

Numerical Prediction of Water Content Distribution of Catalyst Coated Membrane of Fuel Cell Stack using Simulation

Kenji NAGUMO*

Masajiro INOUE*

ABSTRACT

The distribution of the water content of catalyst-coated membranes (CCM) is a factor that affects the durability of polymer electrolyte fuel cells. Because the in-plane distribution of the water content of the CCM in fuel cell stacks changes with changes in fuel cell operating conditions such as the coolant inlet temperature and the cathode electrode gas inlet flow rate, the setting of appropriate operating conditions is important in helping to ensure fuel cell durability.

The research discussed in this paper therefore developed a simulation making it possible to accurately predict operating conditions in a short period, and studied the effect of operating conditions on the in-plane distribution of the water content of the CCM. The results of this study indicated that the in-plane distribution of water content could be controlled via three operating condition parameters: the coolant inlet temperature, the cathode gas inlet pressure, and the cathode gas inlet flow rate.

Using the developed simulation and the results of the study of the effect of operating conditions on the in-plane distribution of the water content of the CCM made it possible to maintain the in-plane distribution within the upper and lower limit values and reduced variation in the distribution. It was also possible to apply accurate operating conditions in the 2016 FCV fuel cell stack in a period of several days.

1. Introduction

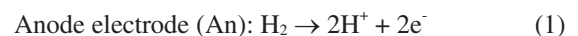
The environmental effects of CO₂, NO_x and other pollutants in automotive exhaust gases and the future depletion of fossil fuels make the development of clean, high-fuel-efficiency vehicles an urgent task. Seeking to realize a sustainable society based on energy circulation, Honda is engaged in research and development efforts in relation to fuel cell vehicles (FCV), which use hydrogen rather than fossil fuels, and fuel cell technologies⁽¹⁾.

Figure 1 shows an image of the structure of a fuel cell and its internal chemical reactions. The fuel cell (simply termed “cell” below) is made up of a membrane electrode assembly (MEA) and separators that form gas channels. A fuel cell stack (termed “stack” below) is created by layering multiple cells together.

The MEA is made up of a polymer electrolyte membrane (PEM), a catalyst layer (CL), and a gas diffusion layer (GDL). The PEM making up the CL are also called catalyst-coated membranes (CCM).

In the cell, a hydrogen oxidation reaction occurs at the anode electrode as shown by Eq. (1), and an oxygen reduction reaction occurs at the cathode electrode as shown by Eq. (2). These chemical reactions between hydrogen and

oxygen produce electricity and water.



In addition to seeking increased compactness and weight reductions, and realizing high-efficiency-generation performance with higher output, it is also necessary for the stacks fitted in FCV to display durability in relation

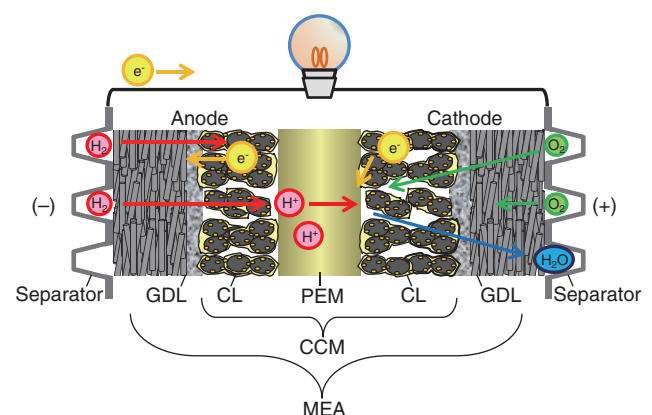


Fig. 1 Schematic image of fuel cell

* Automobile R&D Center

to the fluctuations in current load to which cells used in automotive environments are subjected. To help ensure durability in cells, it is necessary to maintain the surface area of the electrode catalysts, which contributes significantly to the output of the cell, and to maintain the thickness of the PEM, which functions both to separate the oxygen and the hydrogen that serves as fuel, and to electrically insulate the anode and the cathode.

