Abstract: The purpose of this study is to understand the impact of the thickness of Nafion membrane, which is a typical polymer electrolyte membrane (PEM) in Polymer Electrolyte Membrane Fuel Cells (PEMFCs), and relative humidity of supply gas on the distributions of H\textsubscript{2}, O\textsubscript{2}, H\textsubscript{2}O concentration and current density on the interface between a Nafion membrane and anode catalyst layer or the interface between a Nafion membrane and cathode catalyst layer. The effect of the initial temperature of the cell (T\textsubscript{ini}) is also investigated by the numerical simulation using the 3D model by COMSOL Multiphysics. As a result, the current density decreases along with the gas flow through the gas channel irrespective of the Nafion membrane thickness and T\textsubscript{ini}, which can be explained by the concentration distribution of H\textsubscript{2} and O\textsubscript{2} consumed by electrochemical reaction. The molar concentration of H\textsubscript{2}O decreases when the thickness of Nafion membrane increases, irrespective of T\textsubscript{ini} and the relative humidity of the supply gas. The current density increases with the increase in relative humidity of the supply gas, irrespective of the Nafion membrane thickness and T\textsubscript{ini}. This study recommends that a thinner Nafion membrane with well-humidified supply gas would promote high power generation at the target temperature of 363 K and 373 K.

Keywords: PEMFC; higher temperature operation than usual; numerical simulation; thickness of Nafion membrane; relative humidity of supply gas; mass and current density distribution

1. Introduction

The polymer electrolyte membrane fuel cell (PEMFC) is one of the promising fuel cell technologies which can use H\textsubscript{2} as a fuel for co-generation system and vehicles. Recently, it has been conceived that H\textsubscript{2} could be one procedure to realize the target of zero CO\textsubscript{2} emissions by 2050 in Japan. Therefore, it is important to develop the efficient PEMFC system by 2050. It is important to develop the efficient PEMFC system as well as green H\textsubscript{2} production in order to achieve the net target, i.e., a virtually zero CO\textsubscript{2} emission in Japan by 2050. According to the Japanese New Energy and Industry Technology Development Organization (NEDO) road map 2017 [1], a PEMFC system is required to be operated at 363 K and 373 K for stationary and mobility applications, respectively, from 2020 to 2025. However, the normal PEMFC, which uses a Nafion membrane, is usually operated within a lower temperature range, between 333 K and 353 K [2,3]. If PEMFC is operated at a higher temperature than usual, the following advantages can be obtained: (1) promoting electrochemical kinetics in both electrodes, (2) reducing the cooling system for automobile applications due to an increase in the temperature difference between the PEMFC stack and coolant, and (3) endurance enhancement to CO in lower quality reformed H\textsubscript{2} [4]. However, operating the PEMFC system at a higher temperature would present challenges, including: (1) degradation of Nafion membranes; (2) catalyst elution; (3) uneven distributions of gas
flow, pressure, temperature, voltage and current in a cell of PEMFC. It is necessary to solve them in order to commercialize the PEMFC system operated at a higher temperature [5]. In addition, it is also believed that the temperature distribution influences the phase change of H$_2$O and can influence the performance of the polymer electrolyte membrane (PEM), fuel and oxidant flows in gas diffusion layer (GDL) and catalyst layer at high temperatures. Consequently, it is necessary to analyze the heat and mass transfer mechanism in a cell of PEMFC in order to improve the power generation performance and achieve a longer operation time.

According to the literature on high-temperature PEMFC operated over 373 K, newly developed membranes which can be used at a high temperature include polybenzimidazole-based membrane [6] and bulky N-heterocyclic group functionalized poly (terphenyl piperidinium) membrane [7]. Regarding the development of new electrode, polytetrafluoroethylene (PTFE) binder dispersion [8] and 3D numerical simulation for the optimization of electrode thickness [9] have been reported. In addition, the optimization of the flow channel of a gas separator [10] and multi-objective optimization of operating conditions [11] are popular topics being studied. Mass transport phenomena in a cell such as distributions of H$_2$, O$_2$, and H$_2$O concentration are also being investigated [12–14]. The temperature distribution on the back of the separator and the interface between PEM and the cathode catalyst layer has been investigated experimentally [15,16] and numerically [17,18], respectively, by the authors. Though the current density and temperature distribution were studied experimentally [19] and numerically [20] at the same time, there have been no reports investigating the distribution of H$_2$, O$_2$, H$_2$O concentration and current density on the interface between PEM and the anode catalyst layer, where the H$_2$ oxidation reaction occurs or the interface between PEM and the cathode catalyst layer where the O$_2$ reduction reaction occurs. In addition, the previous numerical research [12–14] used the contour figure qualitatively. It is not enough to understand the mechanism of electrochemical reaction deciding the power generation performance of PEMFC. It is important to understand the characteristics on the interface between PEM and the anode catalyst layer where the H$_2$ oxidation reaction occurs and the interface between PEM and the cathode catalyst layer where the O$_2$ reduction reaction occurs quantitatively. Therefore, the analysis of these distributions is important to understand the electrochemical reaction and power generation characteristics of PEMFC. Therefore, the purpose of this study is to clarify the distributions of H$_2$, O$_2$, H$_2$O concentration and the current density on the interface between the Nafion membrane, which is used as a typical PEM, and the anode catalyst layer or the interface between the Nafion membrane and cathode catalyst layer under a higher-temperature operation condition than usual. Some new membranes, e.g., polybenzimidazole-based membrane [6], have recently been developed for the high-temperature operation of PEMFC. However, it is easy to apply and commercialize the PEMFC system if the Nafion membrane can be used at a high temperature such as 363 K and 373 K. Numerical simulation using a 3D model by multi-physics simulation software COMSOL Multiphysics has also been carried out to achieve the aim of this study. In addition, the impact of the Nafion membrane thickness on these distributions has also been investigated. When the thinner Nafion membrane is used, lower Ohmic resistance as well as a higher proton flux ratio and back diffusion can be obtained [21–23]. Therefore, it is also important to investigate the effect of the Nafion membrane thickness on the distributions of H$_2$, O$_2$, and H$_2$O concentration and current density on the interface between the Nafion membrane and anode catalyst layer or the interface between the Nafion membrane and cathode catalyst layer.

2. Numerical Modeling
2.1. Model Description and Governing Equations

This study has conducted the numerical analysis using a 3D model by multi-physics simulation software COMSOL Multiphysics. It has the simulation code for PEMFC composed of a continuity equation, the Brinkmann equation, considering the momentum transfer; the Maxwell–Stefan equation, considering the diffusion transfer; and the Butler–
Volmer equation, considering the electrochemical reaction. This simulation code has been validated well by many previous studies [12,24–26].

