Journal of Power Sources 326 (2016) 264–269

Contents lists available at ScienceDirect

Journal of Power Sources

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

On-line and real-time diagnosis method for proton membrane fuel cell (PEMFC) stack by the superposition principle

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HIGHLIGHTS

- A PEMFC diagnosis is developed based on the superposition principle.
- The diagnosis can be used for on-line and real-time PEMFC diagnosis.
- A stack voltage is merely used for the diagnosis.
- The diagnosis is experimentally proved using the highway fuel economy test cycle.

ARTICLE INFO

Article history:
Received 9 February 2016
Received in revised form
23 June 2016
Accepted 27 June 2016

Keywords:
PEMFC stack
On-line diagnosis
Critical cell voltage
Superposition principle

ABSTRACT

The critical cell voltage drop in a stack can be followed by stack defect. A method of detecting defective cell is the cell voltage monitoring. The other methods are based on the nonlinear frequency response.

In this paper, the superposition principle for the diagnosis of PEMFC stack is introduced. If critical cell voltage drops exist, the stack behaves as a nonlinear system. This nonlinearity can explicitly appear in the ohmic overpotential region of a voltage-current curve. To detect the critical cell voltage drop, a stack is excited by two input direct test-currents which have smaller amplitude than an operating stack current and have an equal distance value from the operating current. If the difference between one voltage excited by a test current and the voltage excited by a load current is not equal to the difference between the other voltage response and the voltage excited by the load current, the stack system acts as a nonlinear system. This means that there is a critical cell voltage drop. The deviation from the value zero of the difference reflects the grade of the system nonlinearity. A simulation model for the stack diagnosis is developed based on the SPP, and experimentally validated.

1. Introduction

To resolve global environmental problems, many automobile companies are making great efforts to develop so-called green cars. One of the solutions is the electric vehicle equipped with a battery and/or PEMFC stack.

The sustainable performance of electrical energy system should be based on on-line diagnosis as well as optimal control of the process parameters. Therefore, the diagnosis of the state of health (SOH) of the systems is getting more important. And two factors are crucial for the market predominance of a fuel cell electric vehicle (FCEV). The one is durability, which is guaranteed by proper diagnosis methods and optimal control. And the other is cost reduction of the next-generation fuel cell electric vehicle, which can be achieved by reducing the platinum-loading [1] or using non-noble metal as a catalyst.

The information gained from a diagnosis module can be passed to a decision module to decide whether an abnormal state of the stack is reversible or irreversible. If the state of the stack is correctable, this information is sent to an electronic control unit (ECU) of the stack to recover the present state. For this, the real-time and on-line diagnosis method is implemented.

The electrochemical impedance spectroscopy (EIS) is an established conventional method to detect process parameters and to diagnose stack [2]. Although EIS is a verified diagnostic tool for characterizing the performance of the stack, it is not suitable for on-line diagnosis. Another diagnosis method is using a nonlinear frequency response [3].

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http://dx.doi.org/10.1016/j.jpowsour.2016.06.113
0378-7753/© 2016 Published by Elsevier B.V.

Nomenclature

CVM	cell voltage monitoring
DAQ	data acquisition
DC	direct current
EIS	electrochemical impedance spectroscopy
ECU	electronic control unit
FCEV	fuel cell electric vehicle
HWFET	highway fuel economy test
I_a, I_b	DC test input current
I_s	stack operating current
ΔI	difference between DC test input current and stack operating current
IM	intermodulation
MMI	man-machine interface
PEMFC	proton exchange membrane fuel cell
S	linear system response
SOH	state of health
SPP	superposition principle for diagnosis PEMFC stack
SVD	stack voltage difference
THD	total harmonic distortion
V	voltage, V
V(x)	stack voltage response at the current x
VCC	voltage-current curve

The current method for monitoring the cell voltages in a stack, for example, to detect an abnormal voltage drop, is to measure each cell voltage. It is named cell voltage monitoring (CVM), which is mechanically and electrically very cumbersome and complex.