The ability to maintain the surface area of the electrode catalysts and the thickness of the PEM is closely related to the in-plane water content of the CCM in the cell. Generally, when the water content is high, the electrode catalysts become coagulated due to dissolution, agglomeration, and redeposition, and the decline in the catalyst surface area accelerates. It has also been reported that when the water content is low, impurities and hydrogen peroxide accumulate in the PEM, promoting chemical degradation and reducing the thickness of the PEM⁽²⁾.

In order to increase the durability of automotive fuel cells, it is therefore necessary to identify the in-plane distribution of the water content of the CCM and control it to within the proper range.

2. Development Goals

The development of the stack for the 2016 model FCV therefore sought to realize increased stack durability through the development of multi-segment impedance sensors⁽³⁾ to identify the in-plane distribution of water content of the CCM in the cells and the calculation of the optimum in-plane water content distribution using simulations.

This paper will discuss prediction of the in-plane distribution of the water content of CCM in fuel cell stacks through the use of simulations.

As indicated above, the in-plane distribution of the water content of the CCM affects the durability of the cells, and the in-plane distribution of water content is itself significantly affected by the operating conditions of the cells. This means that it is important to set appropriate operating conditions in order to realize increased cell durability.

Six operating condition parameters are relevant in this process: the coolant inlet temperature and flow rate, and the inlet flow rate and pressure of the An and Ca gases.

Up to the present, the in-plane water content of the CCM in the cells has been measured using neutron radiography or weight measurement methods. However, in addition to the fact that a large number of combinations of parameters are possible, measurements also take time^{(3), (4)}, and it has been challenging to determine parameters with an adequate level of accuracy in a short period.

In order to address this issue, the research discussed in this paper first developed a simulation that would produce results displaying a good level of accuracy in a short period, and used the simulation in conducting advance studies of the effect of the various operating condition parameters on the in-plane distribution of the water content of the CCM.

The goal of the development of the simulation was to make it possible to accurately predict operating conditions that would maintain the in-plane water content of the CCM within the appropriate range in a short period using both the developed simulation and the results of the advance studies.

3. Development of Simulation

3.1. Simulation of In-plane Distribution of Water Content of CCM in Cells

In order to develop a method of simulation of the in-plane water content of CCM in cells, original Honda functional modifications were made to a commercially available polymer electrolyte fuel cell simulation software⁽⁵⁾.

3.2. Cell and Stack Configuration and Operating Conditions

Figure 2 shows a schematic image of the cells employed in the 2016 model FCV. The main structures applied in the simulation are the wave flow channel separators, the cooling structures serving every two cells⁽¹⁾, the stack transverse mounting structure, the structures generating counter flows of An and Ca gases and co-flows of Ca gas and coolant, and the independent gas flow control structure for the An and Ca via the resin frame structure around the MEA⁽⁶⁾.

These cells are organized into multiple layers to form stacks, and this was applied in the setting of conditions in the simulation (the setting of the periodic boundary conditions).

In the simulations of operating conditions, the current value was held constant, and the effect of the other parameters was determined. The value that has the greatest effect on the durability and reliability of the cells was employed as the current value.

3.3. Simulation Results and Characteristics

Figures 3(a) and (b) show the respective results of in-plane simulation of an A cell and a B cell. In the figures, “Ca in” represents the air inlet, “Ca out” represents the air outlet, “An in” represents the hydrogen inlet, “An out” represents the hydrogen outlet, “Coolant in” represents the coolant inlet, and “Coolant out” represents the coolant outlet.

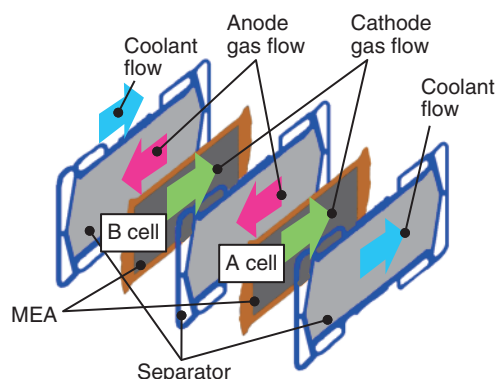


Fig. 2 Schematic image of cell structure

With regard to the characteristics of the cells, the in-plane water content of the CCM is lowest on the Ca gas inlet side, and increases towards the Ca gas outlet side. This is because the water produced on the Ca side flows towards the Ca outlet and accumulates there. However, the position in the cell that displays the highest value for the in-plane water content of the CCM is not the Ca gas outlet (P1 in the figure), but rather a position slightly further towards the Ca gas inlet side (P2 in the figure).

