

## ORIGINAL RESEARCH ARTICLE

# Impact of separator thickness on relationship between temperature distribution and mass & current density distribution in single HT-PEMFC

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## ABSTRACT

Considering the application of the polymer electrolyte membrane fuel cell (PEMFC), the separator thickness plays a significant role to determine the weight, the volume and the costs of the PEMFC. In addition, thermal management, i.e., temperature distribution is also important for the PEMFC system to obtain higher performance. However, there were few reports investigating the relation between the temperature profile and the power generation characteristics e.g., current density distribution of PEMFC operated at higher temperature (HT-PEMFC). This paper aims to study the impact of separator thickness on the temperature profile and the current density profile of HT-PEMFC. The impact of separator thickness on the gases i.e., H<sub>2</sub>, O<sub>2</sub> profile of HT-PEMFC numerically was also studied using CFD software COMSOL Multiphysics in the paper. In the study, the operating temperature and the relative humidity (RH) of supply gas were varied with the separator thickness of 2.0 mm, 1.5 mm and 1.0 mm, respectively. The study revealed that the optimum thickness was 2.0 mm to realize higher power generation of HT-PEMFC. The heat capacity of the separator thickness of 2.0 mm was the biggest among the separators investigated in this study, resulting that the dry up of PEM and catalyst layer was lower compared to the thinner separator thickness. It was also clarified the effects of separator thickness of profile gases, e.g., O<sub>2</sub>, H<sub>2</sub>O, and current density profile became larger under the higher temperature and the lower RH conditions.

**Keywords:** HT-PEMFC; numerical simulation; separator thickness; coupling phenomena

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## 1. Introduction

The Japanese New Energy and Industry Technology Development Organization (NEDO) that is a Japanese government agency has announced that polymer electrolyte membrane fuel cell (PEMFC) should be worked at higher temperature such as 363 K and 373 K for the application use of stationary and vehicle, respectively, during the period from 2020 to 2025 in road map 2017<sup>[1]</sup>. On the other hand, PEMFC which uses Nafion membrane for a polymer electrolyte membrane (PEM) generally worked below 353 K<sup>[2-4]</sup>. The merits for PEMFC operated at higher temperature (HT-PEMFC) include (i) kinetic improvement of catalyst, (ii) down scale effect of the cooling system for the mobility application thanks to increase in temperature gap between PEMFC stack and coolant, and (iii) enhancement of CO endurance allowing the purity of H<sub>2</sub> production from hydrocarbon such as CH<sub>4</sub><sup>[5]</sup>. On the other hand, the following issues which should be overcome: (i) degradation of PEM because of thermal expansion and shrinkage, (ii) electrode erosion, (iii) uneven profile of gas flow,

gas pressure, temperature, voltage and current density in PEMFC<sup>[6]</sup>. In addition, the uneven profiles of H<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>O, temperature and current density would undermine the power generation characteristics as well as the operation life of PEMFC when operated at higher temperature than usual. However, as to the existing PEMFC, the following problems are considered<sup>[7,8]</sup>; (1) low tolerance of catalyst such as Pt to contaminant such as CO, (2) slow electrochemical kinetics, (3) difficulties in water and thermal management. These problems can be solved under the higher operation temperature conditions (HT-PEMFC).

The effect of thickness of PEM and gas diffusion layer (GDL) as well as micro porous layer (MPL) on the coupling phenomena of HT-PEMFC worked at 363 K and 373 K had been studied experimentally and numerically<sup>[9–11]</sup> by authors. Moreover, the effect of separator thickness on H<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>O and current density distributions had been investigated numerically<sup>[12]</sup> and that on the temperature profile on separator's back surface experimentally<sup>[13]</sup>. According to recent works except for the authors' studies<sup>[12,13]</sup>, the impact of interdigitated flow field of separator on mass transport and electrochemical reaction in HT-PEMFC was investigated by CFD software COMCOL multiphysics<sup>[14]</sup>. Compared the performance of interdigitated flow field with that of parallel flow field, the increase in current density with air stoichiometry in the case of interdigitated flow field was approximately three times as large as that in the case of parallel flow field. On the other hand, the polarization curve in the case of interdigitated flow field was almost same as that in the case of single channel serpentine flow field. Though the relationship between O<sub>2</sub> distribution or pressure distribution and the power generation performance was discussed, that between the temperature distribution and the power generation performance was not investigated. The other numerical study using CFD software COMCOL Multiphysics reported that three different types of cathode-enhanced mass transfer flow fields, i.e., tapered, staggered-blocked and blocked were designed and compared their performances<sup>[15]</sup>. As a result, the tapered flow field was the optimum design for HT-PEMFC due to the superior performance and lower flow resistance. Though the relation between the power generation characteristics and O<sub>2</sub> profile or flow field distribution was discussed, that between the power generation characteristics and the temperature distribution was not studied. Regarding the general PEMFC operated below 353 K, the several flow fields of separator such as a modified parallel flow field<sup>[16]</sup>, a blocked flow field<sup>[17]</sup>, a modified serpentine wave flow field<sup>[18]</sup>, a straight channel with baffled obstacles<sup>[19]</sup> and an ultrathin steel separator whose thickness was 0.1 mm<sup>[20]</sup> were investigated. Foam structure separators consisting of graphene or metal to improve the mass diffusion were investigated and compared to the normal separator<sup>[21,22]</sup>. The porous structure foam can improve the convection and diffusion, and reduce the contact resistance between the flow plate and carbon paper. Since the weight ratio of separator to that of total cell is approximately 80%<sup>[23]</sup>, it is important to optimize the design of separator. Especially, the separator thickness provides a big impact on the weight, volume and cost of cell. In addition, a thermal management is important to realize higher performance for the application usage of PEMFC system<sup>[24]</sup>. However, there are few reports investigating the relation between the temperature profile and the power generation characteristics, e.g., current density profile of HT-PEMFC. Therefore, we aim to reveal the impact of separator thickness on the relationship between the temperature distribution and the current density distribution of HT-PEMFC. We have investigated the impact of separator thickness on the relationship between the temperature profile and the current density profile of HT-PEMFC numerically using CFD software COMSOL Multiphysics. The reason why this study selects COMSOL Multiphysics is as follows; fuel cell improves complex multi-physics coupling problems, including charge transfer, water transport, and heat transfer, etc. In COMSOL, users could select or customize various partial differential equations and combine them to achieve direct coupled multi-physics field analysis easily. As described above, some numerical studies on HT-PEMFC were conducted by COMSOL multiphysics<sup>[14,15]</sup>. Given the characteristics of fuel cells and the advantages of COMSOL software, we chose this software for the simulation work in this paper. The relation between the temperature profile and not only the current density profile but also the mass such as O<sub>2</sub> and H<sub>2</sub>O profiles are also discussed. The separator thickness is changed by 2.0 mm, 1.5 mm and 1.0 mm. The separator thickness of 2.0 mm consists of the saddle thickness = 1.0 mm and the channel height = 1.0 mm. The separator

