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Short communication

Measuring method for flow rate distribution between cells in a polymer electrolyte fuel cell stack

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ABSTRACT

The fuel cell transmogrified from a single cell that was the research object to stack is used in various fields such as cars, portable power sources, and fuel–cell cogeneration systems. It is preferable in the stack for the flow rate distribution between cells to be uniform because of the performance gain in power generation efficiency and longevity. As for the flow rate distribution between cells, the method for measuring using smoke in the measuring method and making visible the heat distribution in the stack is reported. However, a research stack was used with these measures, not a stack for practical use. In this report, a method for measuring the flow rate distribution between cells which can also be used for a cell that uses hydrogen limiting current in practical use was examined.

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 the portable power source for cogeneration, electric vehicles, and 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.

This report proposes a new technology that measures the flow distribution between cells in the stack.

The example of measuring the flow distribution between cells in an actual stack has not been reported at all until now. In the field of the molten carbonate fuel cell (MCFC), a method for measuring flow rate distribution in a model stack containing a hot wire sensor [1] was reported. Another method supplying air mixed with smoke in a visualized model stack was also reported [2]. However, these reports are the result of studies using a model stack made of materials different from those in an actual stack. It is important to measure the flow distribution for the structure and size of the PEFC stack after assembly is complete, because the stack is constructed by assembling soft materials and hard materials in layers and the dimensions of the flow channels may change due to the stacking.

In the PEFC field, 20 pressure sensors were installed in the downstream header and the upstream header each in the test stack made from actual stack materials, and the stack internal pressure distribution was measured [3]. However, the flow rate in each cell cannot be obtained because, in this method, there is a possibility that the flow rate is different according to the cell structure even if the differential pressure is the same.

In this study, the proposed new technology is a nondestructive method for measuring the flow distribution between cells applicable to an actual PEFC stack that does not contain an embedded sensor. The principle of the measurement is to measure the flow rate by using the hydrogen limiting current. It is a method for measuring the current value during a rapid voltage change when the supplied hydrogen is consumed completely. This principle is based on Faraday's law, and it is applied to products such as a zirconia-type oxygen flowmeter and hydrogen detectors. However, there is a possibility that electrode damage occurs because hydrogen is lacking when the hydrogen limiting current is measured in the PEFC [4]. In order to avoid electrode damage, a second fuel is mixed with hydrogen. When hydrogen shortage occurs, the second fuel is expected to react and suppress the electrical potential rise at the anode. Dimethyl ether (DME) a fuel for a direct fuel cell [5] is a gas

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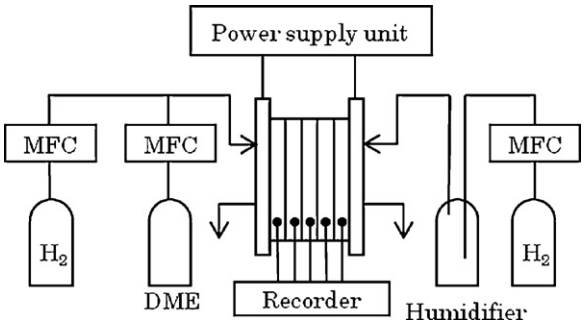

Fig. 1. Scheme of measurement system.

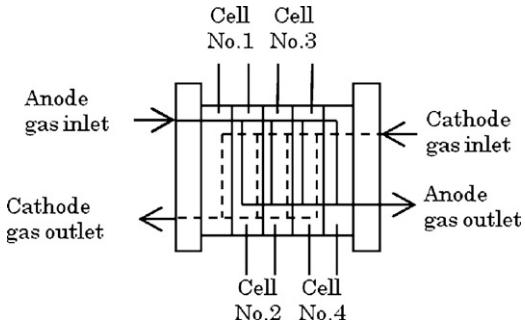

Fig. 2. Scheme of measurement for a 4-cell stack.

at standard temperature and pressure and was used in this study as the second fuel. Hydrogen was supplied to the cathode. We then measured the voltage of each cell while monitoring the anode electrical potential of each cell so as not to reach a high potential that may cause electrode degradation. In this study, the gas is supplied to each cell in parallel via the manifold, and the gas flows through the separator channel in each cell. It is clear that uneven channel size of each cell causes a difference of the flow rate between cells. However, it is known that not all of the gas passes through the channel in the cell and that there is a shortcut gas flow that passes the gas diffusion layer (GDL), etc. [6]. Therefore, not only the difference in the channel size but also the difference in the shortcut passage affects the flow rate distribution between cells.