Figure 4 shows a schematic image that explains the characteristics of the in-plane distribution of the water content of the CCM. Two factors determine the in-plane water content of the CCM: the diffusion of water via the CCM due to the difference in water vapor partial pressure between the Ca and the An [(1) in the figure], and the amount of water carried away from the CCM by gas [(2) in the figure].

It is considered that the in-plane water content of the CCM does not reach its maximum at the position of the Ca gas outlet (P1 in Fig. 3) because the amount of water carried away from the CCM by An gas [(2) in Fig. 4] exceeds the amount of water that diffuses via the CCM [(1) in Fig. 4].

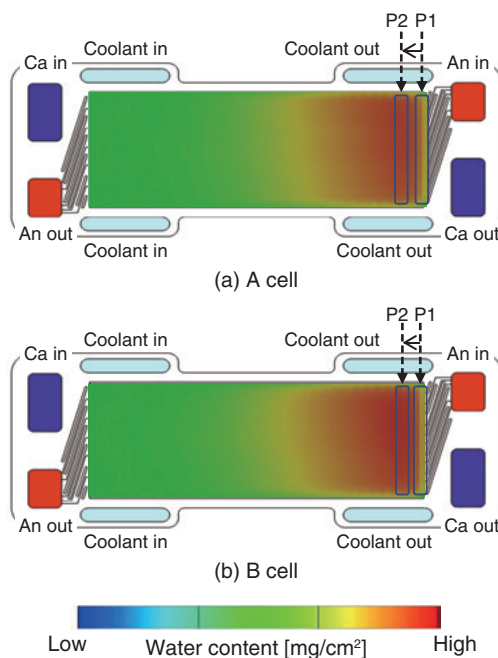


Fig. 3 Simulation results for water content distribution

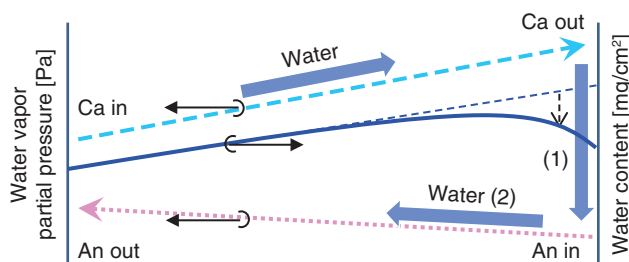


Fig. 4 Schematic image of movement of water in CCM

3.4. Comparison of Simulation and Test Results for Distribution of CCM Water Content

For the development of the stack employed in the 2016 model FCV, a newly developed sensor system⁽³⁾ was used in the measurement of the in-plane distribution of the CCM water content. Figure 5 shows a schematic image of the system. This system divides the cell in the in-plane direction into 75 segments to measure the in-plane distribution of water content. In order to compare the results of measurements using this system with simulation results, the simulation data was also divided into 75 segments, and an averaging procedure was applied to the data in each segment.

Figures 6(a) and (b) compare respective simulation results and measurement results for the A cell and the B cell. The comparison was conducted for the central group of segments, as shown in Fig. 5.

