



Preferred test conditions for measuring flow rate distribution between cells in a polymer electrolyte fuel cell stack

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ARTICLE INFO

Article history:

Received 27 September 2010

Received in revised form

20 December 2010

Accepted 20 February 2011

Available online 1 March 2011

Keywords:

PEFC

Stack

Flow rate distribution

Hydrogen limiting current

ABSTRACT

A new and practical testing technique was developed for measuring the flow rate distribution between cells in a stack that did not contain any internal sensors. The flow rate distribution is obtained by measuring the hydrogen limiting current of each cell in the stack while a mixed gas of hydrogen and dimethyl ether is supplied to the anode and hydrogen to the cathode. In order to measure large flow rate deviations between cells, it is necessary to decrease the flow rate of the anode hydrogen and to sufficiently humidify the cells. The faster the increasing rate of the current, the more the apparent hydrogen limiting current increases than the theoretical electrochemical equivalent current. However, the relative flow rate deviations between cells can be obtained by a practical accuracy using the ratio of the apparent hydrogen limiting current. Humidification of the cell is indispensable for the measurement and a method using dry anode gas and humidified cathode gas is recommended. The preferred test conditions for measuring the flow rate distribution between cells in a PEFC stack are proposed.

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1. Introduction

Fuel cells are a promising option for replacing state-of-the-art energy conversion and storage technologies such as internal combustion engines and batteries due to their high conversion efficiency and their low or even zero-emission operation. Especially, the polymer electrolyte fuel cell (PEFC) is now being paid much attention and is expected to be in practical use as a portable power source for cogeneration, electric vehicles, mobile phones, etc. To make this system practicable, the fuel cell is used in the form of a stack of sufficient single cells to provide the intended voltage. In the stack, the gas needs to be supplied evenly to each cell. However, it is extremely difficult to measure the flow rate of each cell of the PEFC stack. An example of measuring the flow distribution between cells has not been reported though there was an example [1] of having measured the pressure distribution between cells in a stack.

We reported a method of nondestructively measuring the flow rate distribution between cells in a PEFC stack for the first time [2]. This technology is a method of supplying the mixed gas of the

dimethyl ether (DME molecular formula: CH_3OCH_3) and hydrogen to the anode and hydrogen to the cathode as shown in Fig. 1, and measuring the hydrogen limiting current of each cell, then calculating the flow rate of each cell from these values. According to the PEFC performance using mixed fuel gas of H_2 and DME in reference [3], the performance was the same as that using H_2 fuel within the range of the current that hydrogen was not completely consumed and remained in the cell. The performance was not changed by mixing DME to H_2 under full humidifying conditions. Because the performance of a hydrogen fuel cell does not change as long as hydrogen exists in the cell even if DME nine times hydrogen is added to hydrogen [3], DME is not a poisoning gas. It is thought to be possible to use a gaseous methanol instead of DME as the second fuel used for this measurement. However, the methanol is a liquid at the normal temperature, it is easy to dissolve in water and the proton exchange membrane, and might cause the cell size transformation. Therefore, not the methanol but DME was selected as the second fuel. It is considered that there is no material that solubility to water is lower than that of DME and is high performance fuel for the direct fuel cell. The principle of this measuring method is described using Fig. 2. The current in the stack is gradually increased by an external power source. For a cell supplied with only hydrogen to the anode and hydrogen to the cathode, the cell voltage is shown as H_2/H_2 in Fig. 2. In a case of a cell supplied only DME to the anode and hydrogen to the cathode, the cell voltage is shown as DME/H_2 . In a case of a cell supplied

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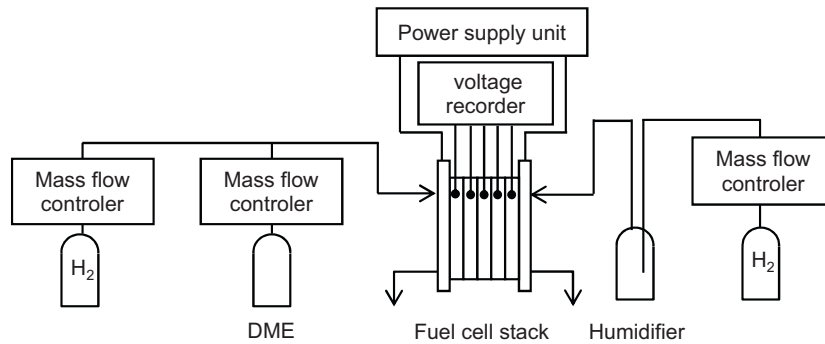


Fig. 1. Measuring system.

