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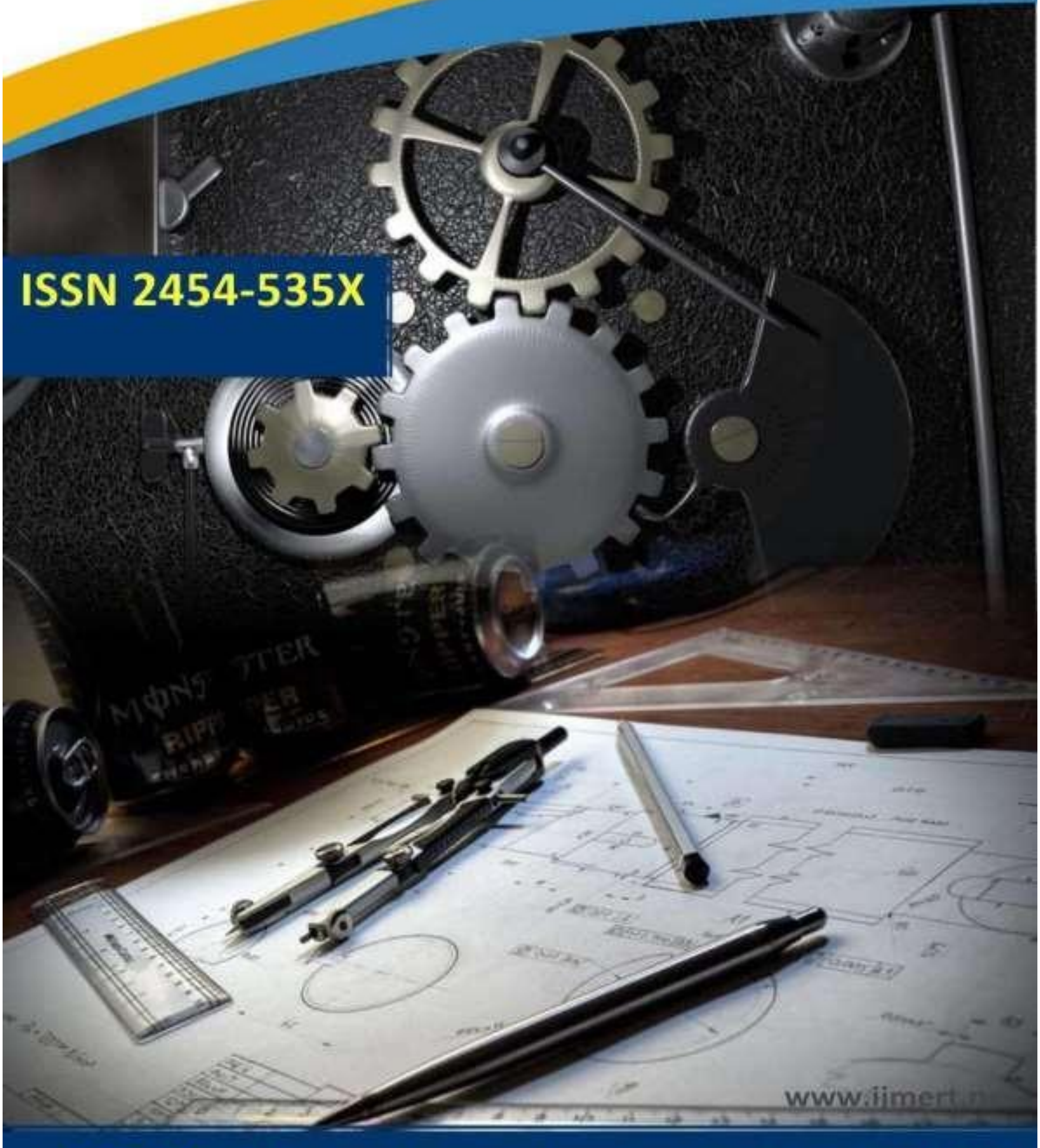

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Understanding the Distribution of Degradation Factors via Enhanced Diagnostics