thickness of 1.5 mm consists of the saddle thickness = 0.5 mm and the channel height = 1.0 mm. The separator thickness of 1.0 mm consists of the saddle thickness = 0.5 mm and the channel height = 0.5 mm. Regarding PEM and GDL, this study adopts Nafion NRE-211 and TGP-H-030, respectively. This selection follows the results obtained by the previous studies conducted by the authors which optimized the thickness of PEM and GDL<sup>[12,13]</sup>. This study changes the operation temperature by 353 K, 363 K and 373 K. This study investigates the characteristics at 353 K, 363 K and 373 K. 353 K is selected as a case of LT-PEMFC for the comparison of the results under higher temperature condition. The higher temperature condition at 363 K and 373 K are selected following the target temperature for the application use of stationary and vehicle, respectively during the period from 2020 to 2025 in the road map of NEDO. Compared to the general operation temperature of HT-PEMFC of 413 K and 473 K, 363 K and 373 K which are investigated in this study are lower. However, the issues such as low tolerance of catalyst such as Pt to CO and SO<sub>2</sub>, difficulties in water and thermal management, and slow electrochemical kinetics can be improved over 373 K according to Zhang et al.<sup>[25]</sup>. Therefore, the 373 K investigated in this study has a point as a case study of HT-PEMFC. This study also examines changing the relative humidity (RH) of supply gases at the anode = 80%RH and cathode = 80%RH (A80%RH-C80%RH), anode = 80%RH and cathode = 40%RH (A80%RH-C40%RH), anode = 40%RH and cathode = 80%RH (A40%RH-C80%RH) and anode = 40%RH and cathode = 40%RH (A40%RH-C40%RH).

## 2. Numerical simulation procedure

### Governing equation

The numerical simulation conducted by a multi-physics software, i.e., COMSOL Multiphysics, ver. 6.1. Fuel cell involves complex multi-physics coupling problems, including charge transfer, mass transfer and heat transfer, etc. In COMSOL, users could select or customize various partial differential equations and combine them to easily achieve direct coupled multi-physics filed analysis. The COMSOL Multiphysics has a simulation function code consisting of Brinkman formula, Maxwell-Stefan formula, Butler-Volmer formula and heat transfer formula considering the heat generated by over-potentials, thermal conduction through each component in the cell and thermal convection via the flow through the channel as well as transferring from the exhaust gas to the ambient air. Some researchers previously carried out the numerical simulation using COMSOL Multiphysics for HT-PEMFC<sup>[4,14,15,25–28]</sup> and achieved good results for the temperature, gases and current density distributions. In addition, the validation was conducted well. Thanks to giving the characteristics of fuel cells and the advantages of COMSOL, the present study adopted it for the numerical simulation of HT-PEMFC. The following governing equations are involved in COMSOL.

Firstly, the continuity equation treating the gas species in porous material in single PEMFC, e.g., catalyst layer, MPL, GDL and the gas channel, can be defined, as following:

$$\frac{\partial}{\partial t}(\varepsilon_p \rho) + \nabla \cdot (\rho \vec{u}) = Q_m \quad (1)$$

where  $\varepsilon_p$  is the porosity of porous material (-),  $\rho$  is the gas density (kg/m<sup>3</sup>),  $\vec{u}$  is the gas velocity vector (m/s),  $Q_m$  is the mass source term balancing this equation (kg/(m<sup>3</sup>·s)) and  $t$  is the time (s).

Brinkman equation considering the relationship between the gas pressure and gas flow velocity which is solved in porous material in single PEMFC, e.g., catalyst layer, MPL, GDL and the gas channel, can be defined, as following:

$$\frac{\rho}{\varepsilon_p} \left( \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla) \frac{\vec{u}}{\varepsilon_p} \right) = -\nabla p + \nabla \cdot \left[ \frac{1}{\varepsilon_p} \left\{ \mu \left( \nabla \vec{u} + (\nabla \vec{u})^T \right) - \frac{2}{3} \mu (\nabla \cdot \vec{u}) \vec{I} \right\} \right] - \left( \kappa^{-1} \mu + \frac{Q_m}{\varepsilon_p^2} \right) \vec{u} + \vec{F} \quad (2)$$

where  $p$  is the gas pressure (Pa),  $\mu$  is the gas viscosity (Pa·s),  $\vec{I}$  is the unit vector (-),  $\kappa$  is the permeability of porous material (m<sup>2</sup>), and  $\vec{F}$  is the force vector (kg/(m<sup>2</sup>·s<sup>2</sup>)) such as a gravity. Maxwell-Stefan equation treating

the mass transfer phenomena, i.e., the diffusion phenomenon, ion transfer phenomenon as well as convection transfer phenomenon can be defined, as following:

$$\vec{N}_i = -D_i \nabla C_i - z_i u_{m,i} F C_i \nabla \phi_l + C_i \vec{u} = \vec{J}_i + C_i \vec{u} \quad (3)$$

$$\frac{\partial C_i}{\partial t} + \nabla \cdot \vec{N}_i = R_{i,tot} \quad (4)$$

where  $\vec{N}_i$  indicates the vector molar flow rate on the interface between PEM and catalyst layer (mol/(m<sup>2</sup>·s)),  $D_i$  indicates the diffusion constant of gas (m<sup>2</sup>/s),  $C_i$  indicates the ion  $i$  concentration (mol/m<sup>3</sup>),  $z_i$  indicates the ion valence (-),  $u_{m,i}$  indicates the ion  $i$  mobility ((s·mol)/kg),  $F$  indicates the faraday constant (C/mol),  $\phi_l$  indicates the electrical potential of liquid material<sup>[28]</sup> (V),  $J_i$  indicates the molar flow rate of the convection transfer phenomenon (mol/(m<sup>2</sup>·s)) and  $R_{i,tot}$  indicates the species' reaction rate (mol/(m<sup>3</sup>·s)).

