

Short communication

Temperature gradients measurements within a segmented H₂/air PEM fuel cell

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Abstract

A commercial Nafion 112 membrane loaded with the catalysts and catalysts' support in a segmented way was used for a H₂/air PEM fuel cell. On this membrane, the anodic and cathodic catalysts with their support were loaded on five consecutive places in a back to back style forming five catalyst islands of same dimensions on each side of the membrane. So, five sub-fuel cells were existed in one fuel cell compartment. These subcells are connected ionically but not electronically. The polarization behavior for these subcells was measured separately when the other subcells were at a zero load and simultaneously when they were polarized at the same load. Also, the temperature gradient within this segmented PEM fuel cell was measured in front of cathode side of the sub-fuel cells when hydrogen and air gases were flown in a parallel or opposite direction to each other. Temperature gradients were correlated with the observed performance of the fuel cell.

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1. Introduction

A fuel cell is simply a galvanic cell, which converts the chemical energy into electrical energy. Proton exchange membrane fuel cell (PEMFC) is one of several types of fuel cells. Although having the lowest efficiency relative to the other types of fuel cells, the PEMFC is considered to be the most suitable fuel cell for small and mobile technologies since it has the least management problems [1]. It has been shown that there are different local environments inside PEMFCs [2,3] which resulted in an irregular performance. A considerable effort has been paid to investigate the different factors affecting the performance of PEMFC, e.g., the gases' humidity [4], flow rate [5,6], and pressure [1], the membrane durability [7] and thickness [8,9], the fluid dynamics [10], the catalyst functionality [11], layer thickness [12], corrosion tendency [13] and catalyst support corrosion [14,15] and the working temperature [16].

The importance of studying the temperature within the fuel cell arises from its effect on the performance through influencing many other parameters. For example, it affects the relative humidity within fuel cells [4], water transport through membranes [17], kinetics of the electrode reactions [18], catalyst support and membrane degradation [19,20]. Besides the above, knowing the temperature distribution is crucial for modeling and improving the performance of the PEMFCs [2,21]. Hakenjos et al. used infrared thermography and optical water surveillance to study the local temperature and water distribution within a PEMFC [22]. Also, tunable diode laser absorption spectroscopy was employed to measure the temperature in the cathode gas channel [16]. Siroma et al. used thermocouples at the surface of 100 catalyst segments on the membrane [23].

The aim of this work is to investigate the temperature gradient within a H₂/air PEM fuel cell and correlate this gradient to the performance of a segmented fuel cell. The reason of using the electronically separated catalyst areas is to simulate a real case when a degradation of the catalyst and catalyst support occurs, which results in disconnected catalyst areas. These disconnected catalyst areas will confront a different local environment due to the temperature gradient and the water distribution which will

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affect the performance of the fuel cell. Different from the work of Siroma et al., the surface area of the individual subcells of the catalyst was larger and their number is less in order to measure a significant increase in temperature. Also, in this work, the polarization behavior of the formed subcells was investigated separately when the other subcells were at a zero load and simultaneously when the other subcells were polarized at the same load. Besides studying the polarization behavior of the subcells, the effect of the flow direction of H₂ and air gases (whether parallel or opposite to each other) on the temperatures within the fuel cell was investigated.

2. Experimental

The membrane electrode assembly (MEA) was purchased from NF Corporation, Japan. Anodic catalyst (platinum–ruthenium) of a surface density 1 mg cm⁻² was hot-pressed on five consecutive places on one side of a Nafion 112 membrane. The membrane thickness was 50.8 μm and its dimensions were 16 cm × 9 cm. Cathodic catalysts (platinum) of the same surface density were also pressed simultaneously on the same five locations on the other face of the membrane in a back to back style. This forms five catalyst islands on each side of the membrane. The dimensions of these islands were 3.0 cm × 1.5 cm and they were parallel to their gas flow channel. Each pair of the islands opposite to each other is acting as a subcell. The distance between successive subcells is 0.6 cm (see Fig. 1). This means there are five sub-fuel cells in one fuel cell compartment. These subcells are connected ionically through the membrane but they are electronically disconnected. Fig. 1 is a schematic diagram showing the Nafion 112 membrane loaded with five anodic catalyst islands and five cathodic catalyst islands. It also shows the gases' flow channels in front of these catalyst islands. The gases' flow direction shown in Fig. 1 is the parallel flow mode. In the opposite flow mode, the hydrogen gas comes in from the same entrance at Subcell 1, but air enters from the opposite direction at Subcell 5 instead of entering at Subcell 1.