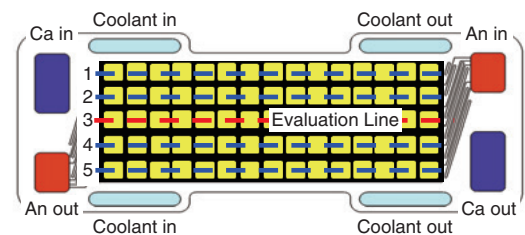
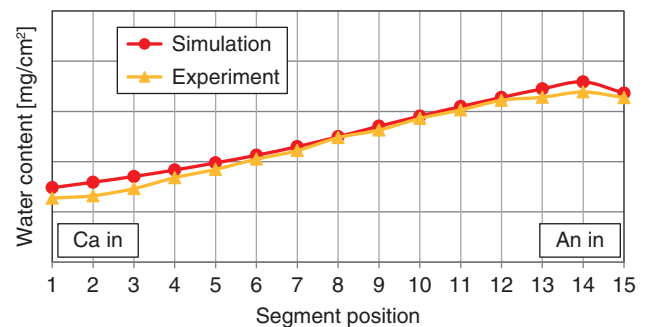
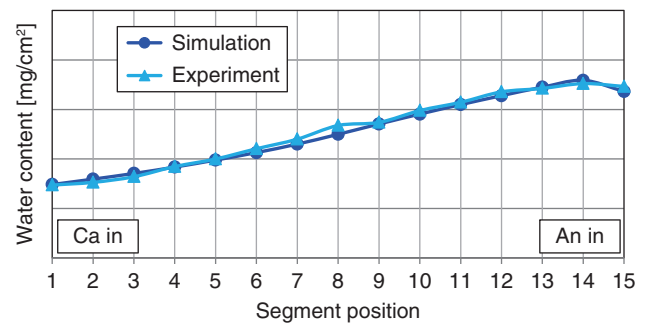


Fig. 5 Schematic image of 75-segment impedance measurement system



(a) A cell



(b) B cell

Fig. 6 Results of comparison of simulation and experiment

The stack employs cooling structures serving every two cells, and the measurement and simulation results correspond well in indicating no difference in the water content of the CCM in the A and B cells. In addition, the average predictive accuracy for the 75 segments was within 5.0%.

4. Study of Degree of Effect of Operating Condition Parameters

The operating condition parameters (the inlet temperature and flow rate of the coolant, the An gas inlet flow rate, the Ca gas inlet flow rate, the An gas inlet pressure, and the Ca gas inlet pressure) were varied, and the effect on the in-plane distribution of the water content of the CCM in the cells was studied. The respective operating conditions were changed $\pm 10\%$, encompassing the range in which the fuel cell system was able to continue to operate in relation to the operating conditions applied in Section 3.2.

Because both the A and B cells displayed identical changes in response to the changes in operating conditions, only the A cell results will be discussed below.

4.1. Effect of Coolant Inlet Temperature

Figure 7 shows simulation results with change in the coolant inlet temperature.

Change in the coolant inlet temperature does not produce any change in the tendency of the in-plane distribution of water content in the CCM, but does produce a significant change in the absolute value of the water content. When the coolant inlet temperature is increased, the temperature in the cell increases, and the in-plane water content of the CCM decreases; when the coolant inlet temperature is reduced, the temperature in the cell decreases, and the in-plane water content of the CCM increases.

4.2. Effect of Coolant Inlet Flow Rate

Figure 8 shows simulation results with change in the coolant inlet flow rate.

Change in the coolant inlet flow rate produces almost

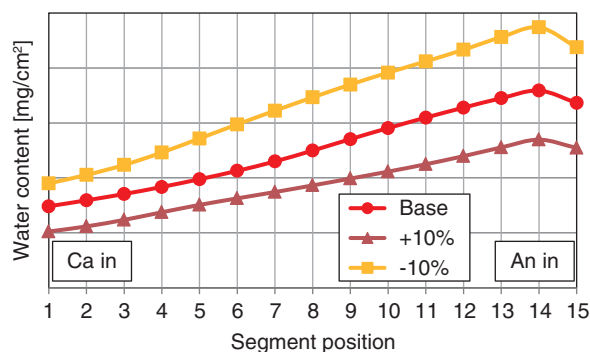


Fig. 7 Simulation results with change in coolant inlet temperature

no change in the in-plane distribution of the water content of the CCM. However, a consideration of detailed values indicates that when the coolant inlet flow rate is increased, a higher quantity of the heat produced by the CCM is carried away by the coolant. As a result, the temperature in the cell close to the coolant outlet (Segment position 14 in the figure) increases in the in-plane direction as the coolant temperature increases, and the water content of the CCM in the cell decreases. By contrast, when the coolant inlet flow rate is decreased, the coolant is less able to carry away the heat produced by the CCM. Because of this, the coolant temperature decreases, and the in-plane water content of the CCM increases close to the coolant outlet (Segment position 14 in the figure).