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ABSTRACT: With the use of resistance, activation, and diffusion polarizations, our diagnostics assesses the transient reaction of the PEFC and uses this response to produce diagnostics parameters. By using this diagnostics method on PEFC short stack or PEFC single cell with a quadrisection separator, we have been able to clarify the distribution and degradation factors (the three polarizations). Ultimately, Tsuruga Electric Corporation has used this diagnostic technique to sell maintenance equipment under the PEFC Performance Diagnostics System (PPSD) "356 TD" name. The cost of conventional diagnostic equipment is prohibitive, and the analysis time is excessive. Nevertheless, our device, the 356TD, is far less expensive than traditional ones and can identify deterioration factors and do an impedance study by monitoring the transient response for roughly a minute. But given that the reflection of

INTRODUCTION

Polymer Electrolyte Fuel Cells (PEFCs) have gained popularity recently and are being used in several industries, including domestic power generation and the automobile industry, due to their low emission and ability to function at low temperatures. PEFC still faces a few issues, nevertheless, including the price of fuel cell products, how to increase life performance, and how to produce basic maintenance tools, among other issues. Our group has spent a great deal of time studying hydrogen synthesis and fuel cell technology. One group has developed a catalyst layer with self-water management since 2015, while another group has produced a separator with self-water management since 2003, with the goal of lowering the cost of the PEFC system. Our team focuses oncomposed of four cells and a PEFC single cell with a quadrisection separator as shown in Figure 1 and 2, and it found the deteriorated cell in the short stack, and it also obtained the degradation distribution in the same cell [3, 4]. However, because the reflection of each deterioration factor in each parameter was small, there is a possibility of causing the diagnostic error. Therefore, a new algorithm for PEFC performance diagnostics has been developed by solving the transient response using the third differential equation directly. A new diagnostic approximates the transient response of PEFC directly using the on-line algorithm of the method of least squa res, and obtains three diagnostic parameters sequentially. The adequacy of a new algorithm is verified by eva lua t ing the cell inner heat flux distribution using heat flux sensors and a special separator

that can measure heat flux distribution as shown in Figure 3 and 4, respectively. the separator has four spaces for installing the heat flux sensor to the back of each electrode as shown in Figure 4 (b). After installing each heat flux sensor as shown in Figure 4 (c), each space was buried so that there was no gap by the carbon block as shown in Figure 4 (d). By installing the sensors close to the reaction area, the sensors could measure how much heat flows from inside to outside and from outside to inside. Figure 5 shows Cross diagram of PEFC.

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Figure 1. Photograph of PEFC short stack

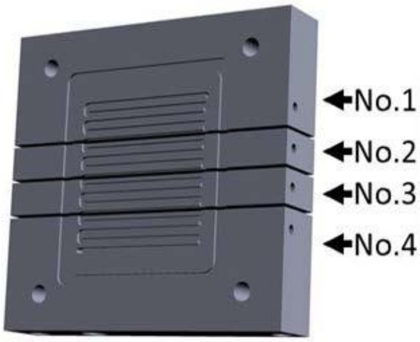
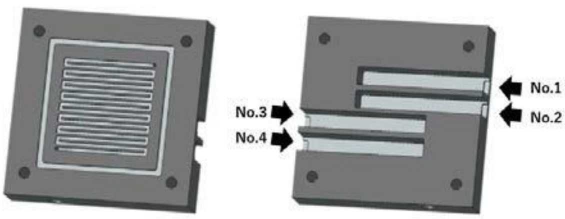
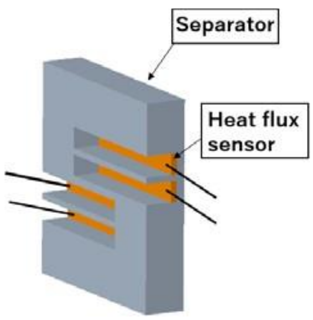


Figure 2. Schematic diagram of four segmented separator

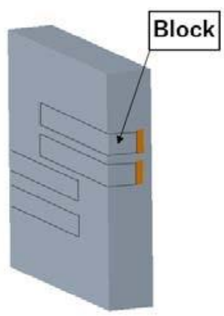
Figure 3. Heat flux sensor



(a) front view (b) back view



(c) Separator installed heat flux sensors



(d) Separator installed heat flux sensors and blocks

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Figure 4. Schematic diagram of separator for measuring heat flux