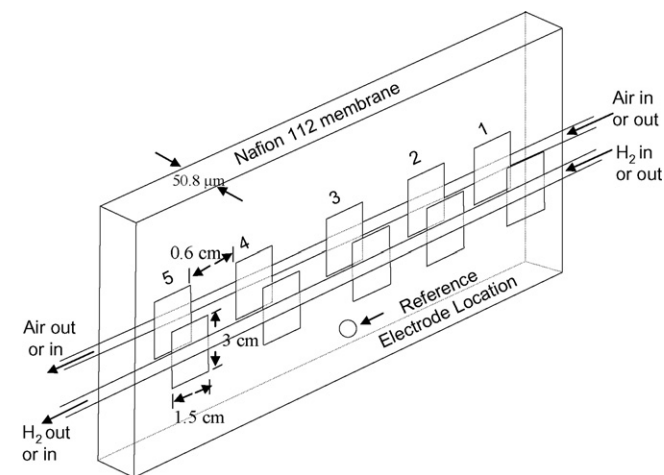


Fig. 1. Schematic diagram shows the Nafion 112 membrane loaded with five anodic catalyst islands and five cathodic catalyst islands and the gas flow channels in front of these catalyst islands.

A PC-interfaced fuel cell system (MTB-36714) purchased from NF corporation, Japan, was used to maintain the gases' pressures, temperatures, humidity and flow rates. An eight-channel multipotentiostat unit purchased from Toho Technical Corporation, Japan, interfaced with the same PC was also used to measure the *I*–*E* characteristic curves for the different subcells simultaneously or separately.

The reference electrode was a reversible hydrogen electrode (RHE) with its own H₂ gas feed, and so it does not use the H₂ that passes in front of the catalyst island on the anode side. This is to assure a constant hydrogen gas pressure for the reference electrode.

Temperatures of the anode and cathode gas lines between the humidifiers and the cell were maintained at the same cell temperature (65 °C in front of Subcell 3) to ensure that water condensation did not occur or a temperature gradient within the fuel cell was created. The temperature of the fuel cell compartment was controlled externally using a temperature controlling unit with two controlled-heating pads and a thermocouple embedded within the fuel cell to achieve the desired temperature of the fuel cell. The thermocouple sensor can be embedded at five different locations in front of the cathodic catalyst islands. For measuring the temperature gradient within the fuel cell, the thermocouple sensor was placed in front of the cathodic catalyst island for 1 min until reaching a steady state reading. It is important to mention that the thermocouple sensor does not interfere with the gas pathway. The initial temperature of the fuel cell compartment was always kept at 65 °C in front of Subcell 3. Since there was always a temperature gradient within the fuel cell even at open circuit potential (OCP), it was necessary to adjust the temperature to a reference Subcell which was chosen to be Subcell 3 as its temperature is an intermediate one.

Polarization curves were measured using a galvanostatic technique by applying 12 different currents, starting from 0 to –160 mA, for 120 s each and measuring the corresponding potentials. The polarization curves for the different subcells were recorded either separately while the other subcells are at open circuit potential or simultaneously while the other subcells are polarized at the same load.

The gases' flow rate was kept constant during all the measurements at 300 and 1000 cm³ min⁻¹ for H₂ and air, respectively. These flow rates satisfy a ratio slightly higher than the stoichiometric ratio of H₂ and O₂ gases. The gas pressure was kept constant accordingly at 1.5 and 25.3 kPa for H₂ and air, respectively. The relative humidity of the hydrogen and air gases (RH) was 100%, i.e., RH was symmetric [4].

It is worthy to mention here that all experiments are confirmed to be quite reproducible.