The continuity equation considering the gas species in porous media such as the catalyst layer, micro porous layer (MPL), and GDL as well as in the gas channel is expressed as follows:

\[
\frac{\partial}{\partial t} (\varepsilon_p \rho) + \nabla \cdot (\rho \vec{u}) = Q_m
\]

(1)

where \(\varepsilon_p\) is the porosity ( ), \(\rho\) is the density (kg/m\(^3\)), \(\vec{u}\) is the velocity vector (m/s), \(Q_m\) is the mass source term (kg/(m\(^2\) s)), and \(t\) is the time (s). The relationship between the pressure and gas flow velocity, which is solved in porous media such as the catalyst layer, MPL, and GDL, as well as in the gas channel, can be expressed by the following Brinkmann equation:

\[
-\nabla p + \nabla \cdot \left[ \frac{\rho}{\varepsilon_p} \left( \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla) \vec{u} \right) \right] = \left( \kappa^{-1} \mu + \frac{Q_m}{\varepsilon_p} \right) \vec{u} + \vec{F}
\]

(2)

where \(p\) is the pressure (Pa), \(\mu\) is the viscosity (Pa s), \(\vec{I}\) is the unit vector( ), \(\kappa\) is the permeability (m\(^2\)), and \(\vec{F}\) is the force vector (kg/(m\(^2\) s\(^2\))), e.g., gravity.

Mass transfer considering the diffusion, ion transfer, and convection transfer can be expressed by the following Maxwell–Stefan equation:

\[
\vec{N}_i = -D_i \nabla C_i - z_i u_{m,i} FC_i \nabla \phi_l + C_i \vec{u} = \vec{J}_i + C_i \vec{u}
\]

(3)

\[
\frac{\partial C_i}{\partial t} + \nabla \cdot \vec{N}_i = R_{i,tot}
\]

(4)

where \(\vec{N}_i\) is the vector of the molar flow rate on the interface between PEM and electrode (mol/(m\(^2\) s)), \(D_i\) is the diffusion coefficient (m\(^2\)/s), \(C_i\) is the concentration of ion \(i\) (mol/m\(^3\)), \(z_i\) is the valence of ion ( ), \(u_{m,i}\) is the mobility of ion \(i\) ((s mol)/kg), \(F\) is the Faraday constant (C/mol), \(\phi_l\) is the potential of liquid [27] (V), \(\vec{J}_i\) is the molar flow rate of the convection transfer (mol/(m\(^2\) s)), and \(R_{i,tot}\) is the reaction rate of species (mol/(m\(^2\) s)).

The electrochemical reaction is calculated following Butler–Volmer equation:

\[
i = i_0 \left\{ \exp \left( \frac{a_c F \eta}{RT} \right) - \exp \left( -\frac{a_c F \eta}{RT} \right) \right\}
\]

(5)

\[
\eta = \phi_s - \phi_l - E_{eq}
\]

(6)

where \(i\) is the current density (A/m\(^2\)), \(i_0\) is the exchange current density (A/m\(^2\)), \(a_c\) is the charge transfer coefficient at anode ( ), \(\eta\) is the activation over-potential [27] (V), \(R\) is the gas constant (J/(mol K)), \(T\) is the temperature (K), \(a_c\) is the charge transfer coefficient at cathode ( ), \(\phi_s\) is the electrical potential of solid [27] (V), \(E_{eq}\) is the equilibrium electric potential [27] (V).

Figure 1 illustrates 3D model of single cell of PEMFC. This structure follows the commercialized single cell used in the previous experimental study [16]. In this model, the outside of the roof of gas separator at anode and cathode sides are omitted. This single cell has a gas separator having a serpentine flow channel which consists not only of five gas channels with the width of 1.0 mm and depth of 1.0 mm but also a rib with a width of 1.0 mm. Table 1 lists the geometrical parameters of the 3D model used in this study. Nafion 115, Nafion NRE-212, and Nafion NRE-211, whose thicknesses are 127 µm, 51 µm, and 25 µm, respectively, have been investigated to assess the impact of the thickness of the Nafion membrane on the distributions of H\(_2\), O\(_2\), and H\(_2\)O concentrations and the current density. Physical parameters and operation conditions for numerical simulation in this study are listed in Tables 2 and 3, respectively. \(T_{\text{ini}}\) is changed by 353 K, 363 K, and 373 K. This study has conducted the numerical simulation at \(T_{\text{ini}} = 353\) K showing
the characteristics at a standard operation temperature to compare the characteristics at a higher temperature. The relative humidity of supply gas at the anode and cathode is changed by 40%RH and 80%RH, respectively. The flow rate of supply gas is set at the stoichiometric ratio of 1.5, where the volume flow rate of supply gas at the anode and cathode is 0.210 NL/min and 0.105 NL/min, respectively. The stoichiometric ratio of 1.0 for the flow rate of supply gas can be defined by Equation (7).

\[
C_{H_2} = \frac{I}{z_{H_2} F}
\]  

(7)

where \(C_{H_2}\) is the molar flow rate of consumed \(H_2\) (mol/s), \(I\) is the loaded current (A) and \(z_{H_2}\) is the electrons moles exchanged in the reaction (=2) ( ). \(C_{H_2}\) is the molar flow rate corresponding to the stoichiometric ratio of 1.0. The \(C_O_2\) is the molar flow rate of consumed \(O_2\) (mol/s), which is a half of \(C_{H_2}\) (which can be defined by Equation (8)).

\[
H_2 + \frac{1}{2} O_2 = H_2O
\]  

(8)

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**Table 1. Geometrical parameters of 3D model for numerical simulation [22,28–31].**

| Components       | Size                                                                 | Characteristics                                                                 |
|------------------|----------------------------------------------------------------------|--------------------------------------------------------------------------------|
| PEM              | 50.0 mm × 50.0 mm × 0.127 mm (for Nafion 115), 0.051 mm (for Nafion NRE-212) and 0.025 mm (for Nafion NRE-211) | Nafion 115, Nafion NRE-212, Nafion NRE-211 (manufactured by Du Pont Corp.)       |
| Catalyst layer   | 50.0 mm × 50.0 mm × 0.01 mm                                          | Pt/C (Pt: 20 wt%)                                                              |
| MPL              | 50.0 mm × 50.0 mm × 0.003 mm                                          | PTFE + carbon black                                                            |
| GDL              | 50.0 mm × 50.0 mm × 0.19 mm                                          | TGP-H-060 (manufactured by Toray Corp.)                                         |
| Rib:             | 5.0 mm × 50.0 mm × 2.00 mm (thickness of gas channel and rib: 1.0 mm, thickness of gas channel: 1.0 mm) | Carbon graphite, serpentine                                                      |
| Gas separator    |                                                                      |                                                                                |

**Table 2. Physical parameters for numerical simulation in this study.**

| Parameter Name                           | Value                                                   |
|------------------------------------------|---------------------------------------------------------|
| Density of \(H_2\) (kg/m³)              | 7.10 × 10⁻² (353 K), 6.89 × 10⁻² (363 K), 6.69 × 10⁻² (373 K) [32] |
| Density of \(O_2\) (kg/m³)              | 1.11 (353 K), 1.08 (363 K), 1.05 (373 K) [32]           |
| Density of \(H_2O\) (kg/m³)             | 2.95 × 10⁻¹ (353 K), 4.26 × 10⁻¹ (363 K), 6.01 × 10⁻¹ (373 K) [32] |
| Viscosity of \(H_2\) (Pa s)             | 9.96 × 10⁻⁶ (353 K), 1.02 × 10⁻⁵ (363 K), 1.03 × 10⁻⁵ (373 K) [32] |
| Viscosity of \(O_2\) (Pa s)             | 2.35 × 10⁻⁵ (353 K), 2.40 × 10⁻⁵ (363 K), 2.45 × 10⁻⁵ (373 K) [32] |
| Viscosity of \(H_2O\) (Pa s)            | 1.16 × 10⁻⁵ (353 K), 1.19 × 10⁻⁵ (363 K), 1.23 × 10⁻⁵ (373 K) [32] |
| Binary diffusion coefficient between \(H_2\) and \(H_2O\) (m²/s) | 9.27 × 10⁻⁵ [33]                                      |
| Binary diffusion coefficient between \(O_2\) and \(H_2O\) (m²/s) | 3.57 × 10⁻⁵ [33]                                      |
### Table 2. Cont.