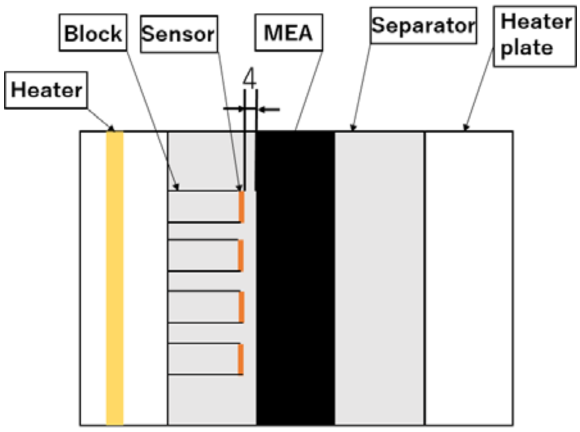

Figure 5. Cross diagram of PEFC

EXPERIMENT

Figure 6 shows the experimental apparatus for the evaluation of the cell performance. Figure 6 (a) a nd (b) show the experimental apparatus for a single cell and a short stuck respectively. The separator was made of carbon a nd have an effective electrode area of 25cm^2 and a serpentine gas channel of 1mm×1mm. Here, each portion of separator used this paper from the top end was numbered from No.1 to No.4. Anode gas and cathode gas were humidified by passing through the humidifier and were supplied to the cell. In order to prevent dew form a t ion in the supply pipe, the piping between the humidifier and the cell was made shorter and was maintained to 80℃with a ribbon heater. A cell temperature, a cell voltage and four heat flux were recorded in a computer online by a da ta logger. A milliohm meter with AC 4 probes was used to measure a cell resistance. The PEFC Performance Diagnostic System (PPDS; 356TD) composed of an A/D converter and a pulse generator measured a transient response of PEFC during 28.2msec. The obtained transient response was converted to CSV data a nd sent to the PC by USB cable, and each fitting parameter was delivered by analyzing this CSV data. Here, because it was clarified that the cell voltage drop from the current interruption to 0.04msec in a transient response means a resistance polarization, the PPDS analyzed transient response that subtracted voltage drop in this section, and derived parameters of an activation and a diffusion polarizations [5]. This paper changed a new analytic algorithm.

The experimental conditions were prepared two humidified conditions and an influence of air utilization ra t io a s shown in Table 1 and 2, respectively. The standard condition was worked under the full humidifying that the temperature of the cell and the humidifier is set at 80℃. On the other hand, the non-humidifying condition was worked without humidifying and the cell temperature was set at 30℃. Here, the cell current density was 0.08A/cm^2. In the experiment that examines the influence of humidifying, the fuel utilization was 70%, and the air utilization was 40%, respectively. In the experiment that examines the influence of oxidant gas utilization, the fuel utilization was fixed at 70%, and the air utilization was varied from 40% to 90% every 10%. The operating pressure was atmosphere.

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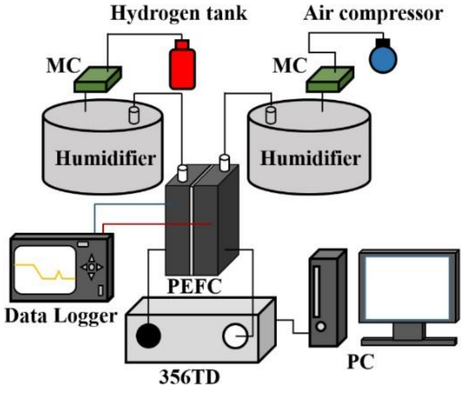
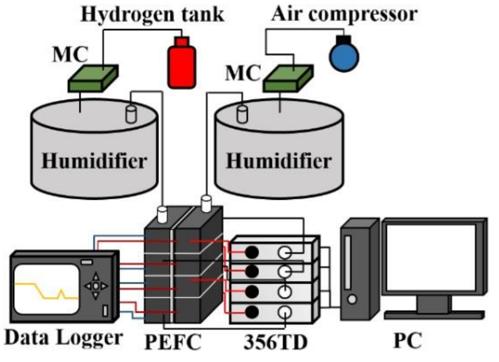