3. Results and discussion

Figs. 2 and 3 show the *I*–*E* characteristic curves for the cathode of each subcell at a temperature of 65 °C. Air and fuel gases flow in a parallel direction to each other starting from Subcell 1. The only difference is as follows: in Fig. 2 all the five subcells were polarized at the same load simultaneously while in Fig. 3 they were polarized separately i.e. only one subcell is polarized

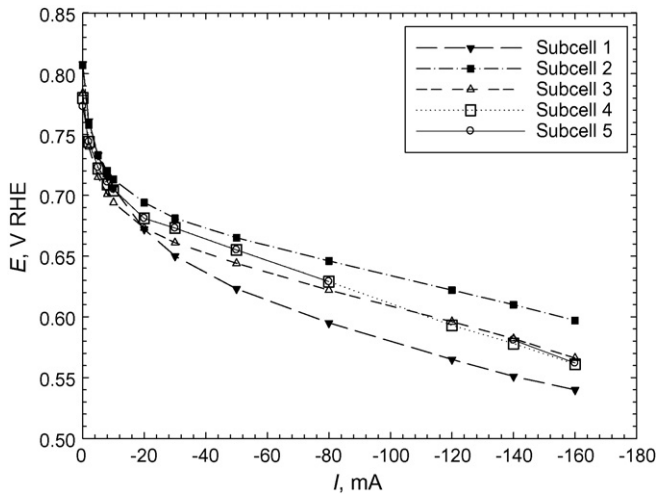


Fig. 2. Polarization curves of the H₂/air PEM fuel cell cathodes. Potentials corresponding to the applied currents were measured simultaneously. T_{cell} : 65 °C, RH: 100%, H₂ and air gases flow was parallel starting from Subcell 1.

while the rest subcells are at a zero load (OCP). An interaction between the five subcells can be recognized from these two figures where the cell performance changes at the same load depending upon whether the measurements were simultaneous (see Fig. 2) or separate (see Fig. 3). Fig. 4a and b shows the I - E characteristic curves for the cathode of Subcell 1 in the case of simultaneous and separate measurements. The more positive potential is required to maintain the applied load, the better the performance of this subcell is. Fig. 4a and b reveals that the performance of each subcell during the separate measurements is better than that during the simultaneous measurements when the load is small. But, at high loads the behavior is reversed where the performance of the subcells is better during the simultaneous measurements than the separate ones. At medium loads (between -70 and -80 mA), the performance of subcells is almost the same. This behavior of Subcell 1 is the same as that of the other subcells (data not shown).

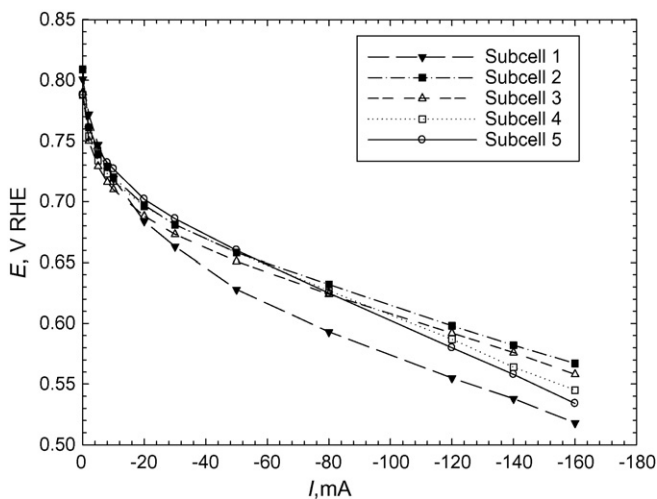


Fig. 3. Polarization curves of the H₂/air PEM fuel cell cathodes. Potentials corresponding to the applied currents were measured separately. T_{cell} : 65 °C, RH: 100%, H₂ and air gases flow was parallel to each other.