A non-linear method to detect abnormal and irreversible cell voltage drop is to measure the stack voltage instead of CVM [4]. The following two diagnosis methods are based on the nonlinearity of the stack characteristic due to an abnormal cell voltage drop. The method is to detect the nonlinearity of the stack system at a nominal operating point which is normally located in the ohmic overpotential region [5]. In the area of the electronic audio amplifier design, the distortion of the amplifier is measured by the intermodulation (IM) method, in which the two sinusoid test input signals are given and the response signal is measured in the time domain and then this signal is transformed into frequency domain by the Fourier transformation. If nonlinearity exists in the amplifier, the harmonics appear in addition to the input signal frequencies in the frequency spectrum. The grade of nonlinearity is dependent on the value of the total harmonic distortion (THD). This method of THD analysis is adopted for the stack diagnosis. The two sinusoid test current signals are inputted to a stack system, and the stack voltage signal as the response is measured in the time domain, and this signal is transformed into the frequency domain subsequently. The nonlinearity can be detected according to the existence of harmonics. The grade of the nonlinearity, by which the intensity of the critical voltage drop can be described, is dependent on the THD value [6].

In this paper, the other on-line and real-time diagnosis method of detecting defective cells is presented, which is based on the superposition principle. The superposition principle (SPP) states that, for every linear system, the output response by two inputs is the sum of the responses which are the outputs by each input separately. This method can be applied for detecting the nonlinearity of the stack system. From here on, this diagnosis method is referred to as "diagnosis by the superposition principle". The linear case, defined as a normal state, and the nonlinear case of the stack, defined as an abnormal state, are shown schematically in Fig. 1. The abnormal state of the cell may be defined when the cell voltage is smaller than 0.5 V at the load current density of 1 A/cm^2, which is referred to as a nominal current density of an FCEV. The objective of this investigation is mainly to detect the defective cell(s) due to stack degradation. The causes of the degradation are not in the scope of our study. For this, the process parameters are kept at the nominal values given by the stack manufacturer due to the fact that the change of a process parameter leads to a cell voltage drop temporary. We developed a simulation model and conducted the simulation for the SPP method. We also proved the practical applicability of the method experimentally.

1.1. Basic superposition principle equations for the stack diagnosis

A linear system, denoted by S, satisfies the property of superposition, which is defined as the Eq. (1)

$$\mathrm{S(a)} + \mathrm{S(b)} = \mathrm{S}(a + b), \tag{1}$$

where a and b are arbitrary inputs.

The equation (1) can be rewritten in Eq. (2).

$$V(I_a) + V(I_b) = V(I_\mathrm{a} + I_\mathrm{b}), \tag{2}$$

Where I_a or I_b is direct current (DC) test input current and $V(I_\mathrm{i})$ is the stack voltage as the response.

Eq. (2) must be satisfied if a stack is operated in the area of the linear ohmic overpotential region, in other words, the linear transfer region as shown in Fig. 1.

For practical purposes, however, Eq. (2) can be modified to maintain the small test input current (ΔI) as shown in Eq. (3).

$$\{V(I_s - \Delta I) - V(I_s)\} - \{V(I_s) - V(I_s + \Delta I)\} = 0, \tag{3}$$

where $V(x)$ is a stack voltage response at the current x, I_s is a stack load operating current and ΔI is the difference between a DC test input current and a stack operating current, which is held small for practical point of view.

Another formulation of Eq. (3) is needed for the fast change of one signal direction of the test currents, here "$+\Delta I$", as follows,

$$\{V(I_s) - V(I_s + \Delta I)\} - \{V(I_s + \Delta I) - V(I_s + 2\cdot\Delta I)\} = 0. \tag{4}$$

The stack voltage difference (SVD) is finally defined as the grade of the nonlinearity of a stack as given by Eq. (5).

$$\mathrm{SVD} = \{V(I_s) - V(I_s + \Delta I)\} - \{V(I_s + \Delta I) - V(I_s + 2\cdot\Delta I)\}. \tag{5}$$

If a stack operating current is in the area of the linear ohmic overpotential region and all cells in a stack have normal cell voltages, the SVD is zero.