(a) for single cell



(b) for quadrisection separator

Figure 6. Schematic diagram of experimental apparatus

Table 1. Experimental conditions that examines the influence of humidifying

	Standard	Non-humidifying
Fuel Utilization [%]	70	
Air Utilization [%]	40	
Standard Current Density [A/cm^2]	0.08	
Cell Temperature [°C]	80	30
Humidifier Temperature (AN) [°C]	80	Not use
Humidifier Temperature (CA) [°C]	80	Not use

Table 2. Experimental conditions that examines the influence of oxidant gas utilization

Operating Condition	
Fuel Utilization [%]	70
Air Utilization [%]	40,50,60,70,80,90
Standard Current Density[A/cm^2]	0.08
Cell Temperature[°C]	80
Humidifier Temperature[°C]	80

Diagnostic principle

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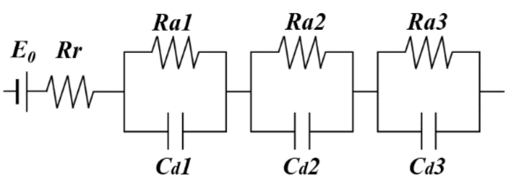
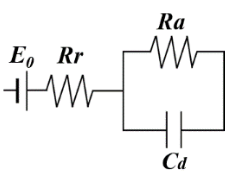
Generally, although the PEFC performance is diagnosed by an I-V curve and a Cole-Cole plot, each m ethod has both merits and demerits. The former diagnostics cannot identify the degradation factor though can dia gnose a deterioration of a cell performance easily. On the other hand, although the latter diagnostics can identify the degradation factor, its dia gnostic time is longer than the former one. The PPDS developed by us can diagnose the degradation factor of PEFC instantaneously by evaluating the transient response. At the first stage of this study, the transient response was expressed by the equivalent circuit composed of three RC circuits where anode reaction, cathode reaction and electrolyte resistance are imitated as shown in Figure 7 (a). At first, a n eva luating equation was derived from this equivalent circuit by Laplace transforming. In Eq. (1), each parameter sta nds f or the following: E_0, *Rr*, *Ra*, and *Cd* mean the open circuit voltage, the resistance of ions in an electrolyte, a diffusion resistance, an activation loss and an electric double layer capacity generated at the interface between the electrode and electrolyte, respectively. Generally, it is well known that the anode polarization is very small compa red with the cathode polarization, and the resistance polarization is also reflected in voltage drop to 0 – 1msec in the transient response. Therefore, in the second stage, we changed to the equivalent circuit shown only in cathode electrode as shown in Figure 7 (b), and the fitting equation was also changed to Eq. (2). Moreover, the resista nce polarization is derived by Eq. (3). As a result of deterioration diagnosis under various conditions by this m ethod,

the correlation between each parameter and each polarization was clarified as following: ΔV para meter, A_1

parameter and t_1 parameter mean the resistance polarization, the activation polarization and the diffusion polarization, respectively. The conventional diagnostics had been diagnosing the deteriorating situation of various cells using these parameters (ΔV , A_1 and t_1) as diagnostic parameters [6]. However, because the reflection of each deterioration factor in each parameter was small, it was necessary to diagnose the parameters strictly. Therefore, as there was a possibility of causing the diagnostic error, it should be improved. We have developed a new algorithm for PEFC performance diagnostics that solves the transient response by the third differential equation as shown in Eq. (4) directly to satisfy the demand mentioned above last year. The third differential Eq. (4) wa s discretize by the difference method and was converted into Eq. (5). The model Eq. (6) was obtained by simplifying Eq. (5). Each parameter (a, b, c) was obtained by fitting the transient response by Eq. (6) using on-line a lgorithm. Here, e_t on the right side of Eq. (6) means error in measurement. although the initial value of each parameter was replaced with the convergence value obtained from this online algorithm sequentially, an increase in the repetition processing lengthens the analytical time. the time variance of the parameter mentioned above should be checked to confirm the applicability to PEFC's transient response of the third differential equation. Figure 8 shows the transition of varying of each parameter by repetitive processing. As you can see, the result stabilized in the third, and the time variance of each parameter stabilizes with an increase in an analytical frequency. Therefore, we were able to confirm that the application for third differential equation to transient response is appropriate [7-10].



