Study of Control Method for Proton Exchange Membrane Fuel Cell Stacks Using Overpotential Calculated from Curve Fitting

Yutaro Akimoto^{1*}, Keiichi Okajima¹

1 Graduate School of Systems and Information Engineering, University of Tsukuba

ABSTRACT

For the stable operation of proton exchange membrane fuel cells, measurements are made using sensors, and the controls are designed to detect, identify, and avoid defects based on the measured values. In this study, the control index using overpotential was calculated by the curve-fitting method. This method can increase the in-service operation and lower the cost because the sensors and measurement system are not used. The effectiveness of the proposed method for biased data is demonstrated, and the results are described.

Keywords: Fuel cell, Overpotential, Curve fitting, Control method

NONMENCLATURE

	Abbreviations	
2	PEMFC	Proton Exchange Membrane Fuel Cell
1	Symbols	
	V	Voltage [V]
ſ	- Eo	Initial voltage [V]
1	η_{act}	Activation overpotential [V]
_	$\eta_{_{ohmic}}$	Ohmic overpotential [V]
	$-\eta_{_{con}}$	Concentration overpotential [V]
	T	Temperature [°C]
- 1		

. INTRODUCTION

"Flooding" is one of the failures during the operation of proton exchange membrane fuel cells (PEMFCs). The generated water interferes with chemical reactions on the membrane electrode assembly surface and in the

fuel flow channels (also known as "plucking"). The other failure, "dry-out," is caused by the drying out of the membrane resulting from the excessive operating temperature and flow rates. The detection of these two contradictory failures is a key issue in the water management of PEMFCs. To solve this problem, various measurements are made using sensors. The controls have been designed to detect, identify, and avoid defects based on the measured values. For example, Song et al. used pressure sensors for water management of PEMFCs¹). Li et al. used many sensors for a data-driven approach²⁾. These methods increase the cost of PEMFC systems because they require a large number of sensors. To ensure reliability while lowering the cost of PEMFCs, it is necessary to measure the minimum number of sensors to avoid failures. For this purpose, the fuel cells must be diagnosed properly, and the control method based on these indicators must be considered.

One of the methods for diagnosing a fuel cell is to measure the overpotentials. In general, the output voltage of a fuel cell is calculated from the theoretical voltage and three overpotentials: activation, ohmic, and concentration.

$$V = E_0 - \eta_{act} - \eta_{ohmic} - \eta_{con} \tag{1}$$

A typical evaluation method is electrochemical measurement, such as the Cole–Cole plot³⁾. This measurement during operation is difficult because of the applied frequency and measurement time. There are also evaluation methods using curve fitting^{4–6)}. In these studies, the overpotential was diagnosed by the curve-fitting method using experimental results of the current and voltage characteristics that were not obtained during actual operation.

Selection and peer-review under responsibility of the scientific committee of the 12th Int. Conf. on Applied Energy (ICAE2020). Copyright © 2020 ICAE

The present curve-fitting method is used to diagnose the overpotentials calculated using currents, voltages, and temperatures during each operation. In this report, first, the curve-fitting equation used and its adaptation to in-service diagnosis are described. Subsequently, the effects of the adaptation strategy and a case study of inservice diagnosis and control of a fuel cell under constant current load are described.

2. EXPERIMENTAL SETTING

Figure 1 shows the system apparatus of the PEMFC. In this study, the PEMFC stack was used in all experiments. This stack was cooled by an air fan installed on the stack. Pure hydrogen was supplied from the cylinder in dead-end mode, and air was supplied by a pump. The cell voltage, stack current, and stack temperature were converted into voltage and connected to a PC via a data logger. In the PC, the program containing curve fitting by the least-squares method and the control strategy, which is described in Section 3, was implemented. The experimental results were fed back to the control unit and stored in the PC.