2. Simulation

The model based on an electrochemical model is shown in Fig. 2, which is modified for SPP diagnosis method simulation and transformed to Matlab© and Simulink© [6], [7]. The electrochemical parameters, the number of abnormal cells and the patterns of the test current can be varied in order to simulate different abnormal

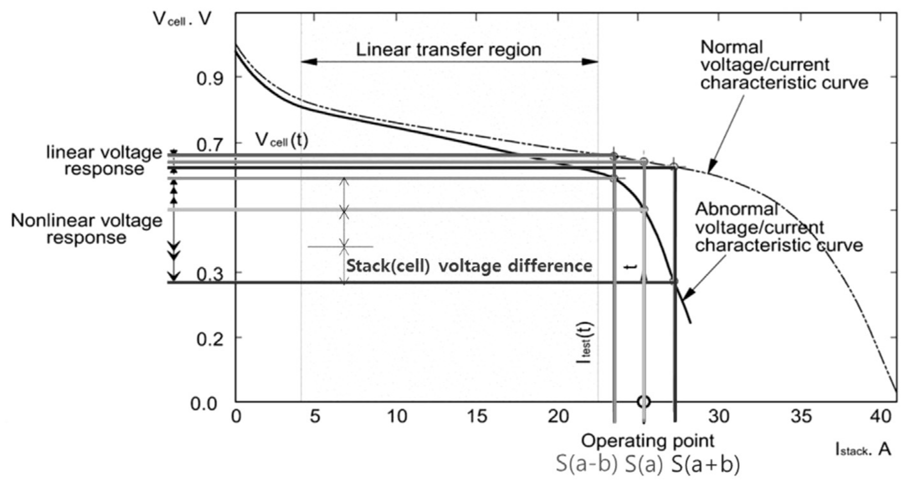

Fig. 1. A voltage-current characteristic curve of a cell/stack.

states. The outputs are stack voltage, cell voltages, SVD and the amplitude spectrum of the stack voltage.

In this simulation, stack model consists of 48 cells, and the number of the abnormal cells can be varied. The typical two voltage-current curves (VCC), one for a normal cell as shown in Eq. (6) and the other for an abnormal cell as shown in Eq. (7), are modeled by a curve fitting method.