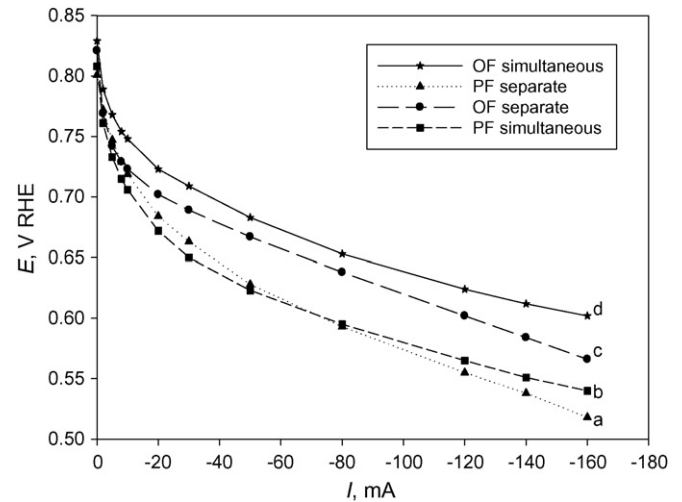


Fig. 4. Polarization curves for the H₂/air PEM fuel cell cathode of subcell 1 at different gases flow modes and different measurement modes: (a) the parallel flow – separate measurements, (b) the parallel flow – simultaneous measurements, (c) the opposite flow – separate measurements and (d) the opposite flow – simultaneous measurements. T_{cell} is 65 °C and RH is 100%.

The behavior might be a consequence of the water produced within the fuel cell which affects the membrane water uptake [24], mass transport of fuel and air [25], temperature gradient within the fuel cell and the resistance of the membrane. The resistance of the membrane is reflected in the IR potential drop which affects the potential (E_{cell}) of the fuel cell according to Eq. (1) [26]

$$E_{\text{cell}} = E^0 - IR_{\text{cell}} \quad (1)$$

where E^0 is the cell potential when the cell resistance (R_{cell}) is zero.

It is worthy to mention here that the RH was maintained at 100% during the whole measurements in this work to ensure that the resistance of the membrane is almost constant during the whole set of measurements at different loads [4]. So, the difference in performance cannot be attributed to the change in the resistance of the membrane which would depend on the amount of the produced water.

Another possibility is that the produced water may affect the rate of the reactions at the electrodes of subcells by changing the rate of mass transfer of the reactants in the gas diffusion layer [16] and/or by changing the temperature which directly influences the exchange current density [1].

For the mass transfer possibility, it is expected that the larger the amount of water produced is, the lower the rate of mass transfer will be. At higher loads, the amount of the produced water is higher, and so it is expected that the performance will be better in the case of separate measurements (less amount of water produced) than in the case of the simultaneous ones (larger amount of water produced). But, in fact the opposite happened where the simultaneous polarization of the subcells resulted in a better performance than the separate ones when high loads were applied. This might indicate that water did not affect the rate of reactions by changing the rate of mass transfer of the reactants (fuel and air) to the electrode surface. The uncertainty in the word “might”

here is because at high loads during the simultaneous measurements, the temperature increases more largely than in the case of the separate measurements (data not shown), and so it is not possible to confirm that water did not affect the rate of mass transfer of the reactants to the electrode surfaces during the separate and simultaneous modes of measurements. In other words, the amount of water is also related to another parameter which is the temperature. In fact water may decrease the rate of mass transfer but the increase in temperature might counterpart this by increasing the exchange current density. The higher temperatures that were recorded during the simultaneous measurements than in the case of the separate ones may be referred to the decrease in the heat dissipation rate when the neighboring subcells are at higher temperature because of the high load on them.

To quantitatively study how local temperatures changed at the different subcells [27] which directly affects the exchange current density [1] at these subcells, the local temperatures were measured and the results are illustrated in Fig. 5. In this figure, the local temperatures in front of each subcell were measured during a simultaneous polarization for the whole subcells at a certain load. Four loads were chosen; 0, -5, -50 and -160 mA. These loads represent low, medium and high loads. Gases' flow mode was parallel. Fig. 5 reveals some interesting points:

- (i) The temperature at each subcell increases as the load increases. This is attributed to the increase in the oxygen reduction reaction rate at the cathode as the load increases, the electronic resistance and also to the frictional resistance of H^+ ions as they transfer through the membrane and the cathode [28]. It is worthy to mention here that the electronic resistance is directly proportional to the operating temperature [29].
- (ii) At low loads, the amount of the produced water is larger at Subcells 3, 4 and 5 than at Subcells 1 and 2 where gases drift the produced water to their exit which is beyond Subcell 5, and so water has a gradient in the whole subcells, that is, its minimum at Subcell 1 and its maximum at Subcell 5.