| Parameter Name                             | Value                                                   |
|--------------------------------------------|---------------------------------------------------------|
| Porosity of catalyst layer ()              | 0.78 \[17,22,29–31\]                                   |
| Permeability of catalyst layer (m²)        | 8.69 × 10⁻¹² \[17,22,29–31\]                           |
| Porosity of MPL ()                         | 0.60 \[17,22,29–31\]                                   |
| Permeability of MPL (m²)                   | 1.00 × 10⁻¹³ \[17,22,29–31\]                           |
| Porosity of GDL ()                         | 0.78 \[17,22,29–31\]                                   |
| Permeability of GDL (m²)                   | 8.69 × 10⁻¹² \[17,22,29–31\]                           |
| Conductivity of Nafion series membrane (S/m) | 10 \[34\]              |
| Conductivity of catalyst layer (S/m)       | 53 \[35\]                                              |
| Conductivity of MPL (S/m)                  | 1000 \[36\]                                            |
| Conductivity of GDL (S/m)                  | 1250 \[37\]                                            |
| Anode reference equilibrium potential (V)  | 0                                                       |
| Cathode reference equilibrium potential (V)| 1.229                                                   |
| Anode reference exchange current density (A/m²) | 1000 \[38\]              |
| Cathode reference exchange current density (A/m²) | 1 \[38\]              |
| Anode charge transfer coefficient ()        | 0.5 \[39\]                                             |
| Cathode charge transfer coefficient ()      | 0.5 \[40\]                                             |

### Table 3. Operation conditions for numerical simulation in this study.

| Each Condition                        | Value                                      |
|---------------------------------------|--------------------------------------------|
| The initial temperature of cell \(T_{\text{ini}}\) (K) | 353, 363, 373                     |
| Cell voltage (V)                     | Experimental data are used \[16,17\]         |
| Supply gas condition                 |                                            |
| Gas type                             | Anode \(\text{H}_2\)                        |
|                                      | Cathode \(\text{O}_2\)                      |
| Temperature of supply gas at inlet (K) | 353, 363, 373                     |
| Relative humidity of supply gas (%RH) | 40, 80                                    |
| Pressure of supply gas at inlet (MPa) | 0.4                                       |
| Flow rate of supply gas at inlet [NL/min] (Stoichiometric ratio ()) | 0.210 (1.5) 0.105 (1.5) |

In this study, \(\text{O}_2\) is adopted as the cathode gas. In the near future, it can be expected that \(\text{H}_2\) will be produced from renewable energy via \(\text{H}_2\text{O}\) electrolyzer mainly in order to realize a zero-\(\text{CO}_2\)-emission society. After the production of \(\text{H}_2\) by \(\text{H}_2\text{O}\) electrolysis, \(\text{O}_2\) is also produced as a by-product. This study suggests that not only \(\text{H}_2\) but also \(\text{O}_2\) produced from \(\text{H}_2\text{O}\) electrolysis are used for PEMFC. The total system consisting of renewable energy, \(\text{H}_2\text{O}\) electrolyzer, and PEMFC can be operated effectively by using \(\text{O}_2\). In addition, if we use \(\text{O}_2\) as a cathode gas, we can obtain a higher current density on the interface between PEM and the catalyst layer, especially under the rib, compared to the case using air \[28\]. This is due to the decrease in over-potential by the increase in the concentration of \(\text{O}_2\). Therefore, it can be expected that higher power generation performance is obtained by using \(\text{O}_2\). They are the reasons why this study adopts pure \(\text{O}_2\) instead of air.

### 2.2. Model Assumption

This study considers the following assumptions:

(i) The distributions of the inlet gas flow rate at the anode side and cathode side are uniform, respectively.

(ii) The pressure of the outlet of the gas channel is the atmospheric pressure.

(iii) No slip on the gas channel wall excluding the inlet and the outlet of the gas channel is considered.

(iv) The cell voltage obtained by the power generation experiment is set at the cathode electrode and the earth ground is set at the anode electrode. The in-plane distribution of cell voltage at the cathode electrode is uniform.

(v) Reactant gases are treated as an ideal gas and incompressible Newton fluid.
(vi) H₂O is treated as a vapor.
(vii) The cell temperature is uniform and the outside boundary of the 3D model is set at $T_{\text{ini}}$.
(viii) The effective porosity and the permeability of the porous media are isotropic. The conductivity in the porous media is also isotropic.

The impact of the Nafion membrane thickness, $T_{\text{ini}}$, and relative humidity of the supply gas on the distributions of the molar concentration of H₂, O₂, and H₂O and current density on the interface between the Nafion membrane and anode catalyst layer or the interface between the Nafion membrane and cathode catalyst layer has been investigated, considering the above-described equations and parameters as well as the assumptions.

3. Results and Discussion

3.1. In-Plane Distribution of Mass and Current Density on the Interface between Nafion Membrane and Anode Catalyst Layer or the Interface between Nafion Membrane and Cathode Catalyst Layer

Figure 2 shows the comparison of the in-plane molar concentration distribution of H₂ on the interface between the Nafion membrane and anode catalyst layer with a different Nafion membrane and $T_{\text{ini}}$ for the relative humidity of the supply gas of the anode of 80%RH and cathode of 80%RH (A80%RH&C80%RH). Figures 3–5 show the comparisons of in-plane molar concentration distribution of O₂ and H₂O and in-plane distribution of the current density on the interfaces, respectively.

![Comparison of in-plane molar concentration distribution of H₂ on the interface between Nafion membrane and anode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (A80%RH&C80%RH; left: 353 K, center: 363 K, right: 373 K).](image-url)
Figure 3. Comparison of in-plane molar concentration distribution of $\text{O}_2$ on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (A80%RH&C80%RH; left: 353 K, center: 363 K, right: 373 K).

Figure 4. Comparison of in-plane molar concentration distribution of $\text{H}_2\text{O}$ on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (A80%RH&C80%RH; left: 353 K, center: 363 K, right: 373 K).

Figure 4. Cont.
According to Figure 2, it is found that the molar concentration of H₂ decreases along with the gas flow through the gas channel irrespective of the Nafion membrane thickness and $T_{ini}$ due to the uniform consumption rate of H₂ caused by the high permeability through porous media [12]. In addition, it is known that the molar concentration of H₂ decreases more with the increase in $T_{ini}$ irrespective of the Nafion membrane thickness. The molar concentration is defined by dividing the molar quantity of gas species by its volume. Since the gas volume increases when $T_{ini}$ increases, the molar concentration of H₂ would decrease with the increase in $T_{ini}$.

**Figure 4.** Comparison of in-plane molar concentration distribution of H₂O on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A80%RH&C80%RH; left: 353 K, center: 363 K, right: 373 K).