(a) equivalent circuit at the beginning of study



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equivalent circuit only on the cathode side**Figure 7.** Equivalent circuit of PEFC

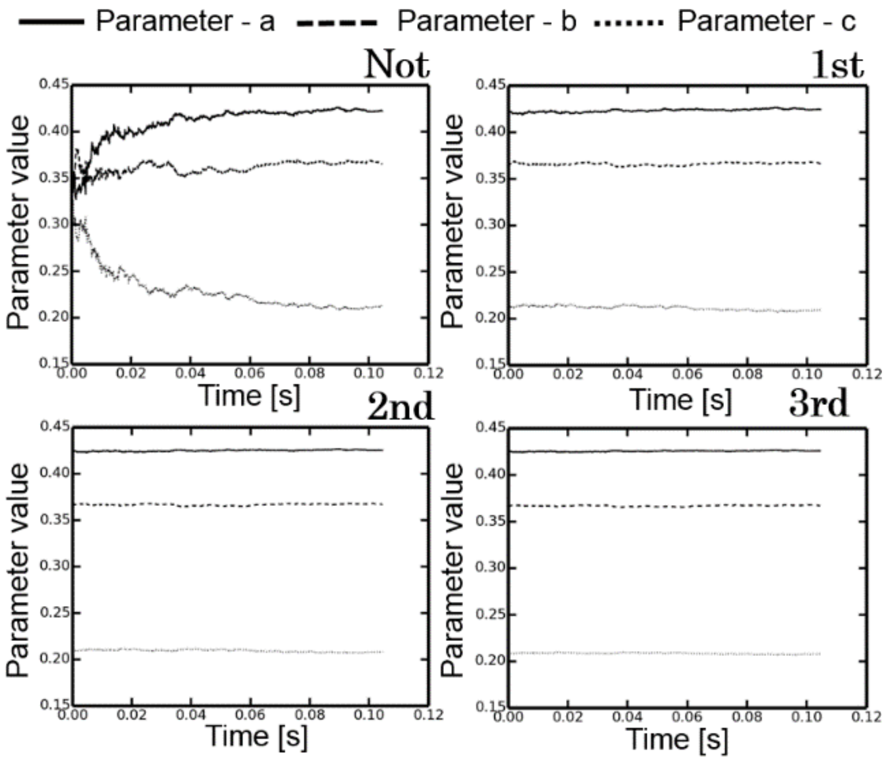
$$V_{out}(t) = y_0 + A_1 e^{-\frac{t}{t1}} + A_2 e^{-\frac{t}{t2}} + A_3 e^{-\frac{t}{t3}} \quad (1)$$

$$V_{out}(t) = y_0 + A_1 e^{-\frac{t}{t_1}} (2)$$

$$\Delta V = V_{out}(0) - V_{out}(100\mu sec) (3)$$

$y(t + 3 \cdot \Delta t) + ay(t + 2 \cdot \Delta t) V_{out}(t) d^3y(t) = \frac{}{dx^3} + {}^2y(t) \frac{}{dx^2}$ $b y(t) dx + cy(t)(4) + by(t + \Delta t) + cy(t) = e_t (5) y_{t+3} + ay_{t+2} + by_{t+1} + cy_t = e_t (6)$

Figure 8. Validation of application for the third differential equation to transient response



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RESULT AND DISCUSSION

Verifying the heat flux distribution by presence of humidifying

We aim to confirm the validity of the new diagnostics by comparing with the conventional diagnostics under condition of examining influence by presence of humidifying. In order to verify the correlation, firstly the heat flux distribution of each humidifying condition was measured by the separator as shown in Figure 4. Figure 9 shows the heat flux distribution in direction of gas flow. Here, the heat flow from the cell to the separator side was defined as a positive. From Figure 9 (a), the heat flux becomes negative by heating with the heater plate because the cell temperature is maintained by the heater plate at 80℃. However, the heat flux at the center pa rt of the cell (No.2) is smaller than other positions because the cell is heated by emitting the latent heat when the steam generated by the cell reaction condenses. Moreover, because the current density at the cell midrange is generally high, the cell temperature becomes higher than other parts in the cell by heating with the cell reaction. In previous study [(4)], It was confirmed that midrange has the highest current density, as shown in Fig 10. It shows heat distribution in the same electrode. Consequently, the heat flux at the center part of the cell becom es smaller. Therefore, we think that a weak flooding phenomenon is caused from the center part of the cell to gas outlet under the standard operating condition, and the cell reaction at the center part is more active than other position. From Figure 9 (b), the change of the heat flux originates in only the cell reaction because the cell is not heated by the heater plate under the non-humidifying condition. Because the steam generated by the reaction is added to the supplied gas with the heat of reaction, the heat flux is accumulated from the center pa rt of the cell to gas outlet. Based on these findings, we verified two degradation factor distributions obtained by the conventional diagnostics and the new diagnostics.