When this method is put to practical use, we expect that determining the cause of the different flow rate between cells becomes possible, and this will lead to the development of a stack with few deviations in the flow rate between cells because the flow rate distribution can be measured. When thin separators are used to decrease the stack height, it is desirable to apply this method carefully so as to measure the exact uniform flow rate distribution between cells. This method is expected to contribute to realizing high fuel utilization of the stack because of the uniform flow rate distribution. The efficiency of the system may increase with fuel utilization because of decreasing wasted hydrogen. High fuel utilization may also allow the reformer to be miniaturized due to decreasing required hydrogen.

Moreover, this method may be applied to non-destructive shipment examination of the product, thus contributing to the enhancement of product reliability.

2. Experimental

Fig. 1 shows the scheme of the system for measuring the flow rate distribution between cells in a stack.

Hydrogen, DME or a gas mixture of hydrogen/DME is supplied to the anode. Hydrogen is supplied to the cathode. The cell voltage then shows the anode potential versus a reversible hydrogen electrode. Because this method was aimed at a convenient examination, the measurement environment was assumed to be room temperature, normal pressure, and no humidification. However, to control the dryness of the electrolyte membrane, the cathode gas is fully humidified at normal temperature. The hydrogen flow rate to the cathode is controlled by a mass flow controller (MFC) that has an accuracy of $\pm 10\,\mathrm{ml\,min^{-1}}$ on a full scale of $1000\,\mathrm{ml\,min^{-1}}$. The hydrogen flow rate at the anode is controlled by an MFC that has an accuracy of $\pm 0.5\,\mathrm{ml\,min^{-1}}$ on a full scale of $50\,\mathrm{ml\,min^{-1}}$. The DME flow rate is controlled by an MFC that has an accuracy of $\pm 5\,\mathrm{ml\,min^{-1}}$ on a full scale of $500\,\mathrm{ml\,min^{-1}}$. The structure of a single cell used in this experiment has a single serpentine channel and the electrode area is $25\,\mathrm{cm^2}$. A stack in this study is composed of 4 cells. Each cell has a 1 mm × 1 mm single serpentine channel and the electrode area $25\,\mathrm{cm^2}$. The gas is supplied to each cell in parallel as shown in Fig. 2 through a manifold of $\varphi 3$. Nafion@NR-212 was used as the electrolyte membrane. Carbon-supported platinum (Pt/C) was used as the electrode catalyst, and the Pt loading of both anode and cathode was $0.5\,\mathrm{mg\,cm^{-2}}$. In the measurement of the hydrogen limiting current, the current was increased step rise at a value of 0.005–0.05 A with a holding time of 1 min. The measuring range of the cell voltage was 0–0.9 V because a potential higher than 0.9 V was known to cause oxidative degradation of the electrode. The electrical current was interrupted when the voltage of any cell reached 0.9 V, and the measurement was stopped.

3. Results and discussion

3.1. Hydrogen limiting current measurement in a single cell

Fig. 3 shows the measured result of the hydrogen limiting current when supplying a mixed gas of hydrogen and DME to the anode. A single cell was used and the cell voltage change was measured on changing the electrical current. At this time, DME flow rate was $50\,\mathrm{ml\,min^{-1}}$ and the hydrogen flow rate was 4, 6, 8, or $10\,\mathrm{ml\,min^{-1}}$ to the anode. The hydrogen flow rate to the cathode was $50\,\mathrm{ml\,min^{-1}}$. In Fig. 3, when the cell voltage rises rapidly at a certain electric current density and the cell voltage reaches about 0.7 V, the rising inclination becomes small. Because the current density where the cell voltage rises rapidly is nearly proportional to the hydrogen flow rate, it is clear that this indicates the hydrogen limiting current. In this report, the current density immediately before the inclination $\Delta V/\Delta I$ of the cell voltage rise became $50\,\Omega\,\mathrm{cm^{-2}}$ or greater was considered to correspond to the hydrogen limiting current. The hydrogen limiting current density calculated from the anode hydrogen flow rate using Faraday's law agrees with the measured hydrogen limiting current density as shown in Fig. 4. This shows that the hydrogen flow rate is computable by measuring the

