and liquid water at anode and cathode are also depicted.

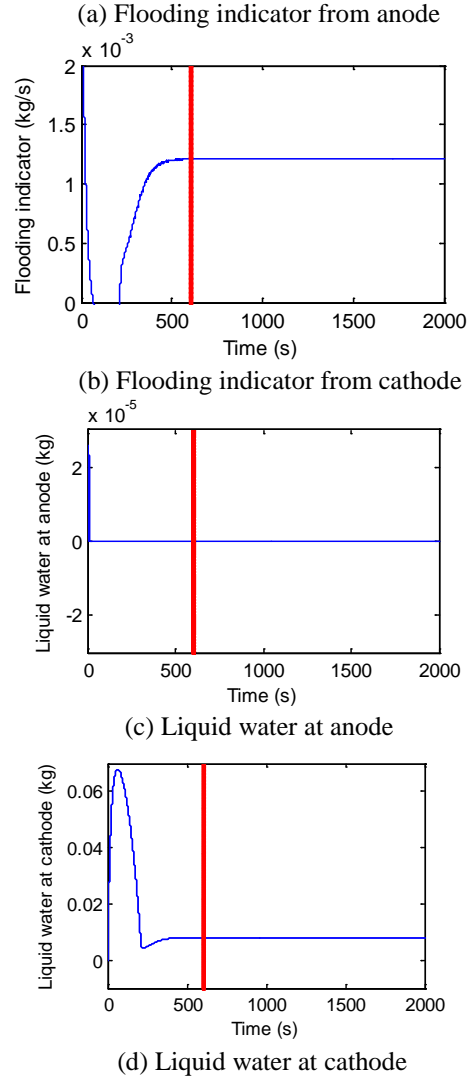
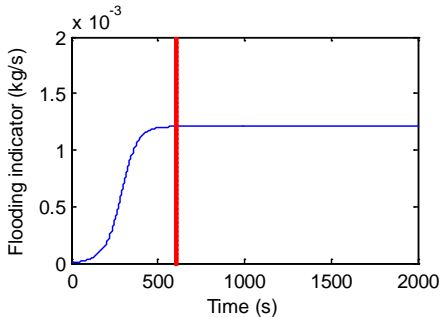


Figure 6 Flooding indicators and liquid water at anode and cathode with increased relative humidity

From figure 6(a) and 6(b), flooding indicators are about 10 times larger than that from the lower cathode relative humidity, but values from the anode and cathode are still the same, meaning water balance inside cell. Moreover, from Fig. 6(d), liquid water can be observed inside the cathode at stack steady state stage, although this does not cause a voltage drop and cell degradation, this further confirm that the fuel cell stack system does not work in the optimum operation condition. It should be noted that in this case, only the cathode relative humidity is increased, thus accumulated liquid water at anode is not found as shown in Fig. 6(c).

Moreover, with further increased cathode relative humidity, liquid water inside the cathode and corresponding flooding indicator are determined, and their relationship is depicted in Fig. 7. It should be mentioned that in these cases, the

water accumulation rate is still zero, which means no degradation within the fuel cell stack.

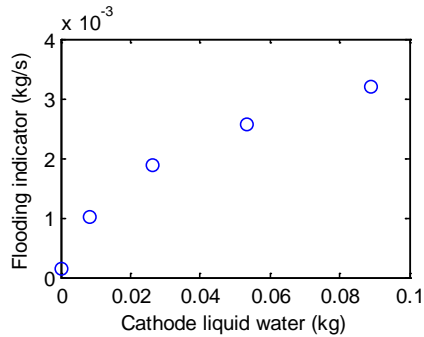


Figure 7 Relationship between liquid water inside cathode and flooding indicator

From figure 7, the flooding indicator will increase clearly with increased liquid water inside the cathode side, which further confirms the possibility of using the proposed indicator for evaluating liquid water inside the fuel cell stack, when the accumulated liquid water inside the fuel cell stack reach a certain level, fuel cell stack begins degradation. It should be noted that when the indicator is employed for flooding diagnosis, its threshold value should be determined to define the amount of liquid water causing fuel cell stack flooding, this value may change for different fuel cell stack systems. Moreover, it will be mentioned in the next section that the proposed indicator will give different values at anode and cathode sides in flooding case due to unbalanced water condition inside the fuel cell stack.

The results can also be explained from a theoretical point of view. Under normal operation condition, water accumulation rates at the anode and cathode are zero, thus the flooding indicator is actually the water across the membrane based on Eqs. (1) and (2). Based on previous studies (Ous and Arcoumanis, 2013, Wang et al. 2013), with an increase of reactant relative humidity, the water activity of the membrane will be increased, which will lead to a higher rate of water flux across the membrane.