DME with a small amount of hydrogen, the cell voltage changes as $(\text{H}_2 + \text{DME})/\text{H}_2$ (1) ($a \rightarrow b \rightarrow c \rightarrow d$). Also, in the case when a cell is supplied DME with a large amount of hydrogen, the cell voltage changes as $(\text{H}_2 + \text{DME})/\text{H}_2$ (2) ($a' \rightarrow b' \rightarrow c' \rightarrow d'$). The cell voltage is low if there is hydrogen in the anode (between $a-b$ and $a'-b'$). The cell voltage rapidly increases if hydrogen in the cell is completely consumed (between $b-c$ and $b'-c'$). However, the DME starts reacting at a high cell voltage, and the increasing rate of the cell voltage is suppressed (between $c-d$ and $c'-d'$). The flow rate of each cell can be calculated from the current (b or b') in which the cell voltage begins to quickly increase. The electrode degradation does not occur at 0.9 V or less versus the hydrogen reference potential. If the hydrogen limiting current of all cells in a stack can be measured without exceeding 0.9 V for every cell voltage, the flow rate of all cells in the stack can then be nondestructively measured.

It becomes possible to determine the cause of flow rate deviation between cells of the PEFC stack when this method is put into practical use, and it is expected to be useful for the development of a stack with few flow rate deviations between the cells. In addition to research, this method is applicable for nondestructive product inspection because there is no need of internally embedding a sensor in the product, which contributes to the reliability improvement of the product.

The measuring principle and the measured example of the flow rate distribution between cells of a stack using the hydrogen limiting current are described in a previous report [2]. This report describes the preferred test conditions to expand the measuring range and to maintain the measuring accuracy. It is necessary to specify the preferred testing conditions so that many people may then easily use this test method.

In Section 2 of this report, the relation between the hydrogen flow rate and the nondestructive, measurable range is analyzed. In Section 3, the effect of the increasing current rate on the measured hydrogen limiting current is examined. In Section 4, humidification of the stack is examined and how to humidify the gas used for the measurement is recommended. In Section 5, the DME flow rate

and the cathode hydrogen flow rate are recommended. The testing method and recommended test conditions for measuring the flow rate distribution between cells in a stack are summarized in Appendix A.

2. Relation between hydrogen flow rate and nondestructive, measurable range

The nondestructive, measurable range is derived from careful consideration of the result in Fig. 3. Fig. 3 is the result from a former report [2] in which the cell voltage of a 4-cell stack is measured by gradually increasing the current while the mixed gas of 4 ml min^{-1} hydrogen and 200 ml min^{-1} DME is supplied to the anode, and 200 ml min^{-1} humidified hydrogen to the cathode at room temperature under ambient pressure. First the cell voltage of No. 1 rapidly increases. After the cell voltage reaches 0.7 V, the rapid increase changes to a slow increase. The measurement was stopped when the cell voltage of any cell reached 0.9 V. This stop is necessary because there is a possibility of electrode degradation when voltage reaches over 0.9 V. The current density when the cell voltage begins to rapidly increase is the hydrogen limiting current density, and the flow rate ratio of each cell can be obtained according to the ratio of the hydrogen limiting current density of each cell. From Fig. 3, the flow rate of every cell can be obtained because the hydrogen limiting current of every cell is measured before the cell voltage of the minimum flow rate cell No. 1 reached 0.9 V. If the current density difference from 0.7 V to 0.9 V is high, the nondestructive measurement is possible for the stack having a high flow rate deviation. However, if the difference is small, the nondestructive measurement is possible only for the stack having a small flow rate deviation.

If the limiting current of the cell having a minimum or maximum flow rate is defined as I_{\min} or I_{\max} , respectively, and the current density from 0.7 V to 0.9 V is ΔI , the condition that the flow rate deviation can be nondestructively measured is given by the

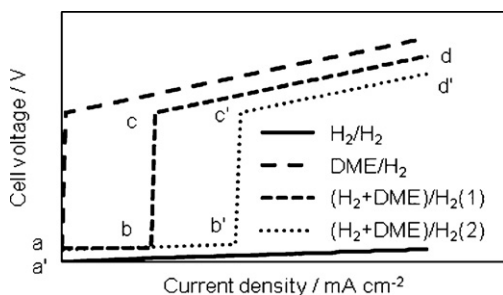


Fig. 2. Cell performance using $(\text{H}_2 + \text{DME})$ mixed gas for the anode and H_2 for the cathode.

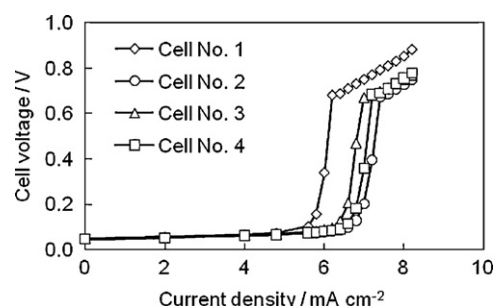


Fig. 3. A measurement result of hydrogen limiting current of a 4-cell stack [2].