Butler-Volmer equation treats the electrochemical reaction phenomenon, as following:

$$i = i_0 \left\{ \exp \left( \frac{\alpha_a F \eta}{RT} \right) - \exp \left( \frac{-\alpha_c F \eta}{RT} \right) \right\} \quad (5)$$

$$\eta = \phi_s - \phi_l - E_{eq} \quad (6)$$

where  $i$  is the current density (A/m<sup>2</sup>),  $i_0$  is the exchange current density (A/m<sup>2</sup>),  $\alpha_a$  is the charge transfer coefficient at anode side (-),  $\eta$  is the activation over-potential<sup>[29]</sup> (V),  $R$  is the gas constant (J/(mol·K)),  $T$  is the operating temperature (K),  $\alpha_c$  is the charge transfer constant at the cathode side (-),  $\phi_s$  is the electrical potential of solid material<sup>[29]</sup> (V),  $E_{eq}$  is the equilibrium electric voltage<sup>[29]</sup> (V).

$$\rho C_p u \cdot \nabla T = \nabla \cdot (k \nabla T) + Q_{jh} + \sum_m a_v Q_m + q_0 \quad (7)$$

$$Q_{jh} = -(\vec{i}_s \cdot \nabla \phi_s + \vec{i}_l \cdot \nabla \phi_l) \quad (8)$$

$$Q_m = \left( \eta + T \frac{\partial E_{eq}}{\partial T} \right) i \quad (9)$$

$$q_0 = -h(T_{ext} - T) \quad (10)$$

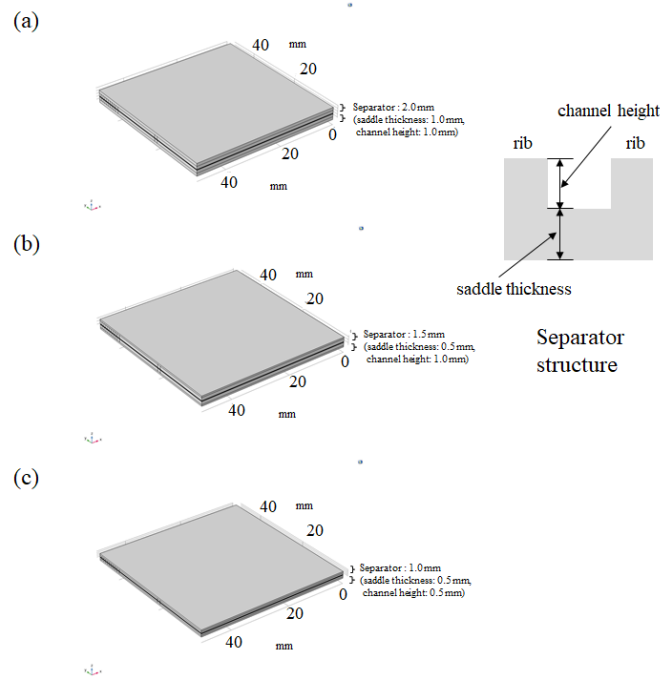
where  $C_p$  indicates the constant pressure specific heat (J/(kg·K)),  $k$  is the thermal conductivity (W/(m·K)),  $a_v$  means the active area ratio (1/m),  $\vec{i}_s$  means the electrode current density vector (A/m<sup>2</sup>),  $\vec{i}_l$  means the electrolyte current density vector (A/m<sup>2</sup>),  $h$  indicates the heat transfer coefficient (W/(m<sup>2</sup>·K)),  $T_{ext}$  indicates the external temperature (K).

The simulation model developed and used in this study was the same as the authors' previous study<sup>[12]</sup>. **Figure 1** illustrates the model for the separator thickness of 2.0 mm, 1.5 mm and 1.0 mm. The structures of these models follow the commercial cell used in the experiments carried out by Nishimura et al.<sup>[11,13]</sup>. The separator has a serpentine flow channel consisting of five gas channels having a gas channel width of 1.0 mm and gas channel width of 1.0 mm. This cell has five gas channels following the structure of the commercial cell<sup>[9,13]</sup>. This separator has the serpentine flow-field as shown in **Figure 2**. **Table 1** shows the geometrical parameters used for the model proposed in this study. **Tables 2** and **3** show physical parameters and operation conditions, respectively. We change the initial operation temperature of a cell ( $T_{ini}$ ) by 353 K, 363 K and 373 K. This study adopts 353 K to compare the characteristics obtained under usual temperature condition with that at higher temperature condition. We also change the RHs of supply gases i.e., A80%RH-C80%RH, A80%RH-C40%RH, A40%RH-C80%RH and A40%RH-C40%RH. We examine the flow rate of supply gas in case of the stoichiometric ratio (s.r.) of 1.5, where the volume flow rate of supply gas at the anode side and the cathode side is equal to 0.210 NL/min and 0.105 NL/min, respectively. The s.r. of 1.0 indicating the flow rate of supply gas can be expressed by Equation (11).

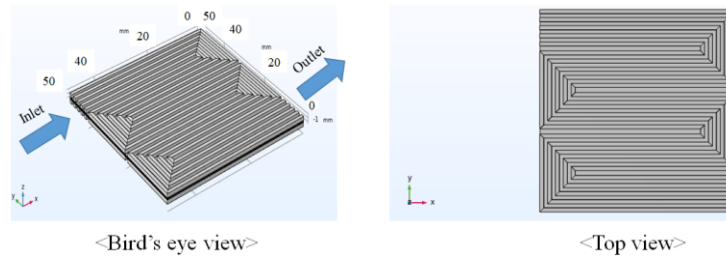
$$C_{H_2} = \frac{I}{Z_{H_2} F} \quad (11)$$

where  $C_{H_2}$  is the molar flow rate of H<sub>2</sub> which is consumed in the electrochemical reaction (mol/s),  $I$  is the

loaded current (A) and  $z_{H_2}$  is the electrons moles which are exchanged in the reaction ( $=2$ ) (-),  $C_{H_2}$  is the molar flow rate for s.r. = 1.0. The  $C_{O_2}$  is the molar flow rate of  $O_2$  which is consumed in the electrochemical reaction (mol/s). The  $C_{O_2}$  is half of  $C_{H_2}$  (refer to Equation (12)).



**Figure 1.** 3D model simulated for single HT-PEMFC ((a): separator thickness of 2.0 mm, (b): separator thickness of 1.5 mm, (c): separator thickness of 1.0 mm).