Based on these results, it can be concluded that the proposed flooding indicator is sensitive to liquid water inside the fuel cell stack and can be used to indicate the liquid water condition. With increased liquid water inside fuel cell stack, flooding indicator will increase significantly. Moreover, as the required parameters in Eqs. (3) and (4) can be monitored continuously during fuel cell stack operation, the indicator can provide instant information about the accumulated liquid water inside the stack.

5. EXPECTATION OF PERFORMANCE OF PROPOSED FLOODING INDICATOR IN THE FUEL CELL FLOODING CASE

According to the results in sections 3 and 4, water accumulation rates at the anode and cathode are zero, and the flooding indicator equals the water across the membrane, thus cathode and anode flooding indicators should be the same under normal fuel cell stack operation, indicating the balanced water condition within fuel cell stack.

However, with fuel cell stack flooding, excess water will be kept inside the fuel cell stack, leading to an unbalanced water condition. In this case, the water accumulation rate should be increased, and by comparison of Eqs. (1)-(4), the flooding indicator should include water across the membrane and the water accumulation rate, thus its value will not follow the trend shown in Fig. 7 and should be increased significantly.

As described before, the flow rates of inlet water vapour and liquid water can be measured directly during fuel cell stack operation, and flow rates of outlet water vapour and liquid water, and generated water product inside fuel cell stack, can be determined using measurements from tests, including anode and cathode pressures, and current. Therefore, during fuel cell stack operation, the proposed flooding indicator can be monitored continuously, and a sudden increase of the flooding indicator value indicates excess water inside the fuel cell stack, thus flooding can be diagnosed.

Further, as the flooding indicator can be calculated at the anode and cathode using Eqs. (3), (4), respectively, the location of the fuel cell stack flooding can also be determined, since flooding can cause a higher value of indicator due to a faster water accumulation rate.

The performance of proposed indicator in on-line fuel cell stack flooding diagnosis is currently being investigated with measurements from a practical fuel cell stack system, and the results can be used to further validate the effectiveness of the flooding indicator.

6. CONCLUSION

In this paper, the mass balance equations at the anode and cathode are employed to indicate the water condition inside fuel cell stack, and the flooding indicator is proposed as on-line method to detect excess liquid water in the fuel cell stack, which can be used for diagnosis of fuel cell stack flooding.

Under normal operation conditions, the water accumulation rates at the anode and cathode are observed to be zero with a numerical study. Based on this, the flooding indicator is proposed as the difference between inlet and outlet water

amounts to express the liquid water condition at the anode and cathode. Under the normal operation condition, the proposed flooding indicator equals water across the membrane, and its performance is investigated using numerical studies by changing the cathode relative humidity. From the results, the flooding indicator is sensitive to the liquid water condition, it will be increased significantly with an increase of liquid water inside fuel cell stack.

Moreover, the performance of the proposed indicator in fuel cell stack flooding is predicted. Under the flooding condition, the flooding indicator includes water across the membrane and water accumulation rate, and excess water inside fuel cell stack will cause sudden increase of proposed indicator. By monitoring the flooding indicator during fuel cell stack operation, it is possible to detect the existence of stack flooding, and location of flooding can also be determined.

Further work will be performed to verify the effectiveness of this flooding indicator using test data from a practical fuel cell stack system, and the flooding indicator will be studied with a fuel cell stack model, which will be modified to express the fuel cell stack flooding scenario. It should be mentioned that in the practical application, the measurements will contain noises, thus before applying proposed indicator for diagnosis, signal processing techniques may be required to minimize the noise effect. Moreover, as in the practical fuel cell stack system tests, multiple degradation factors may occur simultaneously, thus a more robust flooding indicator will be investigated in order to work under more complex conditions.

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Lei Mao is born in Anhui province, China, in 1982. He received his B.E. degree in transportation engineering from Hefei University of Technology, Hefei, China, in 2004, M.S. degree in precision machinery and instrumentation engineering from University of Technology and Science, Hefei, Chian, in 2007, and PhD degree in civil engineering from The University of Edinburgh, Edinburgh, UK, in 2012.

During the period of 2012 and 2013, he worked as research associate at School of civil engineering and surveying, University of Portsmouth, UK.. From November, 2013, he works as research associate at Department of Aeronautical and Automotive Engineering, Loughborough University, UK.

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Andrey Vasilyev received MEng degree in systems and control engineering from the University of Sheffield, UK, in 2013. In October 2013, he continued his studies as a PhD research student at Loughborough University. His research interests include model and data based diagnostic techniques; PEM fuel cells and their health monitoring issues.