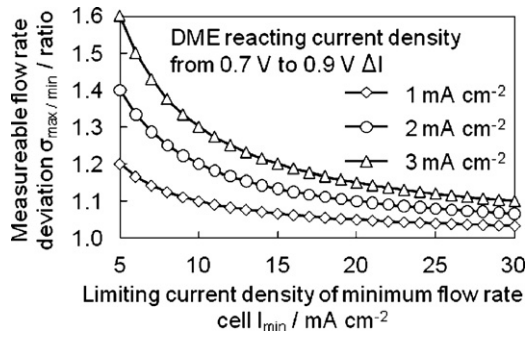


Fig. 4. Measureable flow rate deviation between cells in a PEFC stack.

following equation:

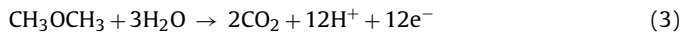
$$I_{\max} < I_{\min} + \Delta I \quad (1)$$

If the ratio of the maximum to minimum flow rate of the cell in the stack is defined as $\sigma_{\max/\min}$, the condition for the nondestructive measurement is shown by the following equation:

$$\sigma_{\max/\min} < \frac{I_{\min} + \Delta I}{I_{\min}} \quad (2)$$

Fig. 4 shows the relation between I_{\min} and $\sigma_{\max/\min}$ using Eq. (2) where ΔI is assumed to be 1, 2 and 3 mA cm⁻². The smaller the I_{\min} , the larger the ΔI , and the larger the $\sigma_{\max/\min}$. If the hydrogen flow rate used for the measurement is decreased, I_{\min} proportionally becomes small. Fig. 4 shows the result that the smaller the hydrogen flow rate, the wider the nondestructive, measurable range. However, another problem influencing the measurement precision occurs if the hydrogen flow rate is significantly decreased. The preferred hydrogen flow rate is discussed in Section 3.

In order to increase ΔI , the DME should be made to easily react with moisture based on Eq. (3).



The significant moisture effect on the direct DME fuel cell performance has already been reported [4]. Humidification of the stack is indispensable for this measurement. The preferred humidifying method is recommended in Section 4.

3. Influence of increasing current rate

The purpose of this method is to obtain the flow rate of each cell by measuring the hydrogen limiting current of each cell. It was demonstrated in a former report [2] that the measured hydrogen limiting current was almost equal to the theoretical electrochemical equivalent current calculated from the hydrogen flow rate. However, the hydrogen flow rate at that time was high enough, and allowed more time for measuring in the stationary state. In the former chapter, a low hydrogen flow rate is expected for the wide nondestructive measuring range. If the hydrogen flow rate becomes low, it takes a long time for the cell voltage to reach equilibrium. Therefore, there is the possibility that the hydrogen limiting current density is excessively measured. This chapter investigates this phenomenon, and recommends the preferred hydrogen flow rate and the increasing current rate.

3.1. Experimental

The stack used in this study is composed of four cells. Each cell has a 1 mm × 1 mm single serpentine channel and the electrode area is 25 cm². The gas is supplied to each cell in parallel through a manifold of \varnothing 3 mm. Nafion@NR-212 was used as the electrolyte membrane. Carbon-supported platinum (Pt/C) was used as

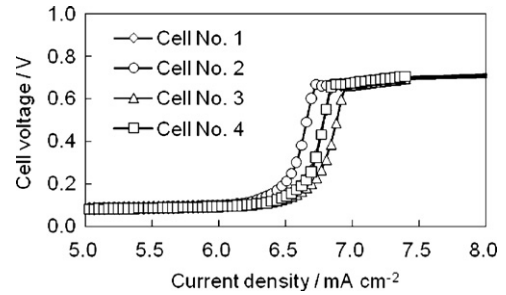


Fig. 5. Hydrogen limiting current characteristics of a 4-cell stack using a mixed gas of 4 ml min⁻¹ H₂ and 200 ml min⁻¹ DME for the anode and 200 ml min⁻¹ humidified H₂ for the cathode by increasing the current density rate to 0.1 mA cm⁻² min⁻¹.

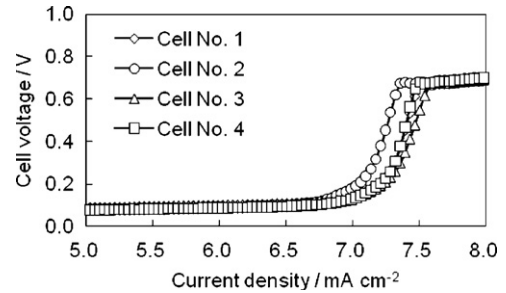


Fig. 6. Hydrogen limiting current characteristics of a 4-cell stack using a mixed gas of 4 ml min⁻¹ H₂ and 200 ml min⁻¹ DME for the anode and 200 ml min⁻¹ humidified H₂ for the cathode by increasing the current density rate to 0.2 mA cm⁻² min⁻¹.

the electrode catalyst, and the Pt loading of both the anode and the cathode was 0.5 mg cm⁻².

When the stack used in the former report [2] was dismantled, it was observed that the thickness of the gas diffusion layers was from 180 μ m to 220 μ m. It was theorized that the difference in the thickness was a cause of the large flow rate deviation in the stack. In this report, a new stack was reassembled using gas diffusion layers whose difference in thickness was \pm 5 μ m.