$$y = -0.086x^3 + 0.26x^2 - 0.37x + 0.87 \tag{6}$$

$$y = -x^3 + 1.5x^2 - 0.96x + 0.91 \tag{7}$$

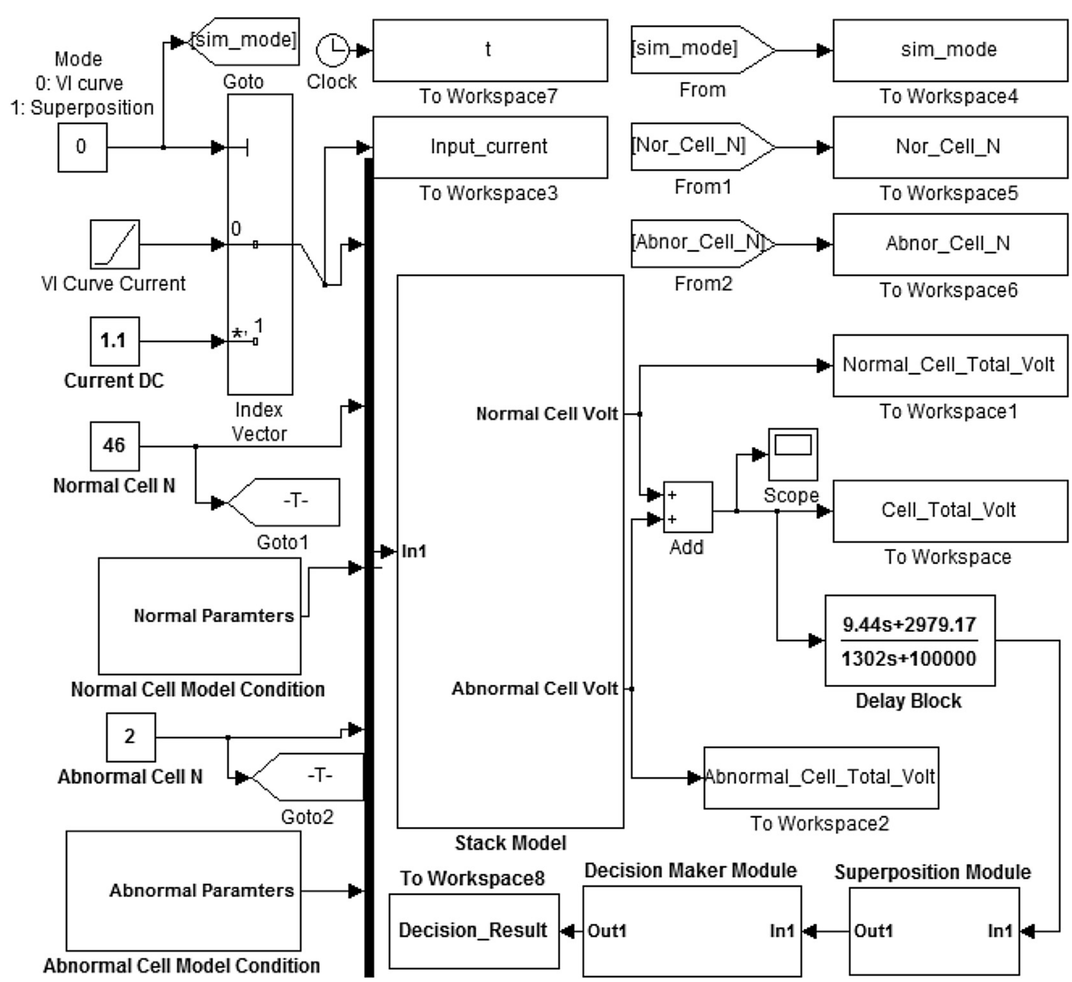

Fig. 2. The electrochemical model (Matlab/Simulink©).

The simulation is carried out with variations of the number of the abnormal cells from zero to 24, while the all electrochemical parameters are held constant. The operating load current is 6 A and the test current is 0.18 A, which is the 3% of the operating current and is determined due to fuel efficiency.

The SVD value of the normal stack is 4.28 mV, which should be zero, if a system is linear. However, as is generally known, the VCC in the ohmic overpotential region is not exactly linear. The SVD values of the abnormal state are larger those that of the SVD of a normal stack and are dependent on the number of abnormal cells. The SVD value of the abnormal stack is linearly dependent on the number of the abnormal cells. The functional relationship between SVD (y) and the number of abnormal cells (x) is denoted in Eq. (8).

$$y = 8.44x + 4.28. \tag{8}$$

3. Experimental

There are two kinds of fault which are identified by a diagnosis system. One is a reversible fault, which is an operative error and is correctable by a control system. And the other is an irreversible fault, which is important for maintenance of FCEV maker. At the first experimental stage, an abnormal state of a stack is emulated by varying the air supply. That is a simulation for fuel starvation, an exemplarily operative fault. The experimental validation of the SPP algorithm is conducted to detect an irreversible faulty cell, defined as "abnormal cell". The next stage of the experiment is a SPP static test, which is carried out at a given operating current. The dynamic characteristic of SPP is also analyzed, for which a dynamic driving cycle is used.