Confirmation of correlation between former diagnostics parameter and new diagnostics parameter

Figure 11 shows polarization distribution obtained using the conventional diagnostics by presence of humidifying. In both conditions, A_1 (the activation polarization) and t_1 parameter (the diffusion polarization)grow toward the gas outlet (No.4) from the gas inlet (No.1) in all current density region though ΔV parameter(the resistance polarization) decreases. This reason is that the flooding/plugging phenomena is caused byflowing as steam generated by cell reaction condensing to the gas downstream, and consequently A_1 and t_1 parameter of the downstream increase. Oppositely, because this excess water wets the membrane, ΔVparameter decreases toward the gas outlet from the gas inlet. Especially, this tendency is remarkable in thecondition without heating and humidifying. However, the difference of the parameter under the same conditionby each place is small though the difference of the parameter by presence of humidifying is great. On the other hand, the diagnostics result by the new diagnostics is shown in Fig.12. The change of parameter-c on the current density is the largest, and because the increase of parameter-c means the improvement of the cellperformance as shown in Eq. (6), the behavior of parameter-c becomes opposite of A_1 in the previousdiagnostics. Therefore, parameter-c may be corresponding to A_1 (activation polarization). On the other hand, the change of parameter-b on the current density is the smallest, and the behavior of parameter-b and ΔV parameter under non-humidifying condition resembles closely. Therefore, parameter-b may be corresponding to ΔV (resistance polarization). Finally, the change of parameter-a decreases with the increase of current density, and especially it decreases drastically under non-humidifying condition. If the parameter-a decreases, it means there are few voltage variations to an electric current density increase. Moreover, if the parameter-a became minus, it means the cell voltage decreases. Therefore, because this behavior and the flooding/plugging phenomena are similar, we are considering that parameter-a is corresponding t1 (diffusion polarization) at present.

From these results, because the correlation between each parameter by the new algorithm and each polarization cannot be identified enough, it should be verified furthermore in the condition that each polarization is greatly reflected. However,

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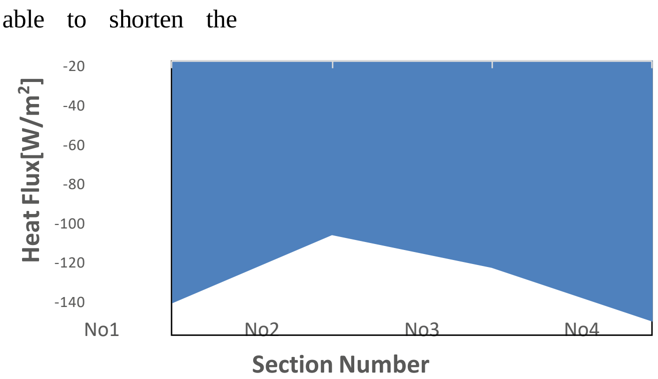
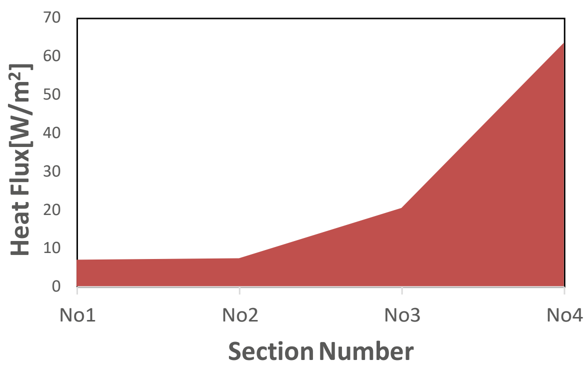
because the new diagnostics can greatly reflect the difference of the humidifying conditions in each parameter as well as the conventional diagnostics and it was able to shorten the diagnosing time, we are considering that its validity was verified.