**Figure 2.** 3D model simulated for single HT-PEMFC which shows the serpentine flow filed structure.

**Table 1.** Geometric parameters for components of model simulating single HT-PEMFC<sup>[10,14,30–33]</sup>.

Components of single cell	Each size[mm]	Specification
PEM	Width: 50.0, length: 50.0, depth: 0.025	Nafion NRE-212 (Manufactured by Du Pont Corp.)
Catalyst layer	Width: 50.0, length: 50.0, depth: 0.01	Pt/C (Weight percentage of Pt: 20)
MPL	Width: 50.0, length: 50.0, depth: 0.003	PTFE + carbon black
GDL	Width: 50.0, length: 50.0, depth: 0.11	TGP-H-030 (Produced by Toray Corp.)
Separator	Width: 75.4, length: 75.4, depth: 2.0 (saddle thickness: 1.0, channel height: 1.0), 1.5 (saddle thickness: 0.5, channel height: 1.0), 1.0 (saddle thickness: 0.5, channel height: 0.5); Width: 50.0, Length: 50.0 (as to gas supply area)	Carbon graphite, serpentine

**Table 2.** Physical parameters for gases, components of cell as well as electrochemical reactions.

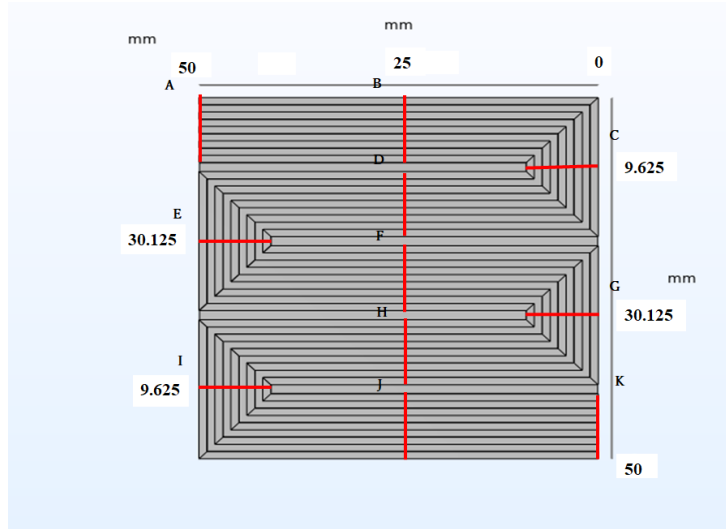
Physical parameters	Values
Gas density (H <sub>2</sub> ) [kg/m <sup>3</sup> ]	$7.10 \times 10^{-2}$ (@ 353 K), $6.89 \times 10^{-2}$ (@ 363 K), $6.69 \times 10^{-2}$ (@ 373 K) <sup>[30]</sup>
Gas density (O <sub>2</sub> ) [kg/m <sup>3</sup> ]	1.11 (@ 353 K), 1.08 (@ 363 K), 1.05 (@ 373 K) <sup>[30]</sup>
Gas density (H <sub>2</sub> O) [kg/m <sup>3</sup> ]	$2.95 \times 10^{-1}$ (@ 353 K), $4.26 \times 10^{-1}$ (@ 363 K), $6.01 \times 10^{-1}$ (@ 373 K) <sup>[30]</sup>
Pressure of supply gas at inlet of cell (absolute based) (MPa)	0.4 <sup>[13]</sup>
Gas viscosity (H <sub>2</sub> ) [Pa·s]	$9.96 \times 10^{-6}$ (@ 353 K), $1.02 \times 10^{-5}$ (@ 363 K), $1.03 \times 10^{-5}$ (@ 373 K) <sup>[30]</sup>
Gas viscosity (O <sub>2</sub> ) [Pa·s]	$2.35 \times 10^{-5}$ (@ 353 K), $2.40 \times 10^{-5}$ (@ 363 K), $2.45 \times 10^{-5}$ (@ 373 K) <sup>[30]</sup>
Gas viscosity (H <sub>2</sub> O) [Pa·s]	$1.16 \times 10^{-5}$ (@ 353 K), $1.19 \times 10^{-5}$ (@ 363 K), $1.23 \times 10^{-5}$ (@ 373 K) <sup>[30]</sup>
Binary diffusion constant (H <sub>2</sub> -H <sub>2</sub> O) [m <sup>2</sup> /s]	$9.27 \times 10^{-5}$ <sup>[31]</sup>
Binary diffusion constant (O <sub>2</sub> -H <sub>2</sub> O) [m <sup>2</sup> /s]	$3.57 \times 10^{-5}$ <sup>[31]</sup>
Porosity (catalyst layer) [-]	0.78 <sup>[10,32,33]</sup>
Permeability (catalyst layer) [m <sup>2</sup> ]	$8.69 \times 10^{-12}$ <sup>[10,32,33]</sup>
Thermal conductivity (catalyst layer) [W/(m·K)]	1.70 <sup>[34]</sup>
Porosity (MPL) [-]	0.60 <sup>[10,32,33]</sup>
Permeability (MPL) [m <sup>2</sup> ]	$1.00 \times 10^{-13}$ <sup>[10,32,33]</sup>
Thermal conductivity (MPL) [W/(m·K)]	1.00 <sup>[35]</sup>
Porosity (GDL) [-]	0.78 <sup>[10,32,33]</sup>
Permeability (GDL) [m <sup>2</sup> ]	$8.69 \times 10^{-12}$ <sup>[10,32,33]</sup>
Thermal conductivity (GDL) [W/(m·K)]	1.70 <sup>[34]</sup>
Porosity (separator) [-]	0.15 <sup>[36]</sup>
Permeability (separator) [m <sup>2</sup> ]	$1.50 \times 10^{-5}$ <sup>[36]</sup>
Thermal conductivity (separator) [W/(m·K)]	0.151 <sup>[36]</sup>
Conductivity (PEM) [S/m]	10 <sup>[37]</sup>
Conductivity (catalyst layer) [S/m]	53 <sup>[38]</sup>
Conductivity (MPL) [S/m]	1000 <sup>[39]</sup>
Conductivity (GDL) [S/m]	1250 <sup>[35]</sup>
Conductivity (separator) [S/m]	83,000 <sup>[36]</sup>
Reference equilibrium voltage (Anode) [V]	0
Reference equilibrium voltage (Cathode) [V]	1.229
Reference exchange current density (Anode) [A/m <sup>2</sup> ]	1000 <sup>[34]</sup>
Reference exchange current density (Cathode) [A/m <sup>2</sup> ]	1 <sup>[34]</sup>
Charge transfer constant (Anode) [-]	0.5 <sup>[40]</sup>
Charge transfer constant (Cathode) [-]	0.5 <sup>[41]</sup>

**Table 3.** Considered operation condition of power generation.

Operation parameters	Conditions	
The initial temperature of cell ( $T_{ini}$ ) [K]	353, 363, 373	
Total cell voltage [V]	Experimental data are used <sup>[9,14]</sup>	
Supply gas condition	Anode	Cathode
Gas type	H <sub>2</sub>	O <sub>2</sub>
Temperature of supply gas at inlet of cell [K]	353, 363, 373	353, 363, 373
RH of supply gas [%RH]	40, 80	40, 80
Pressure of supply gas at inlet of cell (absolute based) [MPa]	0.4	0.4
Flow rate of supply gas at inlet of cell [NL/min] (Stoichiometric ratio [-])	0.210 (1.5)	0.105 (1.5)

Higher operation temperature would cause PEM drying, which would also increase an ionic resistance, an ohmic loss and a material degradation<sup>[42]</sup>. As a result, it would be necessary to manage and control the temperature profile in the cell for the purpose of promotion of power generation performance. Especially, an  $O_2$  reduction reaction produces the heat and  $H_2O$  as well as consumes  $O_2$ , resulting that the complex phenomena occur in the cathode side. These phenomena occur on the interface between PEM and catalyst layer at the cathode side mainly. Since the power generation performance is influenced by temperature and humidification on the interface, this study focuses on the mass such as  $O_2$  and  $H_2O$  distributions, the temperature distribution and the current density distribution on the interface between PEM and catalyst layer at the cathode side.