4.3. Effect of An Gas Inlet Flow Rate

Figure 9 shows simulation results with change in the An gas inlet flow rate.

Change in the An gas inlet flow rate produces almost no change in the distribution of the in-plane water content of the CCM. However, a consideration of detailed values indicates that when the An gas inlet flow rate is increased, the amount of water carried away from the CCM by the An gas on the An inlet side (Segment position 15 in the figure) increases, and the in-plane water content of the CCM decreases; however, because the amount of water carried

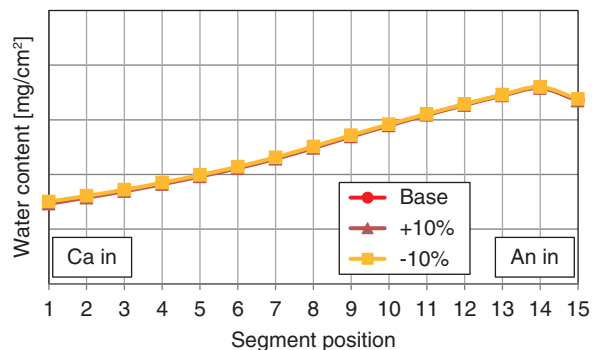


Fig. 8 Simulation results with change in coolant inlet flow rate

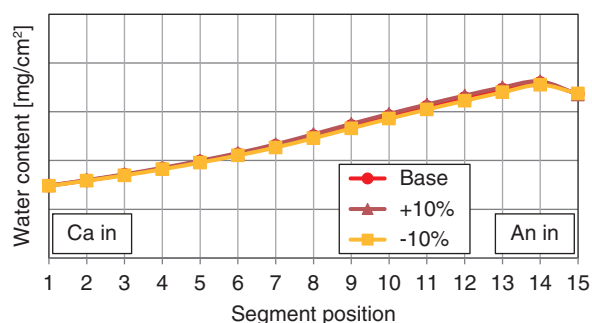


Fig. 9 Simulation results with change in An gas inlet flow rate

away by the An gas decreases downstream of the An gas (Segment positions 1 to 14 in the figure), the in-plane water content of the CCM increases. When the An gas inlet flow rate is reduced, the amount of water carried away from the CCM by the An gas decreases, and the in-plane water content of the CCM on the An inlet side (Segment position 15 in the figure) increases, but because the amount of water carried away by the An gas increases in the downstream section of the An gas flow path (Segment positions 1 to 14 in the figure), the in-plane water content of the CCM decreases.

4.4. Effect of Ca Gas Inlet Flow Rate

Figure 10 shows simulation results with changes in the Ca gas inlet flow rate.

Change in the Ca gas inlet flow rate produces little change in the in-plane water content of the CCM at the Ca gas inlet (Segment position 1 in the figure). However, when the Ca gas inlet flow rate is increased, the maximum in-plane water content of the CCM in the cells (Segment position 14 in the figure) decreases, and when the Ca gas inlet flow rate is reduced, the maximum in-plane water content of the CCM (Segment position 14 in the figure) increases. This is due to the fact that the amount of water carried away increases when the Ca gas inlet flow rate increases, and decreases when the Ca gas inlet flow rate decreases.

4.5. Effect of An Gas Inlet Pressure

Figure 11 shows simulation results with changes in An gas inlet pressure.

Change in the An gas inlet pressure produces almost no effect. However, a consideration of detailed values indicates that, based on the principle of the movement of water in the CCM (Fig. 4), when the An gas inlet pressure increases, the water vapor partial pressure of the An gas increases, and the difference between the Ca and An gas water vapor partial pressures decreases. As a result, the amount of movement of water produced at the Ca to the An decreases, and the in-plane water content of the CCM at all the segment positions