These investigations are led with a PEMC stack, H-200, which is commercially made by the company Horizon©. It has 48 cells with the active area of ca. 8 cm^2, maximum electrical power of 200 W. The operating temperature is ca. 65° and the maximum efficiency is 40% at 19.2 V. The fuel cell stack operates at the hydrogen pressure from 0.45 bar to 0.55 bar, is self-humidified, cooled and fed with air laterally by an air blower. The two same stacks are built in parallel to compare the output responses under the identical process parameters. One stack is for a normal state and the other is for an abnormal state, which means 4 cells are defective of the 48 cells. The process parameters are kept at the nominal values for this investigation given by the stack manufacturer. It is justified that the change of a process parameter is affected by a cell voltage drop temporary. The purpose of the investigation was to detect the defective and irreversible cell voltage drop. The other electrochemical characteristics are not considered in the experiments due to our black box approach for the diagnosis.

The process parameters of the test bench are controlled and measured by National Instrument©'s data acquisition system (DAQ), CompactRio™, which has analog inputs including high voltage and current measurement, and analog outputs for controlling the electrical load and the mass flow controller and also has digital inputs and outputs. The resolution of the A/D converter is 12-bit and the DAQ sampling rate is 250 kS/s. The load current and the test current for SPP is controlled by an electrical load, Agilent© 6060 B (max. 300 W). The block diagram of the test bench is illustrated in Fig. 3.

3.1. SPP diagnosis experiment with the emulation of an abnormal state

The abnormal voltage drop can be emulated by temporarily stopping the air supply. That is the situation of fuel starvation and an operative fault. The purpose of this experiment is a feasibility study of the SPP algorithm without any modification of stack hardware construction. In section 3.2 and 3.3, the investigations are conducted using the stack with defective cells. This emulation begins in a normal state and changes into an abnormal state in 65 s, finally returns to the normal state again in 384 s as described in Fig. 4. The change of the SVD value is illustrated ranging from ca. 0 V to maximum 0.4 V and finally back to 0 V. The validity of the SPP

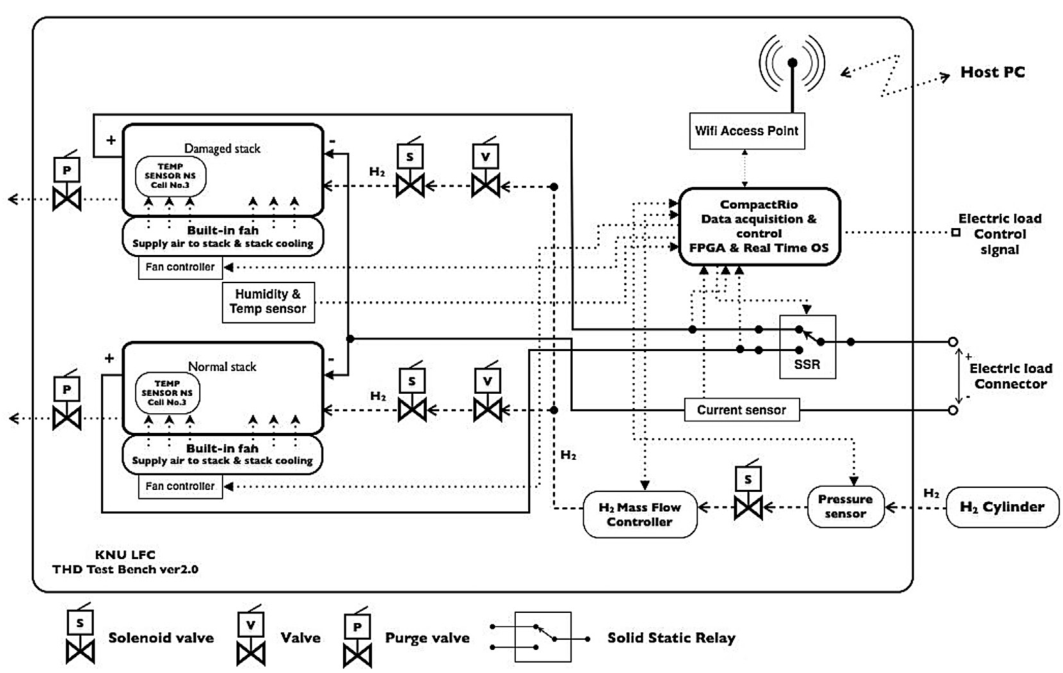

Fig. 3. The block diagram of the test bench.