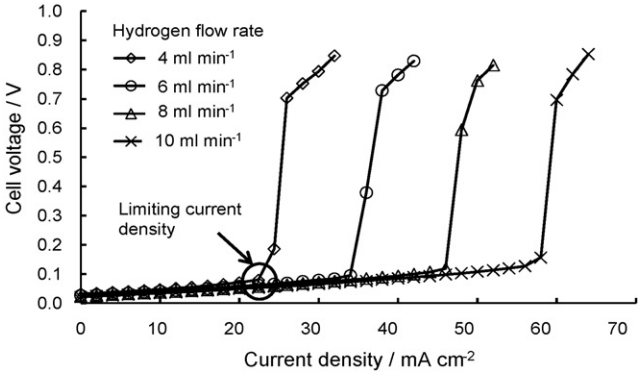

Fig. 3. Measured hydrogen limiting current density for a single cell using a mixed gas of hydrogen and DME.

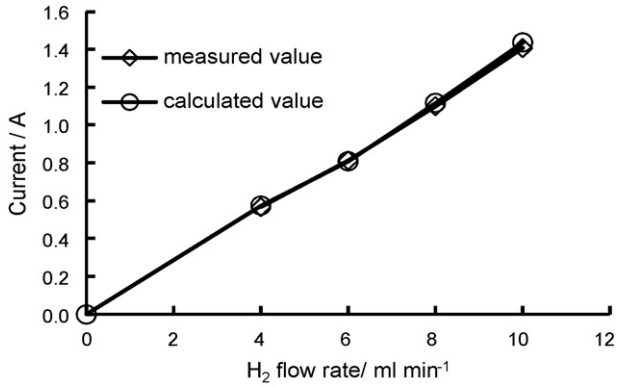

Fig. 4. Comparison between experimental data and the calculated value of hydrogen limiting current.

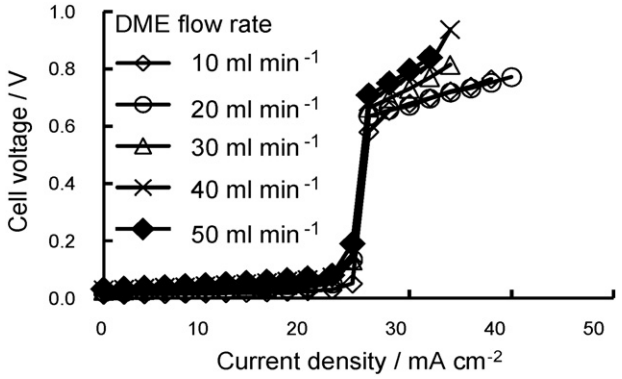

Fig. 5. Influence of DME flow rate on hydrogen limiting current characteristics for a single cell.

hydrogen limiting current characteristic. Moreover, mixing DME causes the inclination in the cell voltage rise to become small at a voltage higher than about 0.7 V as shown in Fig. 3. When DME is not added, it is well-known for the cell voltage to continue rising to more than 1 V, and electrode damage due to the hydrogen shortage occurs. Fig. 3 shows that the cell voltage rise is suppressed by mixing DME, thus is a result showing that measurement of the hydrogen limiting current characteristic is possible without causing electrode damage.

Fig. 5 shows the influence of the DME flow rate on the hydrogen limiting current density when the hydrogen flow rate is constant at 4 ml min^{-1}. Because the hydrogen concentration in the mixed gas is fixed, the flow rate of the mixed gas can be obtained by measuring the hydrogen limiting current.

Moreover, the inclination $\Delta V/\Delta I$ in the region of 0.7 V or higher becomes larger with an increase in DME flow rate as shown in Fig. 5. Because this is a situation in which hydrogen is consumed completely in this region, and DME then reacts, the large inclination in $\Delta V/\Delta I$ indicates the difficulty of the DME reaction. It is known that moisture greatly affects the difficulty of the DME reaction in the research on direct-type DME fuel cells [7]. The difficult DME reaction as shown in Fig. 5 may relate to the cell drying due to the large flow rate of DME gas. The inclination $\Delta V/\Delta I$ in the region of 0.7 V or higher does not influence the measured value of the hydrogen limiting current. It influences the permissible current for the measurement.