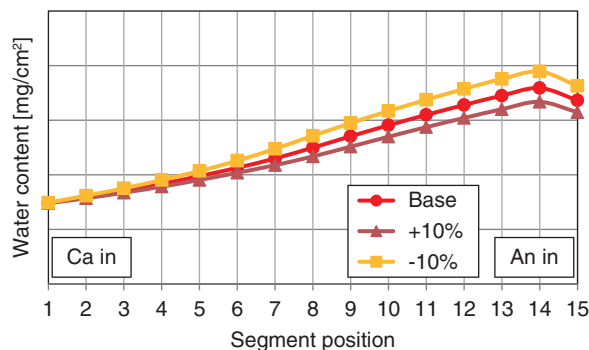


Fig. 10 Simulation results with change in Ca gas inlet flow rate

decreases. Because the An gas water vapor partial pressure decreases when the An gas inlet pressure is reduced, the difference between the Ca and An water vapor partial pressures increases, and the amount of movement of water produced at the Ca to the An increases. As a result, the in-plane water content of the CCM in all segment positions also increases.

4.6. Effect of Ca Gas Inlet Pressure

Figure 12 shows simulation results with changes in Ca gas inlet pressure.

The results show that change in this parameter significantly changes the in-plane distribution of the water content of the CCM. As in the case of the principle of movement of water in the CCM discussed in Section 4.5., when the Ca gas inlet pressure increases, the difference between the An and Ca gas water vapor partial pressure increases, and the amount of movement of water produced at the Ca to the An increases, increasing the in-plane water content of the CCM. Because the Ca gas water vapor partial pressure decreases when the Ca gas inlet pressure is reduced, the difference between the An and Ca gas water vapor partial pressures decreases, and the amount of movement of water produced at the Ca to the An decreases, resulting in a decrease in the in-plane water content of the CCM.

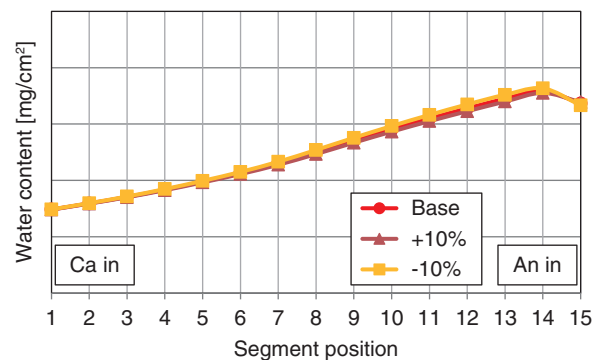


Fig. 11 Simulation results with change in An gas inlet pressure

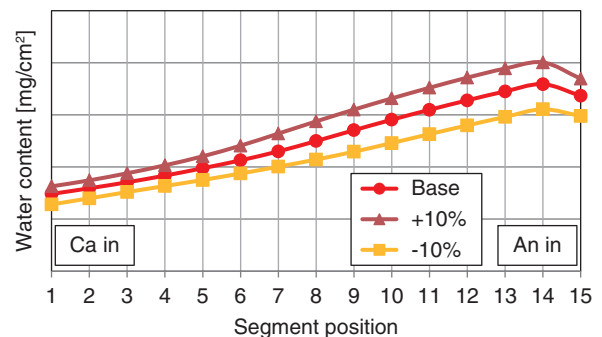


Fig. 12 Simulation results with change in Ca gas inlet pressure

4.7. Sensitivity to Operating Condition Parameters

As indicated in the Introduction, it is important to maintain the in-plane distribution of the water content of the CCM in the cells within the appropriate range. Based on the results shown in Figs. 7 to 12, Fig. 13(a) shows the rate of change in maximum and minimum in-plane water content of the CCM when each parameter is increased by 10%, while Fig. 13(b) shows the rate of change for 10% reductions in the parameters.