This stack was used in the measurement system as shown in Fig. 1. The mixed gas of 4 ml min⁻¹ hydrogen and 200 ml min⁻¹ DME was supplied to the anode and the 200 ml min⁻¹ hydrogen to the cathode at room temperature under ambient pressure. During the measurement of the hydrogen limiting current density, the current was increased in steps by 1 mA, every 3, 6, 12, or 24 s. The increasing current density rate corresponds to 0.8, 0.4, 0.2, or 0.1 mA cm⁻² min⁻¹, respectively.

3.2. Results and discussion

Figs. 5–8 show the measured hydrogen limiting current characteristics of the 4-cell stack. Though the hydrogen flow rate was

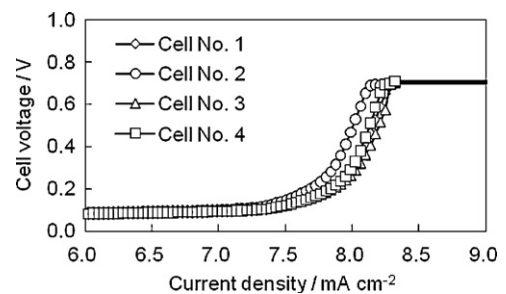


Fig. 7. Hydrogen limiting current characteristics of a 4-cell stack using a mixed gas of 4 ml min⁻¹ H₂ and 200 ml min⁻¹ DME for the anode and 200 ml min⁻¹ humidified H₂ for the cathode by increasing the current density rate to 0.4 mA cm⁻² min⁻¹.

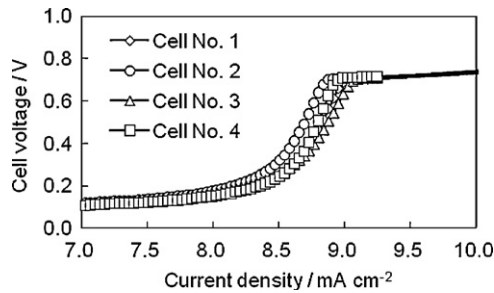


Fig. 8. Hydrogen limiting current characteristics of a 4-cell stack using a mixed gas of $4 \text{ ml min}^{-1} \text{ H}_2$ and $200 \text{ ml min}^{-1} \text{ DME}$ for the anode and 200 ml min^{-1} humidified H_2 for the cathode by increasing the current density rate to $0.8 \text{ mA cm}^{-2} \text{ min}^{-1}$.

the same, the apparent hydrogen limiting current density increased with the increasing current rates. The theoretical hydrogen limiting current density corresponding to the supplied hydrogen flow rate was 5.74 mA cm^{-2} . It is clear that the difference from the theoretical hydrogen limiting current density increased with the increasing current rate. The cell voltage does not increase until the adsorbed hydrogen on the electrode catalyst is consumed. Therefore, when the current is low, and the increasing current rate is fast, it takes time to consume the adsorbed hydrogen, and the difference between the measured apparent hydrogen limiting current and theoretical electrochemical equivalent current increases. The quantity of the electric charge corresponding to the adsorbed hydrogen $C(t)$ in C cm^{-2} at time t is shown by the following equation where K is the increasing current density rate in $\text{A cm}^{-2} \text{ s}^{-1}$, and t is time in seconds after reaching the hydrogen limiting current.

$$t < 0 \quad C(t) = C(0) \text{ constant}$$

$$t > 0 \quad \frac{dC(t)}{dt} = -Kt \quad (4)$$

$$C(t) = \frac{C(0) - Kt^2}{2} \quad (5)$$

Time t_d when $C(t)$ becomes 0 is described by the following equation:

$$t_d = \left(\frac{2C(0)}{K} \right)^{0.5} \quad (6)$$

The difference in the hydrogen limiting current density at time t_d between the measurement and theory is defined as I_d and described by the following equation:

$$I_d = (2KC(0))^{0.5} \quad (7)$$

The difference I_d is proportional to the square root of increasing current density rate K and the square root of the amount of adsorbed hydrogen $C(0)$ on the electrode catalyst. The method that predicts the value of the hydrogen limiting current from the characteristics of the measured hydrogen limiting current has not yet been established. It is difficult to determine the current when the cell voltage begins to increase because of the low slope of the voltage/current. In this report, we then used the current density at 0.4 V at the midpoint of the area when the cell voltage rapidly changed (from 0.1 V to 0.7 V) as the apparent hydrogen limiting current density and expressed as $I_{0.4}$. Fig. 9 shows the relation between the square root of the increasing current density rate K and $I_{0.4}$ obtained from the results of Figs. 5–8, and this relation is almost linear. When extrapolating the line until K is equal to 0 in Fig. 9, the apparent hydrogen limiting current density approaches 5.74 mA cm^{-2} as the theoretical hydrogen limiting current density. The apparent hydrogen limiting current density $I_{0.4}$ is considered to be the value in which I_d expressed by Eq. (7) is added to the theoretical hydrogen limiting current density.