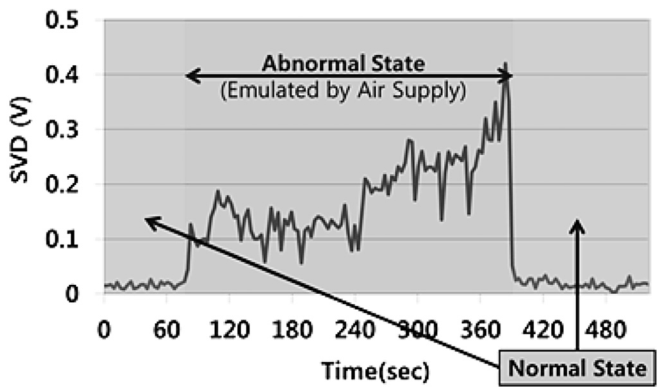

Fig. 4. A SPP experimental result emulated by varying the air supply.

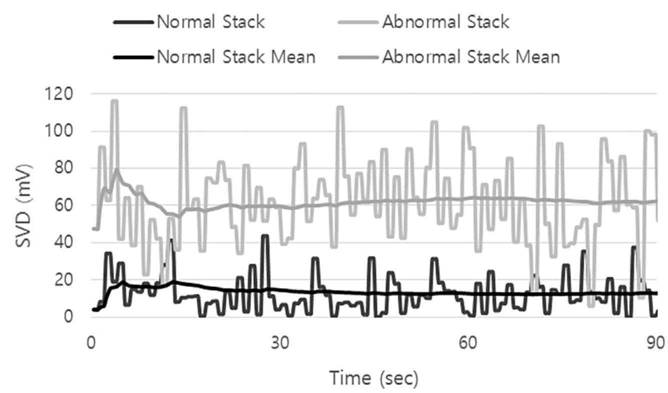

Fig. 6. Static SPP experiment on a normal stack and an abnormal stack.

algorithm is consequently validated by this result.

3.2. SPP diagnosis experiment— static analysis

The two stacks are employed in the test bench for this experiment to compare the SVD values. One is the normal stack which has all normal cells. The other stack is labeled as abnormal, if the stack has defective cells as described above in section 3. The abnormal stack is intentionally built with defective cells whose voltages are smaller than 0.5 V at the nominal operating current density, ca. 1 A/cm^2, and it is operated under the determined process parameters given by the stack manufacturer in order to eliminate the side effect during the test of the SPP diagnosis. The VCC's of these stacks measured are shown in Fig. 5.

Fig. 6 shows the SVD of a normal stack and that of an abnormal stack on the same operation condition. The operating current and ΔI are 6 A and 0.18Arespectively.The two stack voltage responses are compared. The time-averaged, from 0s to 90s, SVD values of the normal and the abnormal stack are12.73 mV and 63.37 mV respectively. The difference of two SVDs is 50.64 mV. If the stack behaves perfectly linear, the SVD value must be zero. The main issue of this study is the fact that if the difference of two SVDs is nonzero value, then there is defective cell(s) in the stack. Thereby, the SPP algorithm is validated.

3.3. SPP diagnosis experiment - dynamic experiment using a driving cycle

A driving cycle of a vehicle, which is called "the highway fuel economy test (HWFET)", is taken to conduct a dynamic SPP experiment. In Fig. 7, the driving cycle of a vehicle denoted by the vehicle speed (HWFET) and the correspondent fuel cell stack load current are shown. As the case of static SPP experiment, the two stacks in the test bench are installed for this experiment to compare the SVDs.