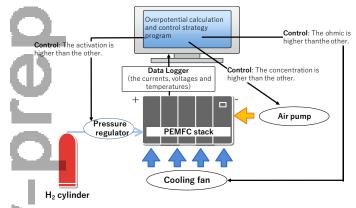


Fig 1 PEMFC and control system apparatus

. CONTROL METHOD AND STRATEGY

Figure 2 shows a flow chart of the control strategy. The voltage was set as the threshold to begin control. When the cell voltage drops below the threshold, controlling of the fuel cell starts. The control index is used as the difference between the initial and calculated values of the overpotential. This value is the greatest, and it is determined to be a significant voltage drop factor and is controlled accordingly. The activation overpotential can be reduced by increasing the operating temperature and pressure due to the catalytic activity, so that the fuel supply pressure is increased in the system. The ohmic overpotential is reduced by lowering the operating temperature owing to the drying of the ion exchange membrane. Because of this possibility, cooling is provided by a fan. The concentration overpotential is caused by the deviation of the reaction and flow rate and the reduction of the reaction surface area. This can be reduced by releasing the hydrogen outlet by purging and controlling the flow rate with an air pump.

Overpotentials were used to identify the output drop factor for the control. In this study, a previous fitting curve model was used for calculating the overpotentials shown in Eq. $(2)^{6}$.

$$V = E_{(T)} - \eta_{act(T)} - \eta_{ohmic(T)} - \eta_{con(T)}$$
⁽²⁾

This model equation is empirical. It differs from other fitting models^{4, 5)} because temperature as a function of each overpotential is included in this equation. Therefore, this model was shown to be the best fit and to be able to separate the overpotentials in previous studies^{6, 7)}.

In this study, there are two methods for the voltage used to separate the overpotentials. The first method uses all the measured data. The second method divides each current into a range and uses the voltage and temperature corresponding to the current range. This categorized method prevents bias in the acquired values when load variations occur.

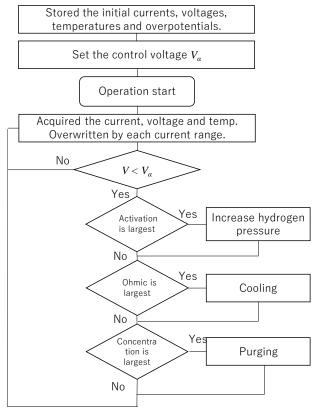


Fig 2 Flow chart of the control strategy

4. **RESULTS AND DISCUSSIONS**

4.1 Effect of overpotentials obtained using these methods

Figure 3 shows a comparison of three fitting I-Vcurves: the previous manual method⁶⁾, the method using all the data acquired every second, and the method proposed in Section 3. The previous and proposed I-V curves show a similar trend. However, the fitting results using all the data show a different curve. This is due to the bias of the data.

Figure 4 shows the plot data for each method. Although the plotted positions of the previous (blue) and proposed (green) methods are close, using all the data (red) plotted, especially in the low current density range, results in 52% of the plots being below 50 mA/cm². This makes it difficult to separate the overpotentials from the acquired data.

Figure 5 shows the overpotentials of each method at 500 mA/cm². The largest difference between the proposed and the previous overpotentials was 0.05 V for the activation. The rest of the overpotential was less than 0.01 V. However, using all the data overestimated the activation overpotential, and the other underestimated it. These results show that the proposed method can maintain the same overpotential separation accuracy, even with biased measured data.

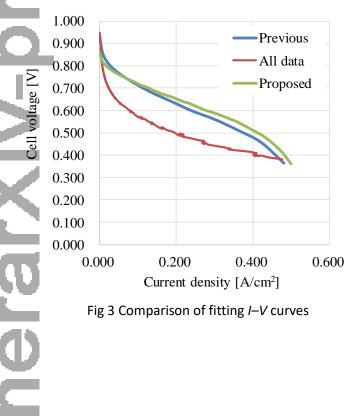


Fig 3 Comparison of fitting I–V curves

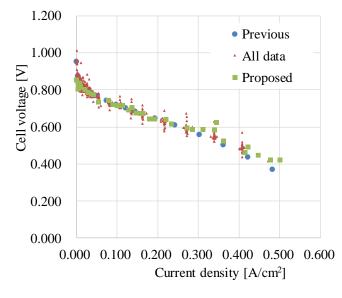


Fig 4 Plot data for each method

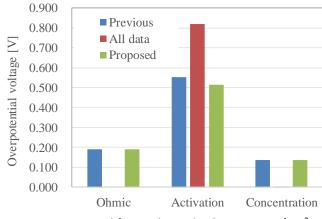


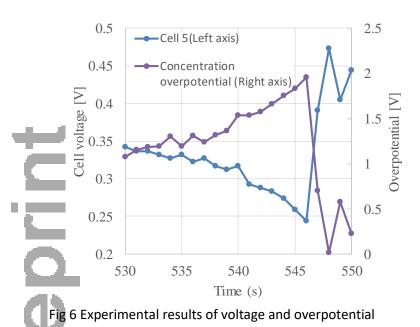
Fig 5 Overpotential for each method at 500 mA/cm²