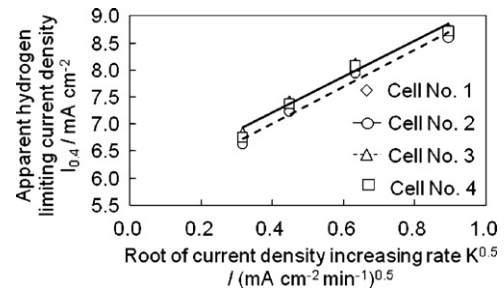


Fig. 9. Effect of the increasing current rate on the apparent hydrogen limiting current density.

Fig. 10 shows the flow rate deviations between cells in the stack by changing the increasing current rates. The flow rate deviations are assigned as the ratio of $I_{0.4}$ of each cell to the average of $I_{0.4}$ of all cells. It is understood from Fig. 10 that the flow rate deviation between cells in the stack used in this measurement is about $\pm 1.5\%$, the flow rate of cell No. 2 is the lowest, and the flow rate of cell No. 3 is the highest. While the flow rate deviation between cells in the stack in a previous report [2] is about 10% , the flow rate deviation of the stack used in this report is significantly improved. Fig. 10 demonstrates that this method can be used to measure small deviations of about 1% .

When the increasing current density rate changed from $0.1 \text{ mA cm}^{-2} \text{ min}^{-1}$ to $0.8 \text{ mA cm}^{-2} \text{ min}^{-1}$ in Fig. 10, the flow rate deviations decreased from 1 ± 0.017 to 1 ± 0.013 , and the absolute value of $I_{0.4}$ changed from 6.7 mA cm^{-2} to 8.7 mA cm^{-2} . However, it is understood that the influence of increasing current density rate is remarkably suppressed if the ratio of $I_{0.4}$ of each cell to the average of $I_{0.4}$ of all cells is used for the flow rate deviation between cells. There is a slight trend in Fig. 10 that the flow rate deviation increased due to a small increasing current density rate. When the increasing current density rate is $0.4 \text{ mA cm}^{-2} \text{ min}^{-1}$ or $0.1 \text{ mA cm}^{-2} \text{ min}^{-1}$, the flow rate deviation is 1 ± 0.013 or 1 ± 0.015 , respectively. It is presumed that the flow rate deviation becomes 1 ± 0.017 as the increasing current density rate approaches 0 by extrapolating the lines in Fig. 9. The flow rate deviation is underestimated only by $\pm 0.4\%$ when measuring it using the increasing current rate of $0.4 \text{ mA cm}^{-2} \text{ min}^{-1}$. It is important to be able to clearly identify the relative flow rate between cells. The underestimated absolute value of $\pm 0.4\%$ is not very important and is considered to be within the acceptable level for this measurement.

It takes 57.4 min for the measurement at 5.74 mA cm^{-2} for the hydrogen limiting current density and a $0.1 \text{ mA cm}^{-2} \text{ min}^{-1}$ increasing current density rate. Actually, it took about 1 h for the measurement of Fig. 5. The measurement time can be shortened to 15 min if the increasing current density rate is increased to $0.4 \text{ mA cm}^{-2} \text{ min}^{-1}$. The measurement does not need to start from a current density of 0. The measurement can also be shortened by

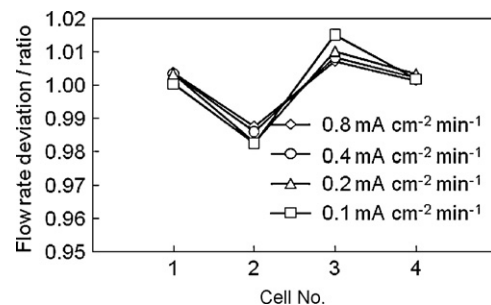


Fig. 10. Effect of the increasing current rate on the flow rate distribution between cells in a 4-cell stack.

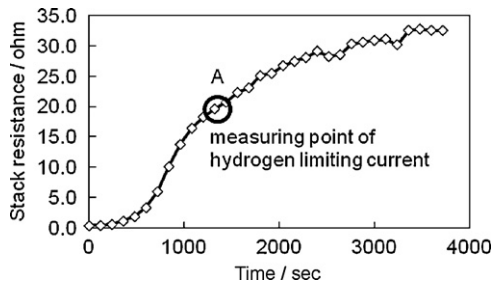


Fig. 11. Resistance change of a 4-cell stack while N₂ dry gas was flowing.

starting it from a slightly lower current density than the hydrogen limiting current. The experiment in this study used electrodes on which Pt was loaded at 0.5 mg cm⁻². Hydrogen is adsorbed on the loaded Pt. Therefore, when the Pt loading amount changes, it is necessary to consider the change in the effect of the increasing current rate on the test results.