The analysis points from A to K, as shown in **Figure 3**, were assigned in this study, which were the same as that in the authors' previous studies<sup>[11,12]</sup> to examine the impact of separator thickness on the mass such as  $O_2$  and  $H_2O$  distributions, the temperature profile and the current density profile. **Figure 3** shows the analysis points from A to K. We have conducted the analysis on the averaged value on the cross-sectional area on the interface between PEM and catalyst layer at the cathode side, covering parts under the gas channel as well as those under the rib.



**Figure 3.** Analysis points from A to K for the quantitative evaluation along the gas flow through gas channel.

### 3. Results and discussion

As to the validation of the model, the authors have used the similar and the same model in the previous studies<sup>[12,43]</sup>. The results and discussions using the model have been recognized by the reviewers of these journals. In addition, the commercial software COMSOL applied in this study was used in many previous studies and their results were validated well<sup>[14,15,18,27,28,44,45]</sup>. Therefore, the model was thought to be validated.

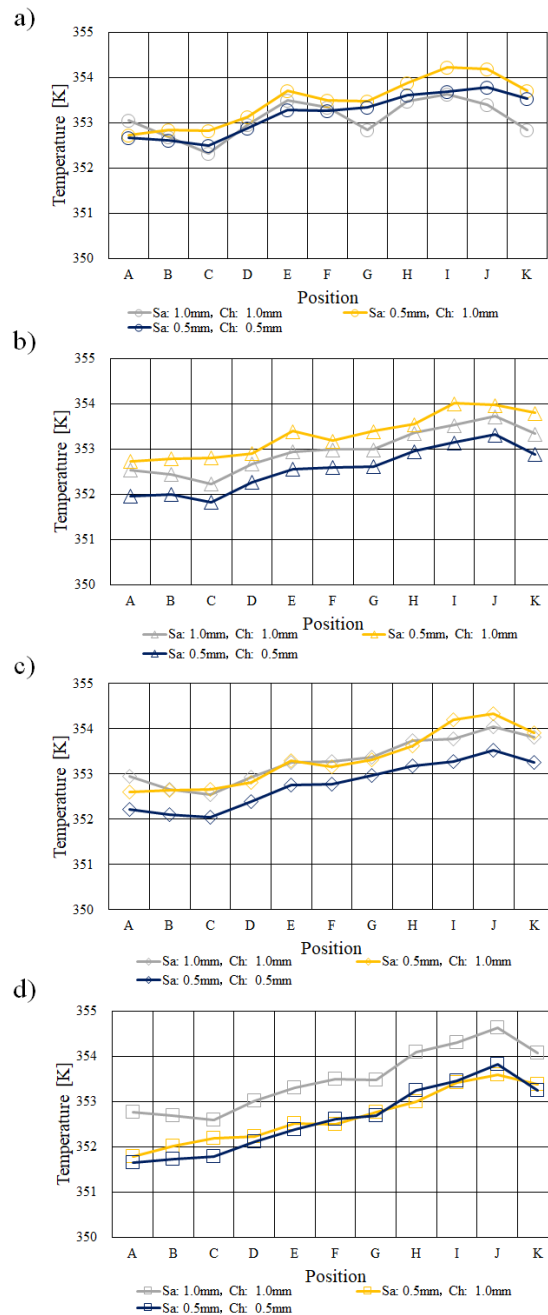
#### 3.1. Comparison of temperature profile

**Figures 4–6** show temperature distributions calculated by 3D numerical simulation model at  $T_{ini} = 353$  K, 363 K and 373 K, respectively. In these figures, the saddle thickness and the channel height are expressed by  $Sa$  and  $Ch$ , respectively. The effect of separator thickness on temperature profile is examined. Moreover, RH of supply gases is also varied.

It can be seen from **Figures 4–6** that the increase in temperature on the interface between PEM and catalyst layer at the cathode side from the inlet to the outlet of cell is smaller with the increase in  $T_{ini}$  irrespective of RH of supply gas. It is known that the saturation pressure of  $H_2O$  increases with the temperature exponentially<sup>[46]</sup>, resulting in easy dehydration of PEM at higher temperature than usual. Namely, it can be easy to reduce the proton conductivity of PEM at higher temperature, causing the decrease in power generation

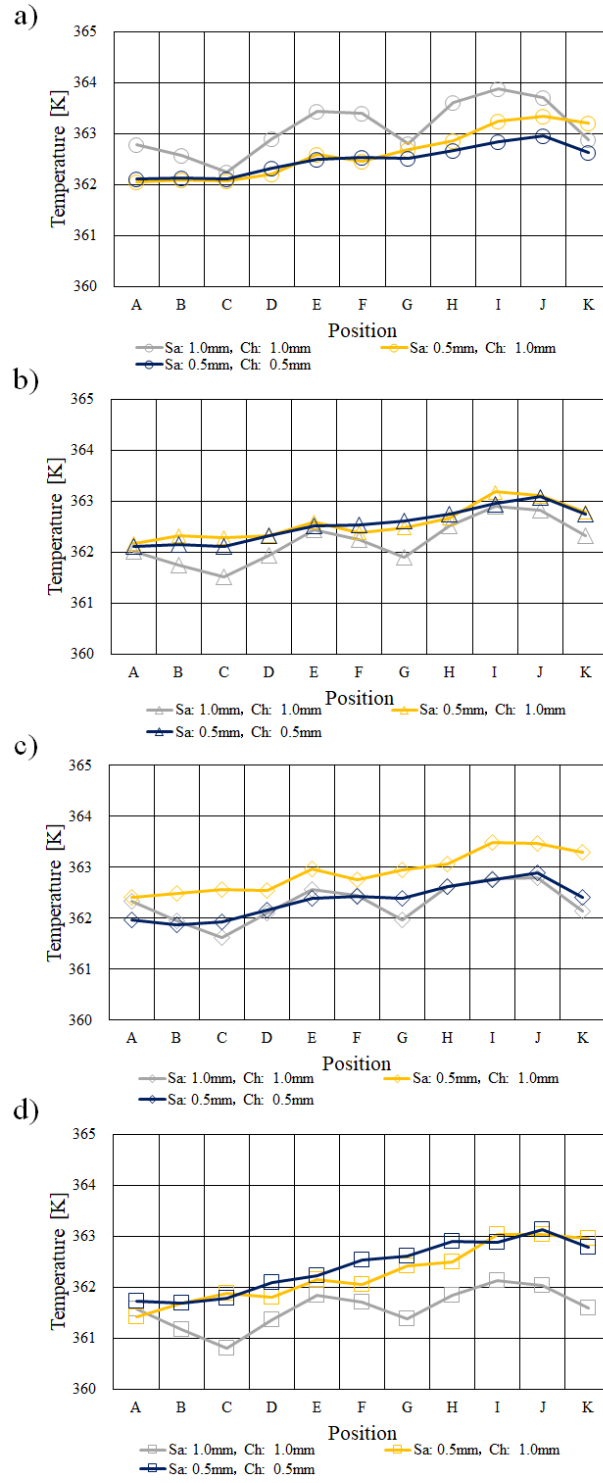
performance at higher temperature because of big ohmic loss. As a result, the generated heat decreases. Since we assume the excess amount of gas which is larger than  $s.r. = 1.0$  as the inlet gas flow rate, the generated heat is accumulated along with the gas flow through the gas channel<sup>[47]</sup>. Therefore, the temperature on the interface between PEM and catalyst layer at the cathode side rises from the inlet of cell to the outlet of cell.