The graphs in both Fig. 13(a) and (b) show that the rate of change is greatest in relation to the coolant inlet temperature. The maximum and minimum in-plane water content each change by more than 20% in response to change in this parameter. The next greatest rate of change was in response to the Ca gas inlet pressure, with the minimum in-plane water content of the CCM changing by between around 5 to 10% and the maximum in-plane water content of the CCM changing by around 10% in response to change in this parameter. The third greatest rate of change was in response to the Ca gas inlet flow rate. While there was minimal change in the minimum in-plane water content

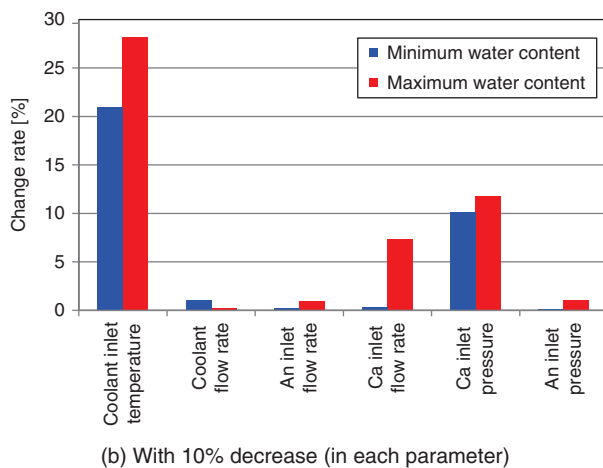
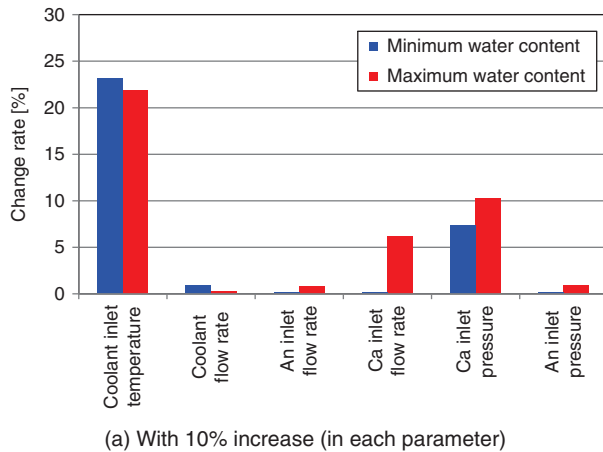


Fig. 13 Rate of change of maximum and minimum water content for individual operating condition

of the CCM in relation to this parameter, the maximum in-plane water content of the CCM displayed a rate of change of around 5%.

These results as a whole indicated that it would be possible to control the minimum in-plane water content of the CCM in the cells by means of the coolant inlet temperature or the Ca gas inlet pressure. The results further indicated that it would be possible to control the maximum in-plane water content in the CCM by using the coolant inlet temperature or the Ca gas inlet pressure as the main parameter and the Ca gas inlet flow rate as a secondary parameter. By this means, it would be possible to control the in-plane distribution of water content in the CCM.

5. Application in Stack

Figure 14 shows the results for in-plane distribution of CCM water content when the results of the studies discussed above were applied to operating conditions for the 2016 model FCV stack. “Upper limit” and “Lower limit” in the figure respectively show the upper limit and lower limit values for in-plane CCM water content necessary for maintaining the durability of the stack.

In the in-plane water content distribution in the CCM for conventional operating conditions (shown as the circles in the figure), the upper limit value was exceeded, meaning that it would not be possible to ensure the durability of the stack.

By contrast, in the case of the operating conditions determined by simulation with consideration of in-plane water content distribution in the CCM (shown as the squares in the figure), increasing the coolant inlet temperature has reduced the overall in-plane water content of the CCM, and increasing the Ca gas inlet flow rate has reduced the maximum in-plane water content of the CCM, making it possible to maintain water content within the upper and lower limit values.

The diamonds in the figure show results for the in-plane distribution of CCM water content in a cell in a stack

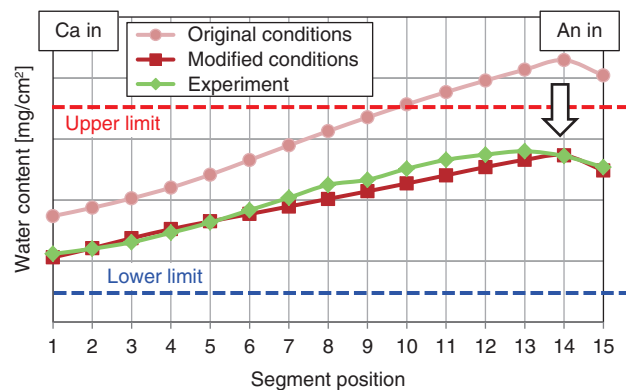


Fig. 14 Original and enhanced water content distribution

measured in measurement tests. A comparison of simulation results (the squares in the figure) and figures from stack measurement (the diamonds in the figure) show that the figures display a good match, reconfirming the judgment that the simulation realized an adequate level of accuracy.