**Figure 5.** Comparison of in-plane distribution of current density on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A80%RH&C80%RH; left: 353 K, center: 363 K, right: 373 K).
It is seen from Figure 3 that the molar concentration of O$_2$ decreases along with the gas flow through the gas channel irrespective of the Nafion membrane thickness and $T_{ini}$. It is found that the O$_2$ reduction reaction progresses along with the gas flow through the gas channel. According to Figure 3, the amount of O$_2$ consumption from the inlet to the outlet is the largest at $T_{ini} = 353$ K, irrespective of the Nafion membrane thickness. The kinetics of the catalyst are faster with the increase in temperature, while the relative humidity influences the performance of the O$_2$ reduction reaction occurring on the ionomer in the cathode catalyst layer [40]. There is the optimum H$_2$O saturation of ionomer in the cathode catalyst layer. On the other hand, the proton conductivity of the Nafion membrane is influenced by the temperature and relative humidity. According to the literature [41,42], the proton conductivity of the Nafion membrane increases with the increase in temperature as well as the increase in relative humidity. Since the saturation pressure of H$_2$O vapor increases with the temperature exponentially [43], it is easy to dehydrate the Nafion membrane at $T_{ini} = 373$ K compared to $T_{ini} = 353$ K, resulting in the proton conductivity of the Nafion membrane decreasing at $T_{ini} = 373$ K. If the proton conductivity of the Nafion membrane decreases, the performance of the O$_2$ reduction reaction drops due to a lack of proton. In addition, since the hydration of the Nafion membrane is not enough at $T_{ini} = 373$ K, the high O$_2$ partial pressure is needed to progress the O$_2$ reduction reaction [43]. Consequently, it is thought that the amount of O$_2$ consumption decreases at $T_{ini} = 373$ K. The proton conductivity of the Nafion membrane increases when the temperature increases, even at a temperature above 373 K [40]. However, we have to consider the degradation of the Nafion membrane under the dehydrated condition [44]. Therefore, it is a challenging issue to control the humidification of the Nafion membrane at a temperature above 373 K.

It is seen from Figure 4 that the molar concentration of H$_2$O increases along with the gas flow through the gas channel, irrespective of the Nafion membrane thickness and $T_{ini}$. This result matches with Figure 3 from the viewpoint of the O$_2$ reduction reaction which produces H$_2$O at the cathode. According to Figure 4, the amount of H$_2$O produced from the inlet to the outlet is the largest at $T_{ini} = 353$ K, irrespective of the Nafion membrane thickness. The reason why this result is obtained can be explained by the above discussion in Figure 3.

According to Figure 5, the current density decreases along with the gas flow through the gas channel, irrespective of the Nafion membrane thickness and $T_{ini}$. The molar concentrations of H$_2$ and O$_2$ are the highest at the inlet, respectively, and they decrease along with the gas flow through the gas channel, as shown in Figures 2 and 3, indicating that the electrochemical reaction progresses along with the gas flow through the gas channel. In addition, the current density is the largest at $T_{ini} = 353$ K among different $T_{ini}$. Moreover, it is evident that the current density decreases with an increase in the thickness of the Nafion membrane. The authors of this study argue that we have to consider the kinetics of catalyst as well as proton conductivity of Nafion membrane for the discussion of this phenomena. The kinetics of the catalyst are faster with an increase in temperature, while the relative humidity influences the performance of the O$_2$ reduction reaction occurring on the ionomer in the cathode catalyst layer [40]. There is an optimum H$_2$O saturation of ionomer in the cathode catalyst layer. On the other hand, the proton conductivity of the Nafion membrane is influenced by the temperature and relative humidity. According to the literature [41,42], the proton conductivity of the Nafion membrane increases with the increase in temperature as well as the increase in relative humidity. Since the saturation pressure of H$_2$O vapor exponentially increases with the temperature [43], it is easy to dehydrate the Nafion membrane at $T_{ini} = 373$ K compared to $T_{ini} = 353$ K, resulting in the proton conductivity of the Nafion membrane decreasing at $T_{ini} = 373$ K. If the proton conductivity of the Nafion membrane decreases, the performance of the O$_2$ reduction reaction drops due to a lack of proton. In addition, since the hydration of the Nafion membrane is not enough at $T_{ini} = 373$ K, the high O$_2$ partial pressure is needed to progress the O$_2$ reduction reaction [43]. As a result, it is thought that the amount of O$_2$ consumption decreases at $T_{ini} = 373$ K. The concentration over-potential increases with a decrease in the
O₂ consumption, resulting in the current density decreasing as \( T_{\text{ini}} = 373 \text{ K} \). In addition, the ohmic loss due to the proton conductivity of the Nafion membrane increases when thickness of the Nafion membrane increases [45]. Consequently, the current density in the case with Nafion 115 at \( T_{\text{ini}} = 373 \text{ K} \) is smaller compared to the other conditions.

3.2. Quantitative Evaluation along with the Gas Flow through the Gas Channel on Mass and Current Density on the Interface between Nafion Membrane and Cathode Catalyst Layer

In order to investigate the effect of the Nafion membrane and \( T_{\text{ini}} \) on the molar concentration distribution of H₂O and the distribution of the current density, which can quantitatively indicate the performance of the electrochemical reaction in PEMFC, this study selected the analysis points of A to K, as shown in Figure 6. The average value on the cross sectional area of the interface between the Nafion membrane and cathode catalyst layer at each point, which covers both part under gas channel and that under rib, has been calculated.

![Figure 6](image)