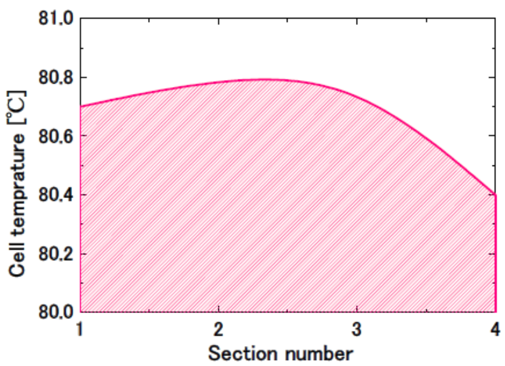


(a)) Standard operating condition



(b) Non-humidifying condition

Figure 9. Heat flux distribution in direction of gas flow



(a)) Standard operating condition

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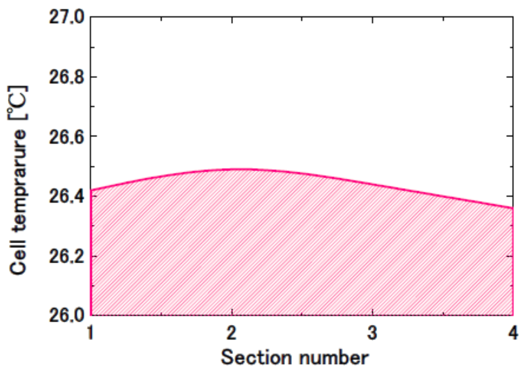

(b) Non-humidifying operating condition

Figure 10. Heat distribution in the same Electrode

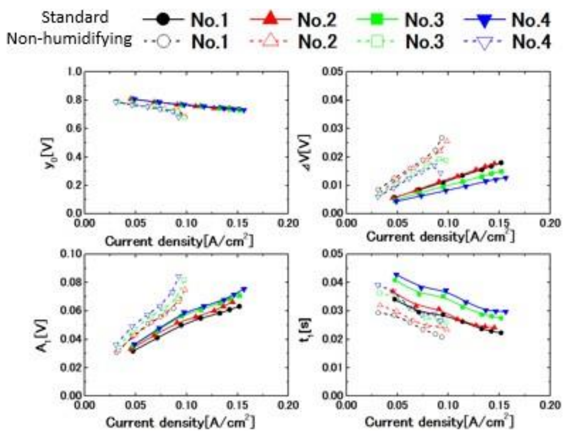

Figure 11. Polarization distribution obtained by the conventional diagnostics

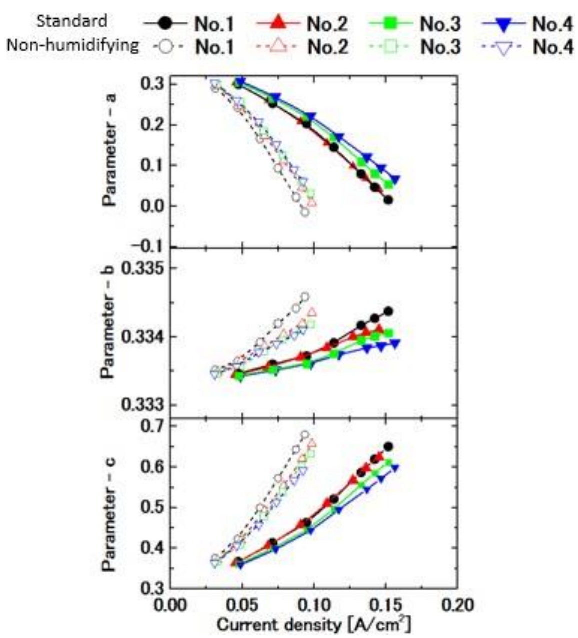

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Figure 12. Diagnostics result by the new diagnostics

CONCLUSIONS

The results obtained in this study are summarized as follows.

- By measuring heat flux inside the PEFC, we verified the degradation factor for PEFC.
- We verified the degradation factors in each part by comparing with the conventional data.
- The new algorithm was able to greatly reflect the difference of the humidifying conditions in each parameter as well as the conventional algorithm.

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