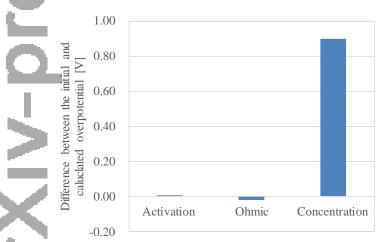
4.2 Failure detection of constant current

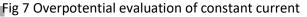
The control threshold of cell voltage V α in the PEMFC stack was set to 0.25 V. In this section, the current was constant at 20 A. The acquired voltage and temperature were overwritten at this ampere level. When the voltage fell below the control threshold V α , the control was performed by overpotential comparison. The operation was performed under flooding conditions, such as low air flow rate and low operating temperature.

Figure 6 shows the cell voltage and concentration overpotential. The cell voltage gradually decreased and the concentration overpotential increased under flooding conditions. When the operation time was 546 s (the control strategy), purging was activated because the cell No. 5 voltage was 0.244 V in the experiment. Then, the concentration overpotential was decreased, and the cell No. 5 voltage recovered. Figure 7 shows the

difference between the initial and calculated overpotentials at 546 s. The ohmic overpotential decreased, while the activation and concentration overpotential increased. At 546 s, the concentration overpotential was larger than the other overpotential. Therefore, purging was performed, and the output voltage was recovered. In these results, failures, such as flooding, were detected and controlled using the proposed method.







CONCLUSIONS

In this study, the PEMFC was controlled using overpotentials calculated by the curve-fitting equation. The proposed method made it possible to maintain the same overpotential separation accuracy with the biased measured data. In failures such as flooding, the method was activated, and it recovered the output voltage. These results demonstrate the effectiveness of the proposed strategy and its viability at constant current.

ACKNOWLEDGEMENT

This work was supported by Yashima Environment Technology Foundation.

REFERENCE

[1] Song M, Pei P, Zha H, Xu H. Water management of proton exchange membrane fuel cell based on control of hydrogen pressure drop. *J Power Sources*.
2014;267:655-663. doi:10.1016/j.jpowsour.2014.05.094
[2] Li Z, Outbib R, Giurgea S, Hissel D, Giraud A, Couderc P. Fault diagnosis for fuel cell systems: A data-driven approach using high-precise voltage sensors. *Renew Energy*. 2019;135:1435-1444. doi:10.1016/j.renene.2018.09.077

[3] Pérez-Page M, Pérez-Herranz V. Study of the electrochemical behavior of a 300 W PEM fuel cell stack by Electrochemical Impedance Spectroscopy. *Int J Hydrogen Energy*. 2014;39(8):4009-4015. doi:10.1016/j.ijhydene.2013.05.121

[4] J.Kim, S.M. Lee, S.Srinivasan GLGB. Modeling of Proton Exchange Membrane Fuel Cell Performance with an Empirical Equation. *J Electrochem Soc*. 1995;142(8):2670-2674.

[5] Pisani L, D'Aguanno B. A new semi-empirical approach to performance curves of polymer electrolyte fuel cells. *J Power Sources*. 2002;108:192-203.
[6] Akimoto Y, Okajima K. Semi-Empirical Equation of PEMFC Considering Operation Temperature. *Energy Technol Policy*. 2014;1(1):91-96.
doi:10.1080/23317000.2014.972480
[7] Balogun EO, Hussain N, Chamier J, Barendse P.

Performance and durability studies of perfluorosulfonic acid ionomers as binders in PEMFC catalyst layers using Electrochemical Impedance Spectroscopy. *Int J Hydrogen Energy*. 2019;44(60):32219-32230. doi:10.1016/j.ijhydene.2019.10.079