**Figure 6.** Analysis points for the quantitative evaluation along with the gas flow through the gas channel.

Figures 7–10 show comparisons of molar concentration of H₂O along with the gas flow through the gas channel with different relative humidities of supply gas among different Nafion membranes and \( T_{\text{ini}} \), respectively. Tables 4–8 list the values shown in Figures 7–10 to compare quantitatively, respectively. According to Figures 7–10, the molar concentration of H₂O increases along with the gas flow through the gas channel, irrespective of the Nafion membrane thickness, \( T_{\text{ini}} \), and relative humidity of the supply gas. Since the O₂ reduction reaction progresses along with the gas flow through the gas channel, H₂O, which is a product of the O₂ reduction reaction, increases as expected. In addition, the molar concentration of H₂O is the highest at \( T_{\text{ini}} = 353 \text{ K} \) among different \( T_{\text{ini}} \), irrespective of the Nafion membrane thickness and relative humidity of the supply gas. As described above, \( T_{\text{ini}} = 353 \text{ K} \) is the most humidified condition among different \( T_{\text{ini}} \), irrespective of the Nafion membrane thickness and relative humidity of the supply gas, resulting in the proton conductivity of the Nafion membrane and the performance of O₂ reduction reaction at cathode being the best. Consequently, the molar concentration of H₂O is the highest at \( T_{\text{ini}} = 353 \text{ K} \) among the investigated \( T_{\text{ini}} \). Regarding the impact of the relative humidity of the supply gas, the molar concentration of H₂O increases when the relative humidity of the supply gas increases, irrespective of the Nafion membrane thickness and
The largest molar concentration of H₂O is confirmed with A80%RH&C80%RH, while the smallest molar concentration of H₂O is confirmed with an anode of 40%RH and cathode of 40%RH (A40%RH&C40%RH). Generally speaking, the increase in humidification enhances the performance of PEMFC, which promotes the proton conductivity of the Nafion membrane, while the decrease in proton conductivity of Nafion membrane causes higher ohmic losses [22,46]. As discussed above, with the low relative humidity, the ionomer in the cathode catalyst layer is hard to be saturated by the H₂O migrated through the Nafion membrane from the anode to the cathode, deciding the performance of the O₂ reduction reaction at the cathode, which produces H₂O [40]. Consequently, it is obtained that the molar concentration of H₂O is the largest with A80%RH&C80%RH, while it is the smallest with A40%RH&C40%RH. Comparing the thickness of the Nafion membrane, the molar concentration of H₂O decreases when the thickness of the Nafion membrane increases, irrespective of \( T_{\text{ini}} \) and the relative humidity of the supply gas. In particular, the molar concentration of H₂O for Nafion 115 is much smaller than that for the other Nafion membranes. H₂O flux of PEM as well as the conductivity of the Nafion membrane is promoted when the thickness of the Nafion membrane decreases, particularly below 50 µm [21,47,48], which corresponds to Nafion NRE-212 and Nafion NRE-211 in this study. In addition, the ohmic loss due to proton conductivity of Nafion membrane decreases when the thickness of the Nafion membrane [49] decreases. Since the proton conductivity and H₂O flux of Nafion membrane are low for a thick Nafion membrane, the performance of O₂ reduction reaction, which produces H₂O at the cathode, declines. Consequently, it is thought that the molar concentration of H₂O decreases when the thickness of the Nafion membrane increases, especially for Nafion 115. Summarizing the above discussion, the largest molar concentration of H₂O, which is 15.1 mol/m³, is obtained at the position K in the case of using Nafion NRE-211 at \( T_{\text{ini}} = 353 \) K with A80%RH&C80%RH according to Tables 4–8. In this study, it is assumed that H₂O is treated as a vapor. To validate this assumption, the saturation of H₂O calculated by the numerical simulation of this study is confirmed. The saturation of H₂O is defined by dividing a partial pressure of H₂O vapor by a saturation H₂O vapor pressure. Figure 11 shows a comparison of the saturation of H₂O along with the gas flow through the gas channel among different Nafion membranes at \( T_{\text{ini}} = 353 \) K with A80%RH&C80%RH. It is seen from Figure 11 that the saturation of H₂O is lower than 1.0 even the case of Nafion NRE-211 near the outlet. When the saturation of H₂O is lower than 1.0, it means that H₂O exists as a vapor. The molar concentration of H₂O is the largest in the case of Nafion NRE-211 under the condition that \( T_{\text{ini}} = 353 \) K with A80%RH&C80%RH among conditions investigated in this study. Therefore, it can be argued that H₂O can be treated as a vapor under the conditions investigated in this study.

Table 4. Comparison of molar concentration of H₂O along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and \( T_{\text{ini}} \) (unit: mol/m³; A80%RH&C80%RH).

| A   | B   | C   | D   | E   | F   | G   | H   | I   | J   | K   |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 353 K | | | | | | | | | | |
| 115 | 0.721 | 2.07 | 3.14 | 4.27 | 5.22 | 6.25 | 7.12 | 8.05 | 8.85 | 9.61 | 10.3 |
| 212 | 0.793 | 2.87 | 4.46 | 6.10 | 7.46 | 8.88 | 10.1 | 11.3 | 12.4 | 13.3 | 14.2 |
| 211 | 0.813 | 3.08 | 4.81 | 6.57 | 8.02 | 9.53 | 10.8 | 12.1 | 13.2 | 14.2 | 15.1 |
| 363 K | | | | | | | | | | |
| 115 | 0.955 | 2.23 | 3.23 | 4.28 | 5.18 | 6.13 | 6.96 | 7.84 | 8.59 | 9.31 | 9.93 |
| 212 | 0.979 | 2.49 | 3.67 | 4.89 | 5.92 | 7.02 | 7.96 | 8.94 | 9.78 | 10.6 | 11.3 |
| 211 | 0.981 | 2.51 | 3.70 | 4.93 | 5.97 | 7.08 | 8.02 | 9.01 | 9.85 | 10.6 | 11.3 |
| 373 K | | | | | | | | | | |
| 115 | 1.17 | 1.58 | 1.91 | 2.27 | 2.58 | 2.93 | 3.23 | 3.56 | 3.85 | 4.13 | 4.38 |
| 212 | 1.19 | 1.77 | 2.24 | 2.74 | 3.17 | 3.65 | 4.06 | 4.52 | 4.91 | 5.29 | 5.63 |
| 211 | 1.20 | 1.86 | 2.39 | 2.95 | 3.45 | 3.98 | 4.45 | 4.95 | 5.40 | 5.82 | 6.20 |
irrespective of $T_{\text{ini}}$ and the relative humidity of the supply gas. In particular, the molar concentration of $\text{H}_2\text{O}$ for Nafion 115 is much smaller than that for the other Nafion membranes. $\text{H}_2\text{O}$ flux of PEM as well as the conductivity of the Nafion membrane is promoted when the thickness of the Nafion membrane decreases, particularly below 50 $\mu$m [21,47,48], which corresponds to Nafion NRE-212 and Nafion NRE-211 in this study. In addition, the ohmic loss due to proton conductivity of Nafion membrane decreases when the thickness of Nafion membrane [49] decreases. Since the proton conductivity and $\text{H}_2\text{O}$ flux of Nafion membrane are low for a thick Nafion membrane, the performance of $\text{O}_2$ reduction reaction, which produces $\text{H}_2\text{O}$ at the cathode, declines. Consequently, it is thought that the molar concentration of $\text{H}_2\text{O}$ decreases when the thickness of the Nafion membrane increases, especially for Nafion 115.

Summarizing the above discussion, the largest molar concentration of $\text{H}_2\text{O}$, which is 15.1 mol/m$^3$, is obtained at the position K in the case of using Nafion NRE-211 at $T_{\text{ini}} = 353$ K with A80%RH&C80%RH according to Tables 4–8. In this study, it is assumed that $\text{H}_2\text{O}$ is treated as a vapor. To validate this assumption, the saturation of $\text{H}_2\text{O}$ calculated by the numerical simulation of this study is confirmed. The saturation of $\text{H}_2\text{O}$ is defined by dividing a partial pressure of $\text{H}_2\text{O}$ vapor by a saturation $\text{H}_2\text{O}$ vapor pressure.