4. Influence of gas humidifying

DME is used as the second fuel in this measurement to suppress the voltage increase. Water is necessary for the reaction of DME as described by expression (3) in Section 2. This chapter examines the influence of moisture on the reaction of DME, and recommends the preferred humidifying method.

4.1. Experimental

An anode gas humidifier was added to the measuring system in Fig. 1. The stack described in Section 3.1 was used for the measurement at room temperature under ambient pressure. The anode gas flow rate of DME was 200 ml min⁻¹ with 4 ml min⁻¹ hydrogen and the cathode gas flow rate was 200 ml min⁻¹ hydrogen. Comparative tests were done under the four gas humidifying conditions of dry/dry, dry/wet, wet/dry and wet/wet for the anode/cathode. The increasing current rate was 0.2 mA cm⁻² min⁻¹.

4.2. Results and discussion

4.2.1. Anode: dry gas/cathode: dry gas

Fig. 11 shows the resistance change of a 4-cell stack while supplying 200 ml min⁻¹ of dry N₂ gas to each electrode. The resistance of the stack began to increase at hundreds of seconds. At A point in Fig. 11, the hydrogen limiting current characteristic was measured while supplying dry hydrogen at 4 ml min⁻¹ and dry DME at 200 ml min⁻¹ to the anode and dry hydrogen at 200 ml min⁻¹ to the cathode. Fig. 12 shows the results. Fig. 12 shows a very different characteristic from the previous one in that the cell voltage rapidly increases at about 3 mA cm⁻² though the theoretical hydrogen limiting current is 5.74 mA cm⁻². It is presumed that the

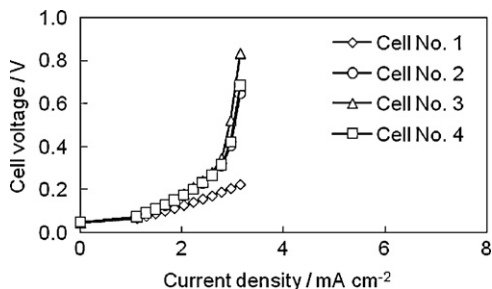


Fig. 12. Hydrogen limiting current characteristics for a 4-cell stack using a mixed gas of H₂ (4 ml min⁻¹) and DME (200 ml min⁻¹) at A point (anode: dry, cathode: dry).

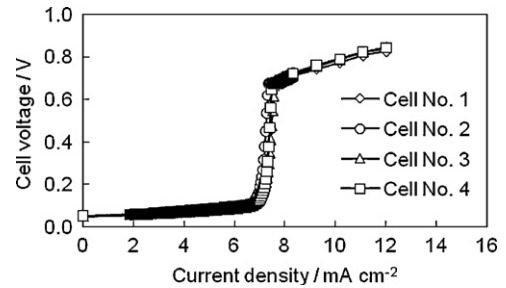


Fig. 13. Hydrogen limiting current characteristics for a 4-cell stack using a mixed gas of H₂ (4 ml min⁻¹) and DME (200 ml min⁻¹) (anode: dry, cathode: wet).

resistance polarization becomes about 0.4 V at 3 mA cm⁻² because the resistance of the stack is 20 ohm. We cannot obtain the flow rate deviation of the stack from Fig. 12, because the hydrogen limiting current was not measured. These results show that moisture is indispensable for the measurement.

4.2.2. Anode: dry gas/cathode: wet gas

In the former chapter, the hydrogen limiting current was successively measured while supplying a dry mixed gas to the anode and wet gas from a humidifier to the cathode. Fig. 13 shows that the DME reacting current density from 0.7 V to 0.9 V is presumed to be about 5 mA cm⁻² under this humidifying condition. Because the observed apparent hydrogen limiting current is 7 mA cm⁻², the measurable flow rate deviation ratio reaches (7 + 5)/7 = 1.71 as explained in Section 2 suggesting that the humidifying is sufficient.

4.2.3. Anode: wet gas/cathode: dry gas

Fig. 14 shows the result of the hydrogen limiting current characteristic for the cathode dry gas and the anode wet gas supplied through a humidifier at room temperature. The observed apparent hydrogen limiting current density *I*_{0,4} and DME reacting current density in Fig. 14 are 6.7 mA cm⁻² and 8 mA cm⁻², while those in Fig. 13 are 7.3 mA cm⁻² and 5 mA cm⁻². The difference of the absolute value of *I*_{0,4} between 6.7 mA cm⁻² and 7.3 mA cm⁻² little influences the flow rate deviations, because the ratios of *I*_{0,4} of each cell to the average of *I*_{0,4} of all cells give the flow rate deviations but the absolute values of *I*_{0,4} influence the flow rate deviations only a little as explained in Section 3.2. DME reacting current density of 8 mA cm⁻² in Fig. 14 gives wide measurable range of flow rate deviation. This suggests that the humidifying is sufficient. However, it took 2 h to obtain a stable anode gas flow rate due to the fact that a large amount of DME was soluble to the water stored in the humidifier. This humidifying method cannot be recommended because it takes a lot of time for preparation of these tests.