Regarding the impact of separator thickness, the temperature change from the inlet to the outlet of cell, i.e., the temperature fluctuation along the gas flow, at  $T_{ini} = 353$  K and 363 K is larger when the separator thickness is 2.0 mm consisting of the saddle thickness = 1.0 mm and the channel height = 1.0 mm. Because the heat capacity of the separator thickness of 2.0 mm was the biggest among the separators investigated in this study, the dehydration of PEM and catalyst layer would be lower compared to the thinner separator thicknesses<sup>[12]</sup>. Consequently, it is thought that the power generation performance was improved with the increase in the separator thickness<sup>[12]</sup>. The reason why the temperature decreases at the positions of C, G and K as the increase in the separator thickness and RH of supply gases occur is discussed in the following sections.

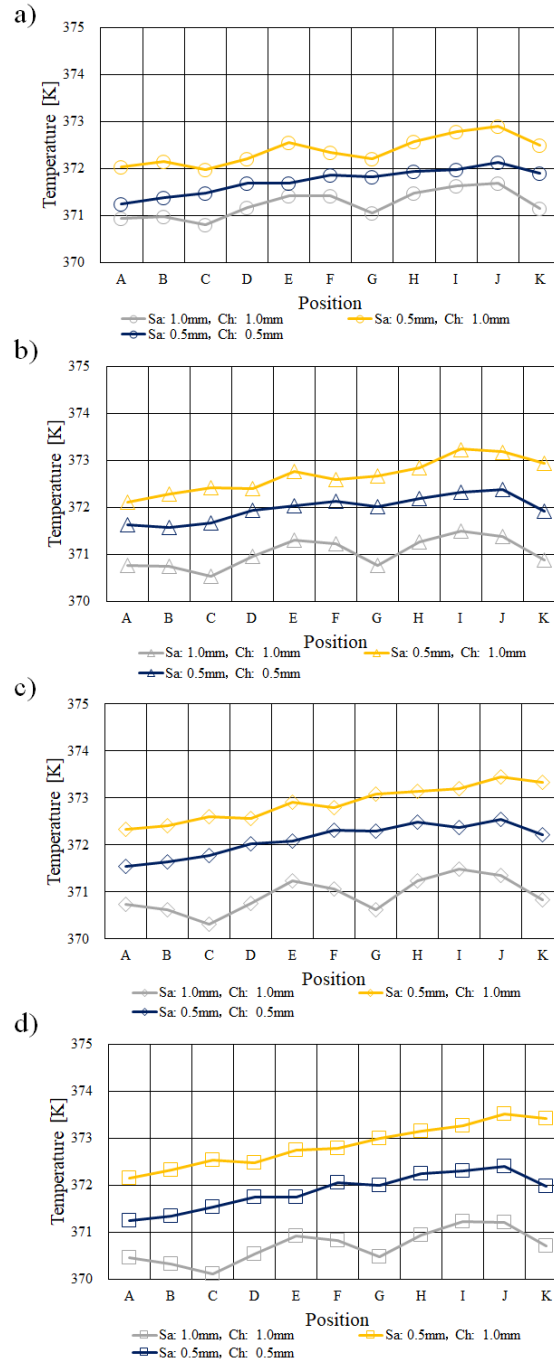


**Figure 4.** Comparison investigation on temperature profile among different separator thickness at  $T_{ini} = 353$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.





**Figure 5.** Comparison investigation on temperature profile among different separator thickness at  $T_{ini} = 363$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.



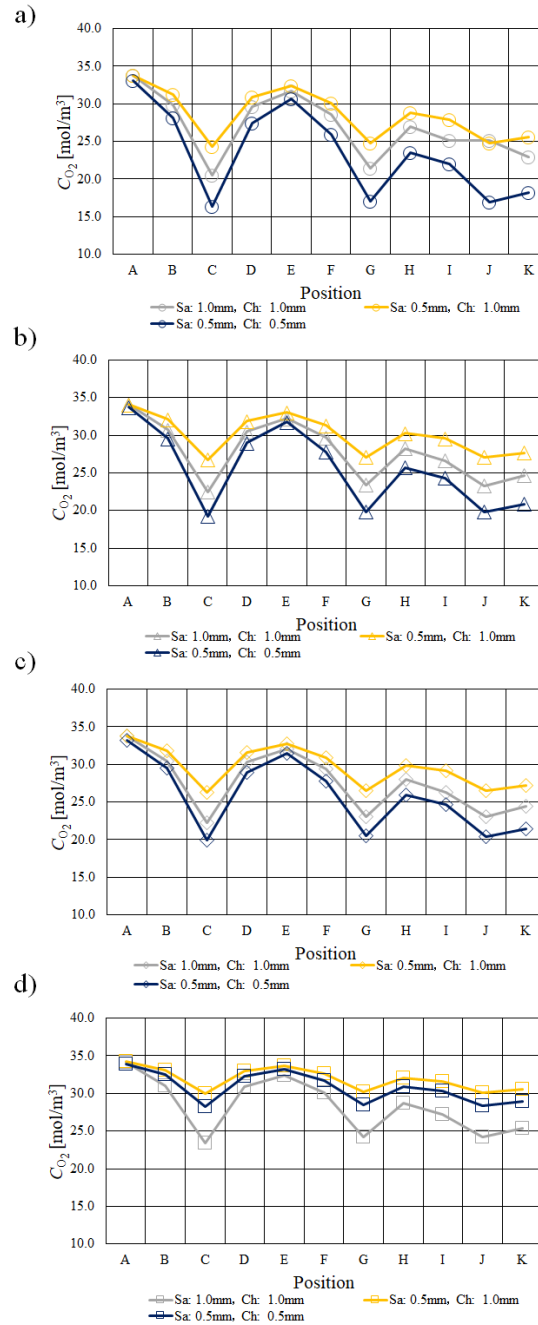
**Figure 6.** Comparison investigation on temperature profile among different separator thickness at  $T_{ini} = 373$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.