Figure 7. Comparison of molar concentration of $\text{H}_2\text{O}$ along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (A80%RH&C80%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Table 5. Comparison of molar concentration of $\text{H}_2\text{O}$ along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (unit: mol/m$^3$; A80%RH&C40%RH).

|     | A   | B   | C   | D   | E   | F   | G   | H   | I    | J    | K    |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|------|------|------|------|
| 353 K |
| 115 | 0.425 | 1.68 | 2.68 | 3.73 | 4.62 | 5.59 | 6.41 | 7.30 | 8.05 | 8.77 | 9.40 |
| 212 | 0.477 | 2.28 | 3.68 | 5.13 | 6.35 | 7.63 | 8.71 | 9.85 | 10.8 | 11.7 | 12.5 |
| 211 | 0.492 | 2.44 | 3.96 | 5.52 | 6.81 | 8.17 | 9.31 | 10.5 | 11.5 | 12.5 | 13.3 |
| 363 K |
| 115 | 0.527 | 1.46 | 2.21 | 3.01 | 3.69 | 4.44 | 5.07 | 5.77 | 6.36 | 6.94 | 7.44 |
| 212 | 0.557 | 1.80 | 2.78 | 3.81 | 4.68 | 5.63 | 6.43 | 7.29 | 8.02 | 8.72 | 9.33 |
| 211 | 0.575 | 2.00 | 3.12 | 4.28 | 5.27 | 6.32 | 7.21 | 8.16 | 8.97 | 9.73 | 10.4 |
| 373 K |
| 115 | 0.644 | 1.02 | 1.33 | 1.66 | 1.95 | 2.28 | 2.56 | 2.87 | 3.14 | 3.40 | 3.64 |
| 212 | 0.664 | 1.25 | 1.72 | 2.23 | 2.67 | 3.16 | 3.57 | 4.03 | 4.43 | 4.82 | 5.16 |
| 211 | 0.675 | 1.37 | 1.92 | 2.52 | 3.15 | 3.60 | 4.08 | 4.61 | 5.07 | 5.51 | 5.91 |
Figure 7. Comparison of molar concentration of H$_2$O along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A80%RH&C80%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Figure 8. Comparison of molar concentration of H$_2$O along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A80%RH&C40%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Table 6. Comparison of molar concentration of H$_2$O along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (unit: mol/m$^3$; A40%RH&C80%RH).

|     | A  | B  | C  | D  | E  | F  | G  | H  | I  | J  | K  |
|-----|----|----|----|----|----|----|----|----|----|----|----|
| 353 K |    |    |    |    |    |    |    |    |    |    |    |
| 115 | 0.706 | 1.91 | 2.86 | 3.87 | 4.73 | 5.67 | 6.46 | 7.31 | 8.05 | 8.74 | 9.36 |
| 212 | 0.758 | 2.49 | 3.83 | 5.23 | 6.40 | 7.64 | 8.69 | 9.79 | 10.7 | 11.6 | 12.4 |
| 211 | 0.773 | 2.65 | 4.10 | 5.50 | 6.85 | 8.17 | 9.27 | 10.4 | 11.4 | 12.3 | 13.1 |
| 363 K |    |    |    |    |    |    |    |    |    |    |    |
| 115 | 0.915 | 1.80 | 2.51 | 3.26 | 3.91 | 4.62 | 5.23 | 5.89 | 6.46 | 7.01 | 7.49 |
| 212 | 0.945 | 2.12 | 3.05 | 4.03 | 4.86 | 5.76 | 6.53 | 7.35 | 8.06 | 8.73 | 9.32 |
| 211 | 0.954 | 2.21 | 3.21 | 4.25 | 5.13 | 6.08 | 6.89 | 7.76 | 8.50 | 9.20 | 9.82 |
| 373 K |    |    |    |    |    |    |    |    |    |    |    |
| 115 | 1.16 | 1.41 | 1.62 | 1.85 | 2.05 | 2.27 | 2.46 | 2.68 | 2.87 | 3.05 | 3.22 |
| 212 | 1.18 | 1.70 | 2.11 | 2.55 | 2.94 | 3.37 | 3.74 | 4.15 | 4.50 | 4.85 | 5.15 |
| 211 | 1.19 | 1.80 | 2.28 | 2.81 | 3.26 | 3.76 | 4.19 | 4.66 | 5.07 | 5.46 | 5.82 |
Figure 9. Comparison of molar concentration of H$_2$O along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A40%RH&C80%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Table 7. Comparison of molar concentration of H$_2$O along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (unit: mol/m$^3$; A40%RH&C40%RH).

|     | A   | B   | C   | D   | E   | F   | G   | H   | I   | J   | K   |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 353 K |     |     |     |     |     |     |     |     |     |     |     |
| 115  | 0.368 | 1.01 | 1.54 | 2.11 | 2.59 | 3.13 | 3.60 | 4.11 | 4.56 | 4.99 | 5.37 |
| 212  | 0.438 | 1.83 | 2.93 | 4.09 | 5.06 | 6.12 | 7.00 | 7.96 | 8.77 | 9.53 | 10.2 |
| 211  | 0.453 | 2.00 | 3.22 | 4.50 | 5.56 | 6.71 | 7.67 | 8.70 | 9.57 | 10.4 | 11.1 |
| 363 K |     |     |     |     |     |     |     |     |     |     |     |
| 115  | 0.464 | 0.727 | 0.944 | 1.18 | 1.39 | 1.62 | 1.82 | 2.04 | 2.24 | 2.43 | 2.60 |
| 212  | 0.529 | 1.09 | 2.25 | 3.06 | 3.75 | 4.51 | 5.16 | 5.86 | 6.47 | 7.05 | 7.55 |
| 211  | 0.539 | 1.59 | 2.43 | 3.32 | 4.08 | 4.90 | 5.61 | 6.37 | 7.02 | 7.64 | 8.18 |
| 373 K |     |     |     |     |     |     |     |     |     |     |     |
| 115  | 0.615 | 0.693 | 0.757 | 0.828 | 0.890 | 0.960 | 1.02 | 1.09 | 1.15 | 1.21 | 1.26 |
| 212  | 0.650 | 1.09 | 1.45 | 1.84 | 2.18 | 2.56 | 2.88 | 3.24 | 3.55 | 3.86 | 4.13 |
| 211  | 0.657 | 1.16 | 1.57 | 2.01 | 2.40 | 2.82 | 3.19 | 3.60 | 3.90 | 4.29 | 4.59 |
Figure 10. Comparison of molar concentration of H$_2$O along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A40%RH&C40%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Table 8. Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (unit: A/mm$^2$; A80%H&C80%RH).

|       | A  | B   | C   | D   | E   | F   | G   | H   | I   | J   | K   |
|-------|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 353 K | 115| 0.197| 0.194| 0.192| 0.190| 0.187| 0.185| 0.182| 0.180| 0.178| 0.175| 0.173 |
|       | 212| 0.307| 0.300| 0.293| 0.289| 0.281| 0.277| 0.270| 0.265| 0.259| 0.254| 0.250 |
|       | 211| 0.336| 0.328| 0.320| 0.314| 0.305| 0.299| 0.291| 0.286| 0.277| 0.272| 0.266 |
| 363 K | 115| 0.197| 0.193| 0.191| 0.189| 0.186| 0.184| 0.181| 0.179| 0.176| 0.174| 0.172 |
|       | 212| 0.234| 0.230| 0.226| 0.223| 0.218| 0.215| 0.211| 0.208| 0.204| 0.200| 0.198 |
|       | 211| 0.237| 0.232| 0.228| 0.225| 0.220| 0.217| 0.212| 0.209| 0.205| 0.201| 0.198 |
| 373 K | 115| 0.066| 0.065| 0.065| 0.065| 0.064| 0.064| 0.063| 0.063| 0.062| 0.062| 0.062 |
|       | 212| 0.094| 0.093| 0.092| 0.092| 0.091| 0.090| 0.089| 0.088| 0.088| 0.087| 0.086 |
|       | 211| 0.107| 0.106| 0.105| 0.104| 0.103| 0.102| 0.101| 0.100| 0.099| 0.098| 0.097 |
As discussed above, a higher relative humidity increases the current density [40]. Since the actual relative humidity of gas in the cell is the highest at \( T_{\text{ini}} = 353 \text{ K} \) among the investigated \( T_{\text{ini}} \), resulting in the current density being the highest at \( T_{\text{ini}} = 353 \text{ K} \). As to the impact of the relative humidity of the supply gas, the current density increases when the relative humidity of the supply gas increases, irrespective of the Nafion membrane thickness and \( T_{\text{ini}} \). The largest current density is confirmed with A80%RH&C80%RH, while the smallest current density is confirmed with A40%RH&C40%RH. The performance of PEMFC is enhanced with the increase in humidification by the promotion of proton conductivity of Nafion membrane, resulting in lower ohmic losses [22,46]. Therefore, it was revealed that the current density is the largest with A80%RH&C80%RH, while it is the smallest with A40%RH&C40%RH. According to Figures 12–15, it is known that the current density increases when the thickness of the Nafion membrane decreases. The thinner Nafion membrane provides lower ohmic losses, indicating that the proton transfers with a shorter distance to reach the cathode and the \( \text{H}_2\text{O} \) produced in the cathode catalyst layer reaches the anode faster [49]. In addition, \( \text{H}_2\text{O} \) flux of Nafion membrane as well as the conductivity of the Nafion membrane is promoted when the thickness of the Nafion membrane decreases, especially below 50 \( \mu \text{m} \) [21,47,48], which corresponds to Nafion NRE-212 and Nafion NRE-211 in this study. Therefore, it is revealed that the current density increases when the thickness of Nafion membrane decreases, especially for Nafion
Nafion NRE-212 and Nafion NRE-211. Summarizing the above discussion, the largest current density of 0.336 A/mm² is obtained at the position A in the case of using Nafion NRE-211 at $T_{ini} = 353$ K with A80%RH&C80%RH according to Tables 8–12.