3.2. Hydrogen limiting current measurement in a stack

A 4-cell stack as shown in Fig. 2 is connected with the measuring system of Fig. 1. Figs. 6–8 show the measured results for the hydrogen limiting current. Fig. 6 shows the hydrogen limiting current for each cell in the stack on supplying hydrogen at 200 ml min^{-1} to the cathode and a mixed gas at a hydrogen flow rate of 8 ml min^{-1} and a DME flow rate of 200 ml min^{-1} to the anode. No. 1 cell has reached 0.9 V before No. 2 and No. 4 cells reach the hydrogen limiting current, although the hydrogen limiting current of No. 1 and No. 3 cells could be observed. Fig. 7 shows the measured result when only the hydrogen flow rate to the anode is changed from 8 ml min^{-1} (Fig. 6) to 4 ml min^{-1}. The hydrogen limiting current of each cell could be measured before No.1 cell reached to 0.9 V, because the hydrogen limiting current of each cell had decreased. When Fig. 7 is compared with Fig. 6, the electric current density of the *X*-axis is 1/2, but the ratio of the hydrogen limiting current density between cells is almost the same. The $\Delta V/\Delta I$ at 0.7 V or higher is also almost the same as that at about 0.2 V at 2 mA cm^{-2}. The permissible excessive current density of the DME reacting region from 0.7 V to 0.9 V is con-

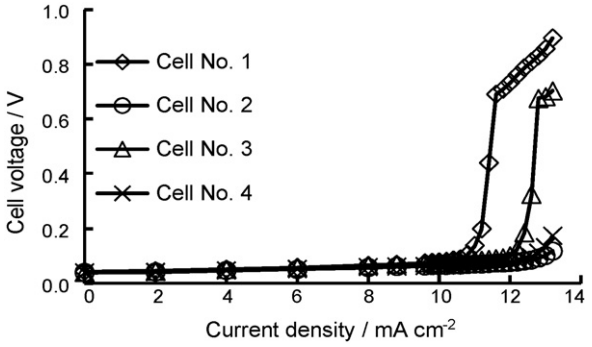

Fig. 6. Hydrogen limiting current characteristics for a 4-cell stack using mixed a gas of H_2 (8 ml min^{-1}) and DME (200 ml min^{-1}).

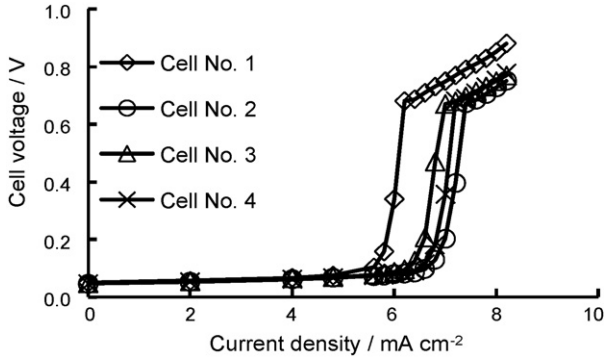

Fig. 7. Hydrogen limiting current characteristics for a 4-cell stack using mixed a gas of H_2(4 ml min^{-1}) and DME(200 ml min^{-1}).

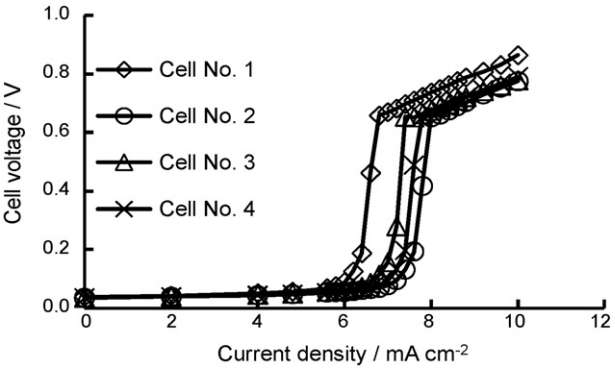

Fig. 8. Influence of DME flow rate on hydrogen limiting current characteristics for a 4-cell stack using a mixed gas of H_2 (4 ml min^{-1}) and DME (100 ml min^{-1}).