For real FCEV application, a diagnosis algorithm should be operated real-time, on-line and tested and proved in a dynamic operating condition. It means that an algorithm can be performed on the condition that the stack current is dynamically varied as shown in Fig. 7.

The load current range in Fig. 7 is reduced to 7% of a current of a FCEV to be adapted to the specification of test bench stack. To apply the SPP algorithm for the dynamic driving cycle test, the operating current is sampled. The sampling interval of the driving current is ca. 280 ms inclusive sampling time and hardware delay of the data acquisition system. The three voltage responses, $V(I_s)$, $V(I_s + \Delta I)$ and $V(I_s + 2\cdot\Delta I)$ in Eq. (5), are measured according to the currents (I_s,$I_s + \Delta I$ and $I_s + 2\cdot\Delta I$). The VCC progression in Fig. 7 contains the low and the high current, which represent the activation and the concentration overpotential region respectively.

Fig. 8 is the dynamic response of SVD in the SPP experiment. The dynamic characteristic of the SVD differs from that of the static experiment in section 3.2 due to the low and the high operating current. The instantaneous SVD values are dependent on the driving current and varied rapidly. These SVD values are time-averaged to be clearly distinguishable. The time-averaged SVD value of the normal and the abnormal stack are43.59 mV and 160.67 mV respectively. The difference of the SVDs is117.08 mV.

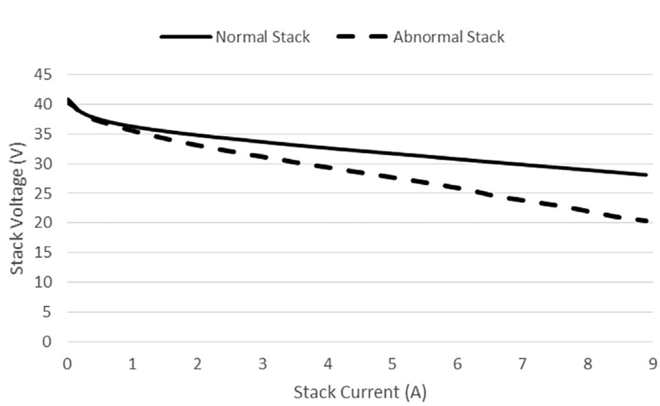

Fig. 5. The VCC of the two stacks for the experiment.

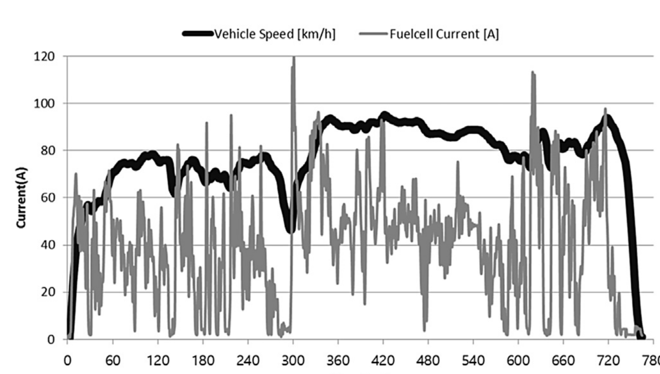

Fig. 7. The driving cycle of a vehicle denoted – the vehicle speed (HWFET) and The correspondent fuel cell stack load current.

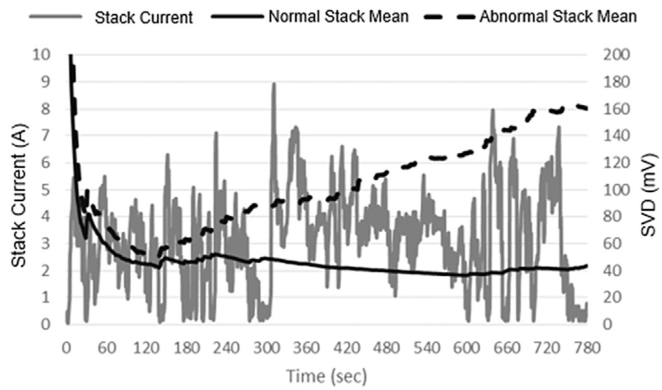

Fig. 8. The dynamic SVD using HWFET driving cycle.