![Graph](image)

**Figure 12.** Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A80%RH&C80%RH): (a): 353 K, (b): 363 K, (c): 373 K.

**Table 9.** Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (unit: A/mm²; A80%H&C40%RH).

|     | A | B | C | D | E | F | G | H | I | J | K |
|-----|---|---|---|---|---|---|---|---|---|---|---|
| 353 K |   |   |   |   |   |   |   |   |   |   |   |
|     | 0.175 | 0.172 | 0.170 | 0.168 | 0.166 | 0.164 | 0.162 | 0.160 | 0.158 | 0.156 | 0.154 |
| 212 | 0.253 | 0.248 | 0.244 | 0.240 | 0.235 | 0.231 | 0.226 | 0.223 | 0.218 | 0.214 | 0.211 |
| 211 | 0.276 | 0.270 | 0.246 | 0.260 | 0.253 | 0.249 | 0.243 | 0.239 | 0.233 | 0.229 | 0.225 |
| 363 K |   |   |   |   |   |   |   |   |   |   |   |
|     | 0.135 | 0.133 | 0.132 | 0.131 | 0.129 | 0.128 | 0.127 | 0.125 | 0.124 | 0.123 | 0.122 |
| 212 | 0.180 | 0.178 | 0.175 | 0.173 | 0.170 | 0.168 | 0.165 | 0.163 | 0.160 | 0.158 | 0.156 |
| 211 | 0.208 | 0.204 | 0.201 | 0.198 | 0.194 | 0.191 | 0.187 | 0.185 | 0.181 | 0.178 | 0.176 |
| 373 K |   |   |   |   |   |   |   |   |   |   |   |
|     | 0.057 | 0.056 | 0.056 | 0.055 | 0.055 | 0.055 | 0.054 | 0.054 | 0.054 | 0.053 | 0.053 |
| 212 | 0.088 | 0.087 | 0.086 | 0.086 | 0.085 | 0.084 | 0.083 | 0.083 | 0.082 | 0.081 | 0.081 |
| 211 | 0.104 | 0.103 | 0.102 | 0.101 | 0.100 | 0.099 | 0.098 | 0.097 | 0.096 | 0.095 | 0.094 |
Figure 12. Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (A80%RH&C80%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Figure 13. Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (A80%RH&C40%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Table 10. Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membranes and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (unit: A/mm$^2$; A40%RH&C80%RH).

|     | A  | B  | C  | D  | E  | F  | G  | H  | I  | J  | K  |
|-----|----|----|----|----|----|----|----|----|----|----|----|
| 353 K |
| 115 | 0.175 | 0.173 | 0.171 | 0.169 | 0.167 | 0.165 | 0.163 | 0.161 | 0.159 | 0.157 | 0.155 |
| 212 | 0.254 | 0.249 | 0.245 | 0.241 | 0.236 | 0.233 | 0.228 | 0.225 | 0.220 | 0.216 | 0.213 |
| 211 | 0.277 | 0.270 | 0.265 | 0.261 | 0.255 | 0.251 | 0.245 | 0.241 | 0.235 | 0.231 | 0.227 |
| 363 K |
| 115 | 0.136 | 0.134 | 0.133 | 0.132 | 0.130 | 0.132 | 0.128 | 0.127 | 0.125 | 0.124 | 0.123 |
| 212 | 0.181 | 0.178 | 0.176 | 0.174 | 0.171 | 0.170 | 0.167 | 0.165 | 0.162 | 0.160 | 0.159 |
| 211 | 0.195 | 0.192 | 0.189 | 0.187 | 0.183 | 0.181 | 0.178 | 0.176 | 0.173 | 0.170 | 0.168 |
| 373 K |
| 115 | 0.041 | 0.041 | 0.041 | 0.041 | 0.040 | 0.040 | 0.040 | 0.040 | 0.039 | 0.039 | 0.039 |
| 212 | 0.083 | 0.082 | 0.082 | 0.081 | 0.080 | 0.080 | 0.079 | 0.079 | 0.078 | 0.077 | 0.077 |
| 211 | 0.098 | 0.097 | 0.096 | 0.096 | 0.095 | 0.094 | 0.093 | 0.092 | 0.091 | 0.091 | 0.090 |
Figure 14. Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (A40%H&C80%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Table 11. Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{ini}$ (unit: A/mm$^2$; A40%RH&C40%RH).

|     | A | B | C | D | E | F | G | H | I | J | K |
|-----|---|---|---|---|---|---|---|---|---|---|---|
| 353 K |   |   |   |   |   |   |   |   |   |   |   |
| 115 | 0.089 | 0.088 | 0.087 | 0.086 | 0.086 | 0.085 | 0.085 | 0.084 | 0.083 | 0.083 |   |
| 212 | 0.195 | 0.191 | 0.189 | 0.186 | 0.183 | 0.181 | 0.178 | 0.176 | 0.173 | 0.170 | 0.168 |
| 211 | 0.218 | 0.214 | 0.210 | 0.207 | 0.203 | 0.200 | 0.196 | 0.194 | 0.190 | 0.190 | 0.184 |
| 363 K |   |   |   |   |   |   |   |   |   |   |   |
| 115 | 0.038 | 0.037 | 0.037 | 0.037 | 0.037 | 0.037 | 0.037 | 0.036 | 0.036 | 0.036 | 0.036 |
| 212 | 0.138 | 0.136 | 0.135 | 0.133 | 0.132 | 0.130 | 0.129 | 0.128 | 0.126 | 0.125 | 0.123 |
| 211 | 0.153 | 0.151 | 0.149 | 0.147 | 0.145 | 0.143 | 0.141 | 0.140 | 0.138 | 0.136 | 0.135 |
| 373 K |   |   |   |   |   |   |   |   |   |   |   |
| 115 | 0.012 | 0.012 | 0.012 | 0.011 | 0.011 | 0.011 | 0.011 | 0.011 | 0.011 | 0.011 | 0.011 |
| 212 | 0.066 | 0.066 | 0.065 | 0.065 | 0.064 | 0.064 | 0.064 | 0.063 | 0.062 | 0.062 | 0.062 |
| 211 | 0.076 | 0.075 | 0.075 | 0.074 | 0.074 | 0.073 | 0.072 | 0.072 | 0.072 | 0.071 | 0.070 |
Figure 15. Comparison of current density along with the gas flow through the gas channel on the interface between Nafion membrane and cathode catalyst layer among different Nafion membranes and $T_{\text{ini}}$ (A40%RH&C40%RH; (a): 353 K, (b): 363 K, (c): 373 K).