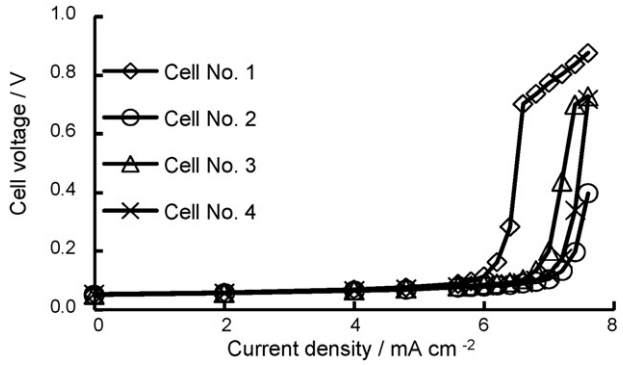

Fig. 9. Influence of DME flow rate on hydrogen limiting current characteristics for a 4-cell stack using a mixed gas of H_2 (4 ml min^{-1}) and DME (300 ml min^{-1}).

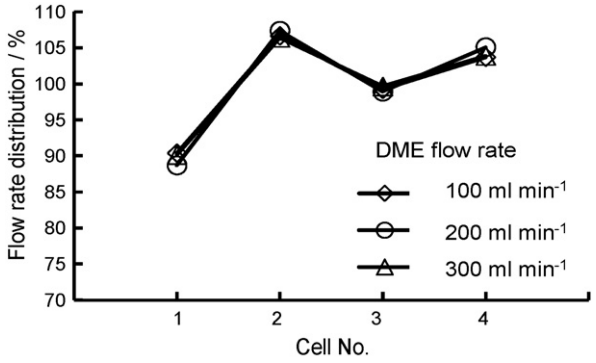

Fig. 10. Flow rate between cells calculated from the measured results of Figs. 7–9.

stant in spite of the DME flow rate. If the amount of the hydrogen stream is small, the measurement becomes possible even if there is a large flow rate deviation between cells.

Fig. 8 shows the hydrogen limiting current characteristic in which the DME flow rate is changed to 100 ml min^{-1}, and Fig. 9 shows the hydrogen limiting current characteristic in which the DME flow rate is changed to 300 ml min^{-1} with the hydrogen stream of 4 ml min^{-1} kept constant.

In this measuring method, the ratio of the flow rate between cells is the ratio of the hydrogen limiting current. If F_i is the ratio (%) of flow rate for each cell to the average flow rate for the cell in the stack, F_i is calculated from the measured hydrogen current density I_i using the flowing formula.

$$F_i = \frac{I_i}{\bar{I}} \times 100$$
$$\bar{I} = \frac{1}{n_i} \times \sum_{i=1}^{n} I_i \quad (1)$$

Fig. 10 shows the results of the flow rate between cells calculated from the measured results of Figs. 7–9 using Eq. (1).

Though the flow channel dimensions of each cell are the same, a flow rate distribution from 89% to 108% is detected. The No. 1 cell has the lowest flow rate ratio. Though the No. 1 cell is located at a position nearest the gas inlet of the stack, the lowest flow rate may not be related to the cell position because the effect of the DME flow rate on the flow rate distribution is small. The cause of nonuniform flow distribution in this experiment may be a shortcut flow in the cell such as that through the gas diffusion layers. The purpose of this report is to present the new measuring method for the flow rate distribution between cells. Future tasks are to investigate the cause of the flow rate deviation between the cells in the stack and to develop stacks with a uniform flow rate distribution.

4. Conclusions

(1) The purpose of this report is to present the new measuring method for the flow rate distribution between cells.
(2) The method of this report involves measuring the hydrogen limiting current of each cell in the stack while supplying a mixed gas of hydrogen and DME to the anode and hydrogen to the cathode.
(3) Because DME functions as a fuel, the electrode potential is suppressed. Degradation of the electrode then does not occur even under the condition of insufficient hydrogen.
(4) This method does not measure the flow distribution under a real operating condition but is a method of examining whether the developed stack has a uniform flow distribution.

We expect to apply the methodology reported in this paper to the product inspection, etc., of the PEFC stack as a non-destructive test at normal temperature and normal pressure.

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