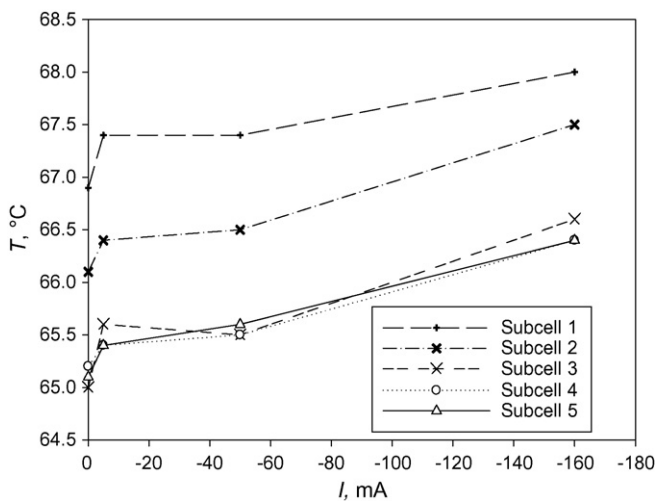


Fig. 5. Temperature profiles within the segmented H_2 /air PEM fuel cell for each subcell as a function of load. Gas flow direction was in the parallel mode and T_{cell} was $65^\circ C$. Subcells were polarized simultaneously.

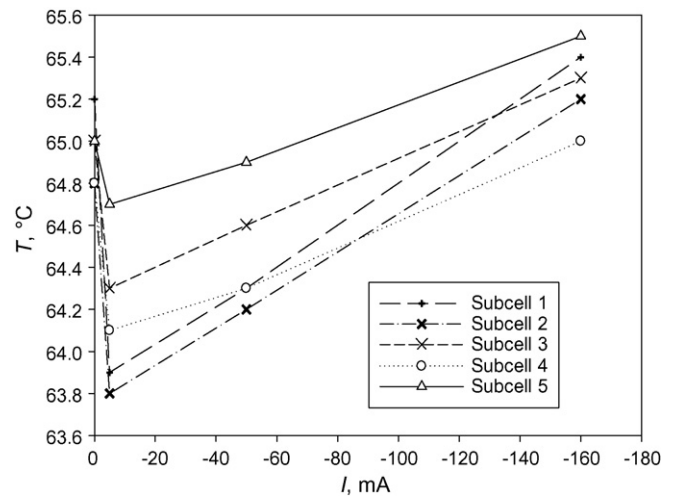


Fig. 6. Temperature profiles within the segmented H_2 /air PEM fuel cell for each subcell as a function of load. Gas flow direction was in the opposite mode and T_{cell} was $65^\circ C$. Subcells were polarized simultaneously.

- (iii) Although the amount of water produced at high load was large, it was not enough to decrease the temperature below the starting temperature ($65^\circ C$) or to keep it.
- (iv) Regardless of the value of the applied load, the highest temperature was always found to be at Subcell 1. For the same load, temperature decreases gradually through Subcell 2 to Subcell 3 before it becomes almost constant at Subcells 4 and 5. It is always found that the temperature difference between the maximum and the minimum temperature at any load is almost $2^\circ C$. This temperature gradient might be attributed to the water gradient in the whole subcells.

To study the correlation between the water gradient and the temperature gradient, the gases' flow direction was reversed. In this case, a different water distribution took place and a different temperature profile was observed, which was expected to produce a different polarization behavior. Fig. 6 shows the temperature profile within the fuel cell in front of each subcell. The subcells were polarized simultaneously but the flow of gases was in the opposite mode. We can see that temperature gradient is scarcely found. Temperatures are fluctuating up and down. Regardless of the applied load, the temperatures of Subcell 2 are always lower than those of Subcells 1 and 3. Also the temperature of Subcell 4 is always lower than those of Subcells 3 and 5. The largest temperature difference between the subcells at the same load was only $0.8^\circ C$ at a load of -5 mA. As the applied load increases the temperature difference decreases. It reached to only $0.3^\circ C$ at -160 mA. The reason for that is the increase in the amount of the uniformly distributed water as the load increases which is not the situation in the case of the parallel flow of air and fuel gases. Usually the highest temperatures recorded are at the subcells next to the air and the hydrogen gas inlets (Subcells 1 and 5). Similar to the case of the parallel flow, it was found that temperature of Subcell 2 is always lower than those of Subcells 1 and 3 and the temperature of Subcell 4 is always lower than those of Subcells 3 and 5. Fig. 4c and d compare the I - E characteristic curves for Subcell 1's cathode at a