### 3.2. Comparison of $O_2$ distribution

**Figures 7–9** show  $O_2$  distributions calculated by 3D numerical simulation model at  $T_{ini} = 353$  K, 363 K and 373 K, respectively.

It can be seen from **Figures 7–9** that the decrease in the molar concentration of  $O_2$  ( $C_{O_2}$ ) from the inlet of cell to the outlet of cell, i.e., the consumption of  $O_2$ , becomes smaller with the increase in  $T_{ini}$  and the decrease in RH of supply gas irrespective of separator thickness. The  $O_2$  reduction reaction is carried out along the gas channel<sup>[43]</sup>. It is known that the saturation pressure of  $H_2O$  increases with the temperature exponentially<sup>[43]</sup> as described above, resulting in easy dehydration of PEM at higher temperature than usual. The proton conductivity of PEM reduces under higher temperature and low RH conditions due to the dehydration of PEM<sup>[43]</sup>. As a result, the ohmic over-potential becomes larger. On the other hand, the ionomer in the catalyst

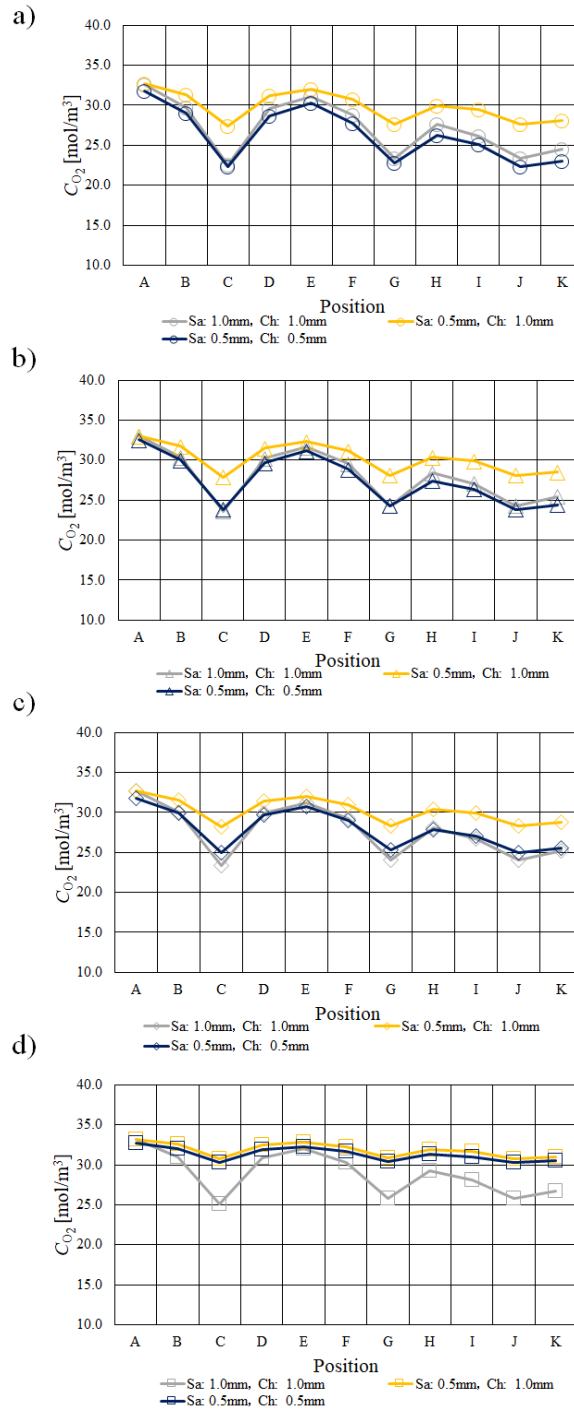
layer at the cathode side is not easy to be humidified by  $H_2O$  migrated through PEM from the anode side to the cathode side, which is significant issue for the performance of  $O_2$  reduction reaction at the cathode side<sup>[12,43]</sup>. The big ohmic over-potential is provided due to ionic and electronic resistances. The ionic resistance is related with the resistance of PEM as well as the ionomer of catalyst layer<sup>[48]</sup>. Therefore, the decrease in the molar concentration of  $O_2$  from the inlet of cell to the outlet of cell is smaller with the increase in  $T_{ini}$  as well as the decrease in RH of supply gas due to lower humidification.



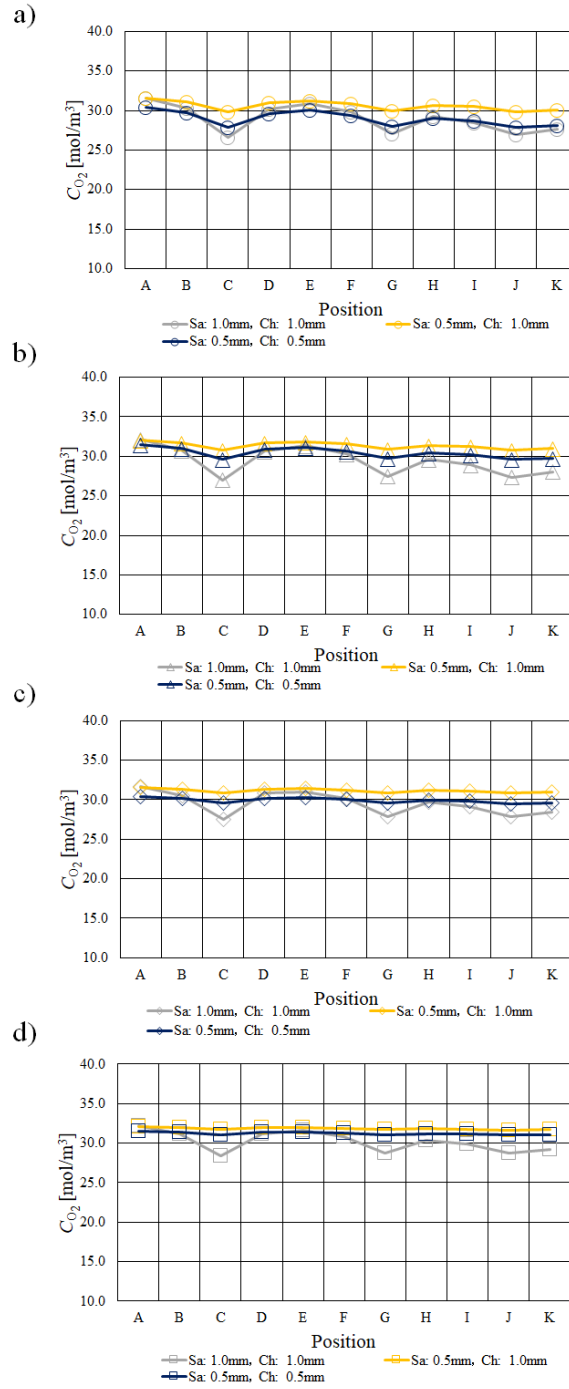
**Figure 7.** Comparison investigation on  $O_2$  profile among different separator thickness at  $T_{ini} = 353$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.