4.2.4. Anode: humidified gas/cathode: humidified gas

Fig. 15 shows the result of the hydrogen limiting current characteristic by the cathode wet gas and the anode wet gas supplied

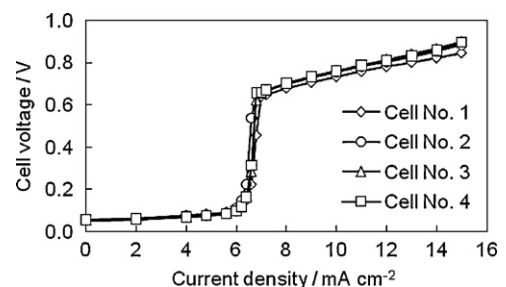


Fig. 14. Hydrogen limiting current characteristics for a 4-cell stack using a mixed gas of H₂ (4 ml min⁻¹) and DME (200 ml min⁻¹) (anode: wet, cathode: dry).

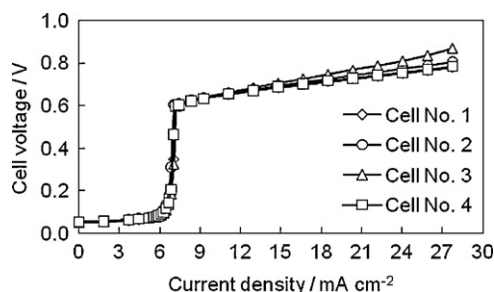


Fig. 15. Hydrogen limiting current characteristics for a 4-cell stack using a mixed gas of H₂ (4 ml min⁻¹) and DME (200 ml min⁻¹) (anode: wet, cathode: wet).

through each humidifier at room temperature. The usual hydrogen limiting current characteristic was observed. The DME reacting current density from 0.7 V to 0.9 V was 21 mA cm⁻² while the observed hydrogen limiting current was 7 mA cm⁻². The measurable flow rate deviation ratio was estimated to be (7 + 21)/7 = 4 that is a high value not usually needed. This humidifying method cannot be recommended because it also takes a lot of time for preparation of these tests.

Fig. 16 shows the flow rate deviations between the cells of the stack obtained from the measured results of Figs. 13–15. The same flow rate deviations between the cells occurred if anode or cathode or both are humidified. The method using the anode dry gas and cathode wet gas is recommended because the measurement is easy.

5. Other measuring conditions

5.1. Dimethyl ether flow rate

The DME is the main gas that flows to the anode in this measurement. The hydrogen flow rate when mixed with the DME is low. In general, the flow rate deviation between cells increases with the flow rate. This is caused by the divergence and the confluence between the manifold and each cell in the stack. The density and the viscosity of the gases used during the flow rate measurement are different from that during real operating conditions of the stack. Therefore, it is desirable to set the DME flow rate using the following equation so that the Reynolds number of both may become the same.

$$Q_{\text{DME}} = Q_{\text{mix}} \cdot \frac{\eta_{\text{DME}}}{\eta_{\text{mix}}} \cdot \frac{\rho_{\text{mix}}}{\rho_{\text{DME}}} \quad (8)$$

where Q_{mix} , η_{mix} , and ρ_{mix} are the flow rate, the viscosity, and the density of the mixed gas under real operating conditions, and Q_{DME} , η_{DME} , and ρ_{DME} are the flow rate, the viscosity, and the density of the dimethyl ether at the test temperature.

For example, the real operating condition of the anode using the reformed gas assumes a current density of 0.3 A cm⁻², utilization of 70%, and full humidifying at 80 °C.

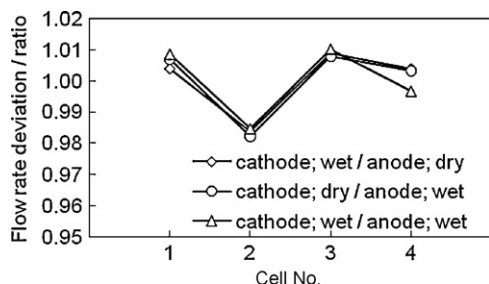


Fig. 16. Flow rate between cells calculated from the measured results of Figs. 13–15.

The viscosities of H₂, H₂O, and CO₂ at 1 atm are each 8.8 × 10⁻⁶ Pa s (20 °C), 1.21 × 10⁻⁵ Pa s (100 °C), and 1.81 × 10⁻⁵ Pa s (100 °C) [5], and the viscosities are 9.99 × 10⁻⁶ Pa s, 1.14 × 10⁻⁵ Pa s, and 1.72 × 10⁻⁵ Pa s when converted at 80 °C using a temperature compensation formula, respectively [6].