Table 1
Temperature profile at the subcells at open circuit potential

Subcells	T (°C)
Subcell 1	29.2
Subcell 2	29.2
Subcell 3	29.1
Subcell 4	29.1
Subcell 5	28.9

Temperature is 28 °C (room temperature) and RH is 100%. Gases' flow mode is parallel. All measurements were taken after 1 h from running the gases.

cell temperature of 65 °C and a RH of 100%. Gases' flow was in the opposite direction. In Fig. 4c, the cathode of Subcell 1 was polarized separately, while in Fig. 4d it was polarized simultaneously with the other subcells at the same load. In these figures it is clear that the performance of the subcell during the opposite flow measurements is better than in the case of the parallel flow ones even though the temperature is higher within the fuel cell in the case of the parallel flow (compare Figs. 5 and 6). This can be understood in the frame of water distribution within the fuel cell where in the case of the opposite flow mode, the water distribution is more uniform [30] across the five subcells than in the case of the parallel flow.

In Figs. 5 and 6, it is easy to recognize that the temperatures at the different subcells are not the same at the open circuit potential (zero load). Now one may ask whether these temperature differences of the different subcells at the open circuit potential might be attributed to the tube heating which increased the temperature of the inlet gases and might have resulted in a local dryness for the membrane areas near the gas inlet. To check this possibility, the temperatures of the different subcells were measured at room temperature (28 °C) and RH is 100% to eliminate the possibility of a local dryness of the membrane as a result of the tube heating. Table 1 summarizes the results of these measurements. It can be easily recognized from the table that the temperature inside the fuel cell is higher than the room temperature and there is also a temperature gradient within the fuel cell with a maximum at Subcells 1 and 2 (29.2 °C) and a minimum at Subcell 5 (28.9 °C). These measurements eliminate the possibility that the tube heating is the reason for the temperature gradient observed at a load of 0 mA. In fact, the fuel crossover and/or the internal (stray) current due to electron conduction through the electrolyte membrane are considered as the reason for this temperature gradient. This hydrogen permeation or electron crossover is by several orders of magnitude lower than hydrogen consumption rate or total electrical current generated when high loads are applied. However, at small loads or open circuit potential, the internal current density which ranges from 6 to 12 mA cm⁻² with an estimated uncertainty of ±7%, has a significant effect on the cell potential [29,31]. This crossover phenomenon occurs with a higher possibility as the temperature increases [29].

4. Summary and conclusions

Using the segmented fuel cell, where the segments are connected ionically not electronically, is a promising technique to study the localized environment within the fuel cell. A temper-

ature profile within the segmented H₂/air PEM fuel cell was measured. It was shown that there is a temperature gradient exists and it depends on the gases' flow direction. This temperature gradient exists even at zero load and that phenomenon was attributed to the internal current and/or diffusion of hydrogen (fuel crossover) through the electrolyte. Also it is found that this gradient depends on whether the measurements in front of each segment were taken simultaneously where all segments are polarized at the same load or separately where the rest segments (subcells) are at a zero load. During the parallel and opposite flow modes, the temperature of Subcells 2 and 4 are less than those subcells located before and after them. The difference between the minimum and maximum temperatures of the different subcells was 2 °C when the air and H₂ gases flow parallel to each other. This difference is constant regardless the value of the applied load. In the case of the opposite flow mode, the temperature differences was 0.8 °C at low loads (0 and -5 mA) but as the load increases this difference decreases until it reached to 0.3 °C at the largest load (-160 mA). This was attributed to the increase in the amount of uniformly distributed water. Although the temperature within the fuel cell was higher in case of the parallel flow mode, the performance was worse than in case of the opposite flow mode. This was attributed also to the difference of water distribution within the fuel cell. Water management is very crucial for increasing the rate of the oxygen reduction reaction to improve the performance of PEMFC.

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