Considering the above results and discussion, this study can suggest that the thinner Nafion membrane under well-humidified conditions is desirable in order to obtain a higher power generation performance operated at higher temperatures such as 363 K and 373 K. In addition, the uniform distribution of the current density along with the gas flow through the gas channel is obtained by using the thinner Nafion membrane according to Figures 12–15. However, the value of current density is still low at high temperatures such as 363 K and 373 K, even using a thinner Nafion membrane. According to Tables 8–12, the current density is 0.237 A/mm² and 0.107 A/mm² at the position A in the case of using Nafion NRE-211 at $T_{\text{ini}} = 363$ K and 373 K with A80%RH&C80%RH, respectively. To increase the current density in the case of a thinner Nafion membrane, this study suggests the optimization of the catalyst layer [9], MPL [45], and gas channel flow of the gas separator [10], not only in order to control the mass and heat transfer phenomena but also to improve the electrochemical reaction. We have to consider the degradation of the Nafion membrane if we operate PEMFC at a higher temperature than usual. This study conducted the experimental investigation using a thin Nafion membrane at higher temperatures such as 363 K and 373 K [16]. In this experiment, it was confirmed that the thin Nafion membrane kept the performance over the power generation operation of 200 h. However, it is necessary to investigate the characteristics of a thin Nafion membrane by operating for a longer time, e.g., 90,000 h (=10 years), which is the target time according to the NEDO road map 2010 in Japan for the practical application of a PEMFC system.
Table 12. Comparison of relationship between current of 20 A and voltage among investigated conditions in this study.

|                  | A80%RH&C80%RH | A80%RH&C40%RH | A40%RH&C80%RH | A40%RH&C40%RH |
|------------------|----------------|---------------|---------------|---------------|
| $T_{ini}$ [K]    | 353            | 363           | 373           | 353           | 363           | 373           | 353           | 363           | 373           | 353           | 363           | 373           |
| Nafion type      | 115            | 212           | 211           | 115           | 212           | 211           | 115           | 212           | 211           | 115           | 212           | 211           |
| Voltage [V]      | 0.581          | 0.631         | 0.636         | 0.601         | 0.611         | 0.606         | 0.461         | 0.501         | 0.516         | 0.561         | 0.601         | 0.606         |
|                  | 0.541          | 0.571         | 0.576         | 0.441         | 0.481         | 0.501         |               |               |               |               |               |               |
|                  | 0.541          | 0.571         | 0.576         | 0.441         | 0.481         | 0.501         |               |               |               |               |               |               |
|                  | 0.461          | 0.561         | 0.571         | 0.371         | 0.531         | 0.541         | 0.291         | 0.451         | 0.466         |               |               |               |

4. Conclusions

The numerical simulation using a 3D model by multi-physics simulation software COMSOL Multiphysics has been conducted in order to investigate distributions of $\text{H}_2$, $\text{O}_2$, and $\text{H}_2\text{O}$ concentration and current density on the interface between Nafion the membrane and anode catalyst layer, and the interface between the Nafion membrane and cathode catalyst layer when operated at higher temperatures. The impacts of the Nafion membrane thickness, $T_{ini}$, and relative humidity of the supply gas on these distributions have been investigated. The conclusions have been drawn as follows:

(i). The molar concentration of $\text{H}_2$ and $\text{O}_2$ decreases along with the gas flow through the gas channel, irrespective of the Nafion membrane thickness and $T_{ini}$.

(ii). The $\text{O}_2$ consumption in the fuel cell is the largest at $T_{ini} = 353$ K, irrespective of Nafion membrane thickness.

(iii). The molar concentration of $\text{H}_2\text{O}$ increases along with the gas flow through the gas channel, irrespective of the Nafion membrane thickness and $T_{ini}$, which can be explained by the $\text{O}_2$ reduction reaction at cathode.

(iv). The current density decreases along with the gas flow through the gas channel, irrespective of Nafion membrane thickness and $T_{ini}$. The current density is the largest at $T_{ini} = 353$ K, irrespective of the Nafion membrane thickness.

(v). The molar concentration of $\text{H}_2\text{O}$ increases when the relative humidity of the supply gas increases, irrespective of the Nafion membrane thickness and $T_{ini}$. The molar concentration of $\text{H}_2\text{O}$ for Nafion 115, whose thickness is 127 µm, is much smaller than that for the other thin Nafion membranes.

(vi). The molar concentration of $\text{H}_2\text{O}$ generally decreases when the thickness of the Nafion membrane increases. The molar concentration of $\text{H}_2\text{O}$ for Nafion 115, whose thickness is 127 µm, is much smaller than that for the other thin Nafion membranes.

(vii). It is revealed that the largest molar concentration of $\text{H}_2\text{O}$ is 15.1 mol/m$^3$ near the outlet in the case of using Nafion NRE-211 at $T_{ini} = 353$ K with A80%RH&C80%RH among the conditions investigated in this study.

(viii). The current density is the highest at $T_{ini} = 353$ K.

(ix). The current density increases when the relative humidity of the supply gas increases, irrespective of the Nafion membrane thickness and $T_{ini}$, which indicates that the
power generation performance is enhanced with the increase in relative humidity due to the promotion of proton conductivity of the Nafion membrane.

(x). The current density increases with the decrease in the Nafion membrane thickness since the H₂O flux of the Nafion membrane as well as the conductivity of the Nafion membrane is promoted with the thinner Nafion membrane.

(xi). It is revealed that the largest current density is 0.336 A/mm² near the inlet in the case of using Nafion NRE-211 at Tini = 353 K with A80%RH&C80%RH among the conditions investigated in this study.

(xii). This study reveals that the thinner Nafion membrane under well-humidified conditions is more desirable to obtain a higher power generation performance at higher temperatures, i.e., 363 K and 373 K. Thinner Nafion membranes can provide a uniform distribution of current density as well.

(xiii). Since the current density at high temperatures of 363 K and 373 K, which are 0.237 A/mm² and 0.107 A/mm², respectively, is still low, even using Nafion NRE-211 with A80%RH&C80%RH, this study suggests the optimization of catalyst layer, MPL, and gas channel flow of gas separator in order to control the mass and heat transfer phenomena as well as to improve the electrochemical reaction.

Author Contributions: Conceptualization and writing—original draft preparation; A.N.; numerical analysis and investigation; K.T.; data curation; Y.K. and S.I.; writing—review and editing; E.H. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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