The flow rates of H₂, CO₂, and H₂O for the anodes having a 100 cm² total area are 386 ml min⁻¹, 101 ml min⁻¹, and 423 ml min⁻¹, respectively, and the total mixed gas flow rate Q_{mix} is 905 ml min⁻¹. The viscosity of the mixed gas is 1.32 × 10⁻⁵ Pa s that is calculated by the Bromley & Wilke expression [7]. The density ρ_{mix} of the mixed gas is 0.482 kg m⁻³.

On the other hand, the viscosity η_{DME} and density ρ_{DME} of DME at 1 atm and 15 °C are 9.06 × 10⁻⁶ Pa s and 1.95 kg m⁻³, respectively. If these values are substituted in expression (8), the flow rate Q_{DME} of the DME is 154 ml min⁻¹.

In this report, the stack with a total anode area of 100 cm² was used for the measurement while supplying DME at 200 ml min⁻¹ to the anode. This flow rate is near the flow rate of 154 ml min⁻¹ at the assumed real operating condition from the viewpoint of fluid mechanics.

As another example, the real operating conditions of the cathode using air, a current density 0.3 A cm⁻², air utilization of 50%, and full humidifying at 80 °C, are assumed. The DME flow rate for the measurement can be calculated in a way similar to that mentioned above. As a result, the DME flow rate Q_{DME} becomes 564 ml min⁻¹ for the total electrode area of 100 cm². The DME flow rate of the about 600 ml min⁻¹ is necessary if one is interested in the air flow rate deviation between cells.

5.2. Cathode hydrogen flow rate

Though a low cathode hydrogen flow rate is sufficient for maintaining the cathode to hydrogen reference potential, a high cathode hydrogen flow rate is necessary for humidifying the cell when the anode dry gas and cathode wet gas are used.

Based on the result in Section 4, when 400 ml min⁻¹ of dry DME is supplied to the anode, a 400 ml min⁻¹ cathode hydrogen flow rate is shown to be sufficient to supply the necessary moisture for the DME reaction. The relative humidity of 50% is considered to be sufficient for the total gas supplied to the stack.

6. Conclusions

In the former report [2], we showed the principle and measured examples of a new method for nondestructively measuring the flow rate distribution between cells in a PEFC stack without including a sensor for the first time.

The preferred testing conditions for this measurement were examined in this report, and the obtained results are listed below.

- (1) It is necessary to lower the anode hydrogen flow rate and to sufficiently humidify the cell in order to measure the higher deviation of the flow rate between the cells of a stack.
- (2) When the increasing current rate is high, the difference between the theoretical current corresponding to the electrochemical equivalent and the measured hydrogen limiting current increases. However, if the flow rate deviation between cells is derived using the ratio of the measured apparent hydrogen limiting current of each cell, the effect of the increasing current rate on the flow rate deviation can be reduced.
- (3) When supplying dry gas to both electrodes, the measurement of the flow rate deviation becomes impossible because the cell resistance significantly increases. The method of supplying dry gas to the anode and wet gas to the cathode is recommended.

- (4) It was clearly demonstrated that this method is able to detect about a 1% flow rate deviation between cells in a PEFC stack.

It is necessary to determine the testing condition used for this measurement according to the specification and the testing purpose of the users. However, a typical testing condition is described in Appendix A because many people should be able to easily use this method.

Appendix A.

The recommended method and conditions are as follows.

- (1) Temperature and pressure
Room temperature and atmospheric pressure.
- (2) Gas flow rate
For an electrode with a total area of 100 cm², the anode hydrogen flow rate is 4 ml min⁻¹, the anode DME flow rate is 200 ml min⁻¹, and the cathode hydrogen flow rate is 200 ml min⁻¹. If the target is the cathode flow rate distribution, the anode DME flow rate and the cathode hydrogen flow rate are both 600 ml min⁻¹.
- (3) Gas humidity
The anode gas is dry, and the cathode gas is fully humidified at room temperature. There is not a necessity to worry about the influence of the liquid water because the liquid water is not generated in the anode in this condition.
- (4) Protection of stack deterioration
The current should begin to flow after hydrogen is supplied to both electrodes. The cell voltage of all cells should be monitored, and when the cell voltage of any cell reaches

0.9 V, the measurement should be interrupted by cutting off the current.

- (5) Current density increasing rate
The cell voltage of all the cells is measured while increasing the current density from 5 mA cm⁻² at the rate of 0.4 mA cm⁻² min⁻¹.
- (6) Ending of measurement
The measurement ends when the cell voltage of all cells reaches 0.7 V without any cell reaching 0.9 V. If the measurement is interrupted by a cell reaching 0.9 V, the measurement condition of the hydrogen flow rate and humidifying condition should be changed. Please refer to the text about how to choose the testing conditions.
- (7) Calculation of flow rate deviation between cells in the stack
The current density $I_{0.4}$ when the cell voltage becomes 0.4 V is measured for all cells in the stack. The flow rate deviation ratio is obtained as the ratio of $I_{0.4}$ of each cell divided by the average $I_{0.4}$ of all cells.

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