Figure 15(a) shows a schematic image of indexes that express the in-plane distribution of CCM water content in the cells.

Two indexes have been established: the difference between the maximum value and the average value for in-plane CCM water content ("Maximum-average" in the figure), and the difference between the average value and the minimum value for in-plane CCM water content ("Average-minimum" in the figure). In both cases, the in-plane distribution of CCM water content is more desirable the lower the figure.

Figure 15(b) shows the in-plane distribution of CCM water content in terms of these indexes before and after the modification of the operating conditions.

Following the modification of the operating conditions, the difference between the maximum value and the average value for in-plane CCM water content ("Maximum-average" in the figure) was reduced by 39%, and the difference between the average value and the minimum value for in-plane CCM water content ("Average-minimum" in the

figure) was reduced by 31%. It was possible to reduce variations in the distribution of water content in the CCM at the same time as maintaining the in-plane distribution within the upper and lower limits⁽⁴⁾.

In addition, the use of the developed simulation made it possible to determine operating conditions in several days that would previously have necessitated several weeks using tests.

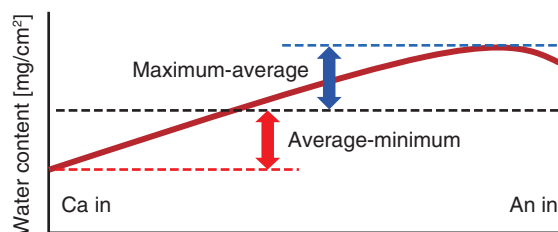
6. Conclusion

A simulation of the in-plane distribution of the water content of CCM in cells was developed. The following results were obtained from the use of the simulation.

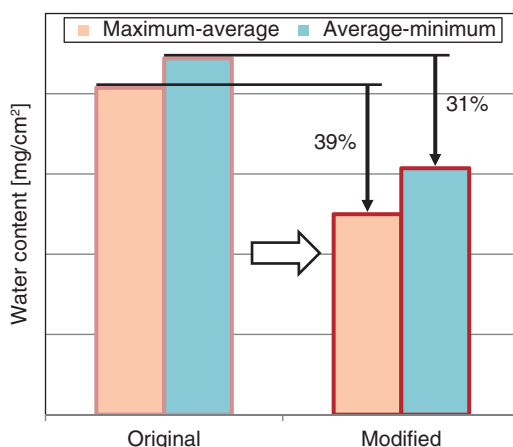
- (1) A simulation was conducted for the stack configuration employed in the 2016 model FCV, and the results of measurements using a sensor system and simulation results for the in-plane distribution of the water content of the CCM were compared. The results indicated that the simulation achieved a predictive accuracy of within 5.0%.
- (2) An advance study of the effect of operating condition parameters on the in-plane distribution of the water content of the CCM was conducted. The results of this study indicated that the minimum in-plane water content of the CCM could be controlled by means of the coolant inlet temperature or the Ca gas inlet pressure. The study also showed that the maximum in-plane water content of the CCM could be controlled by employing the coolant inlet temperature or the Ca gas inlet pressure as the main parameter and the Ca gas inlet flow rate as a secondary parameter. In this way, it would be possible to control the in-plane distribution of the water content of the CCM.
- (3) Using the developed simulation and a coolant inlet temperature, Ca gas inlet pressure, and Ca gas inlet flow rate helping to enable control of the in-plane distribution of water content in the CCM made it possible to maintain the in-plane distribution within the upper and lower limit values and reduced variation in the distribution. It was also possible to apply accurate operating conditions in the stack in a period of several days.

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(a) Schematic image of maximum-average and average-minimum



(b) Maximum-average and average-minimum water content change for original and modified operating conditions

Fig. 15 Enhancement of water content distribution

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■ Author ■



Kenji NAGUMO



Masajiro INOUE