The SPP diagnosis method in the dynamic case is verified successfully.

4. Conclusion

The main objective of this investigation is to detect the defective cell(s) due to stack degradation. A VCC of a defective cell shows nonlinear characteristics in the ohmic overpotential region. The proposed SPP diagnosis algorithm is based on the fact that the SPP is only valid, when a system is linear. If one or more critical cell voltage drops exist, which means that the cells are defective, the stack behaves as a nonlinear system. In this paper, only one type of critical and irreversible voltage drop is analyzed to verify the functionality of the SPP diagnosis method without any consideration of an abnormal change of the process parameters. The SPP method is applicable to an on-line and a real-time diagnosis, which is the requirement for real-world application, for example for FCEV.

The usability of the method is showed by the simulation and experiments on the test bench. The two types of stacks are used for experimental proof. One is normal stack and the other is abnormal stack with defective cells. The static experiment and the dynamic experiment are conducted to verify the SPP diagnosis method.

The SVD value defined in this paper is an indicator for nonlinearity and the existence of defective cells in a stack as well. The time-averaged SVD values of a static SPP experiment for both stacks are 12.73 mV and 63.37 mV each. The difference between both SVD values is 50.64 mV. And a dynamic driving cycle, HWFET, is taken to conduct a dynamic SPP experiment. The SVD time-averaged values of the normal and the abnormal stack are compared in the dynamic SPP experiment. The value of the dynamic characteristic of SPP diagnosis method is 43.59 mV and 160.67 mV respectively. The difference between both SVD values is 117.08 mV. There is discrepancy in the SVD values of the static and the dynamic case as described above. The causes of this discrepancy can include the difference between the operating conditions, the alteration of the operating current in the case of the dynamic operation and non-ideal-linearity of the VCCs, which differs from what we assumed. The investigation of this diagnosis algorithm on a FCEV is under way. Our future study will also include an intelligent decision maker for enhanced diagnosis approach based on fuzzy logic and on artificial neural network.

References

[1] Seungyeol Yoo, Young-Hyun Lee, Methods for diagnosis and minimum PT-loading of PEM fuel cell stack, Trans. KIEE 62 (1) (2013) 105–108.

[2] Xiaozi Yuan, Jian Colin Sun, Mauricio Blanco, Jaijiang Wang, Jiujun Zhang, David P. Wilkinson, AC impedance diagnosis of a 500W PEM fuel cell stack Part I: stack impedance, J. Power Source 161 (2006) 920–928.

[3] Thomas Kadyk, Richard Hanke-Rauschenbach, Kai Sundmacher, Nonlinear frequency response analysis of PEM fuel cells for diagnosis of dehydration, flooding and CO-poisoning, J. Electroanal. Chem. 630 (1) (2009) 19–27.

[4] D. Web, S. Moller Holst, Measuring individual cells voltages in fuel cell stacks, J. Power Sources 103 (1) (2001) 54–60.

[5] Erich Ramschak, Volker Peinecke, Peter Prenninger, Thmas Schaffer, Wolfgang Baumgartner, Viktor Hacker, Online stack monitoring tool for dynamically and stationary operated fuel cell systems, Fuel Cells Bull. (2006) 12–15.

[6] Young-Hyun Lee, Seungyeol Yoo, JongHyeon Kim, Development of real-time diagnosis method for PEMFC stack via intermodulation method, Trans. KSAE 22 (7) (2014) 76–83.

[7] Youngwoo Noh, SaeHoon Kim, KwiSeong Jeong, Ikjae Son, Kookil Han, Byung Ki Ahn, Modeling and parametric studies of PEM fuel cell performance, Trans. Korean Hydrogen New Energy Soc. 19 (3) (2008) 209–216.