As to the impact of separator thickness, the molar concentration of  $O_2$  drops at analysis positions of C, G and J (and K) when the separator thickness is 2.0 mm, especially at  $T_{ini} = 353$  K and for A80%RH-C80%RH, which matches approximately the points of the temperature drop shown in **Figures 4–6**. The heat capacity of the separator thickness of 2.0 mm is the biggest among the separators investigated in this study, resulting that the dry up of PEM and catalyst layer would be lower compared to the thinner separator thicknesses<sup>[12]</sup>. In

addition, the humidification of PEM and catalyst layer is higher for A80%RH-C80%RH. Consequently, the  $O_2$  reduction reaction generating  $H_2O$  is improved with the increase in the separator thickness and RH of supply gas. The analysis points of C and G are located at the corner parts of the serpentine separator. Therefore, it can be thought  $H_2O$  accumulate there<sup>[49,50]</sup>. Additionally, it is considered that  $H_2O$  remaining in gas flowing through the gas channel accumulates near the outlet of cell<sup>[9,51]</sup>, which means the analysis points of J and K. As a result, the  $O_2$  diffusion is inhibited at analysis positions of C, G and J (and K)<sup>[13]</sup>, causing the reduction of the molar concentration of  $O_2$ . The impacts of separator thickness as discussed above become larger under higher temperature and lower RH conditions, which are thought to be easy dehydration conditions.



**Figure 8.** Comparison investigation on  $O_2$  profile among different separator thickness at  $T_{ini} = 363$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.



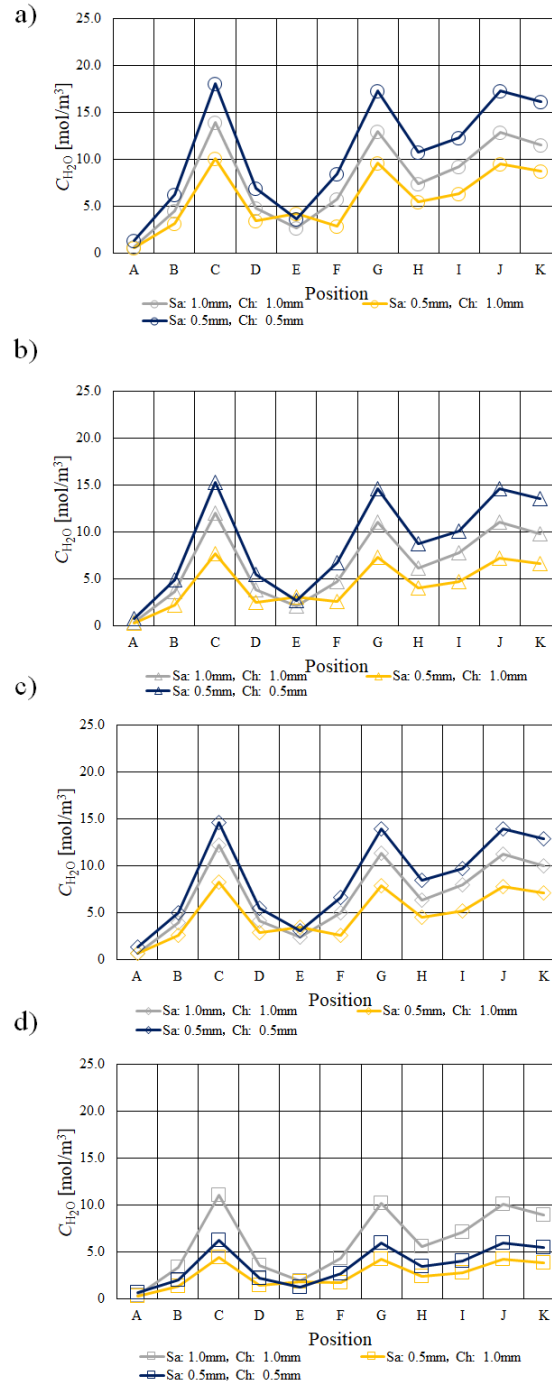
**Figure 9.** Comparison investigation on O<sub>2</sub> profile among different separator thickness at  $T_{ini} = 373$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.

### 3.3. Comparison of H<sub>2</sub>O profile

**Figures 10–12** show H<sub>2</sub>O profiles calculated by the 3D numerical simulation model at  $T_{ini} = 353$  K, 363 K and 373 K, respectively.

It can be seen from **Figures 10–12** that the increase in the molar concentration of H<sub>2</sub>O ( $C_{H_2O}$ ) from the inlet of cell to the outlet of cell becomes smaller with the increase in  $T_{ini}$  and the decrease in RH of supply gas irrespective of separator thickness. It is considered that H<sub>2</sub>O remaining in gas flowing through the gas channel accumulates along the gas flow<sup>[49,50]</sup>. It is known that the saturation pressure of H<sub>2</sub>O increases with the temperature exponentially<sup>[46]</sup>, resulting in easy dehydration of PEM at higher temperature than usual. The proton conductivity of PEM reduces under higher temperature and low RH conditions due to the dehydration of PEM<sup>[14]</sup>, causing larger ohmic over-potential. On the other hand, the ionomer in the catalyst layer at the

cathode side is not easy to be humidified by  $H_2O$  migrated through PEM from the anode side to the cathode side. It is significant for the performance of  $O_2$  reduction reaction at the cathode side<sup>[12,46]</sup>. The big ohmic over-potential is provided due to ionic and electronic resistances. The ionic resistance is related with the resistance of PEM as well as the ionomer of catalyst layer<sup>[48]</sup>. Since the humidification is lower under higher temperature and low RH conditions, the performance of  $O_2$  reduction reaction generating  $H_2O$  is smaller.

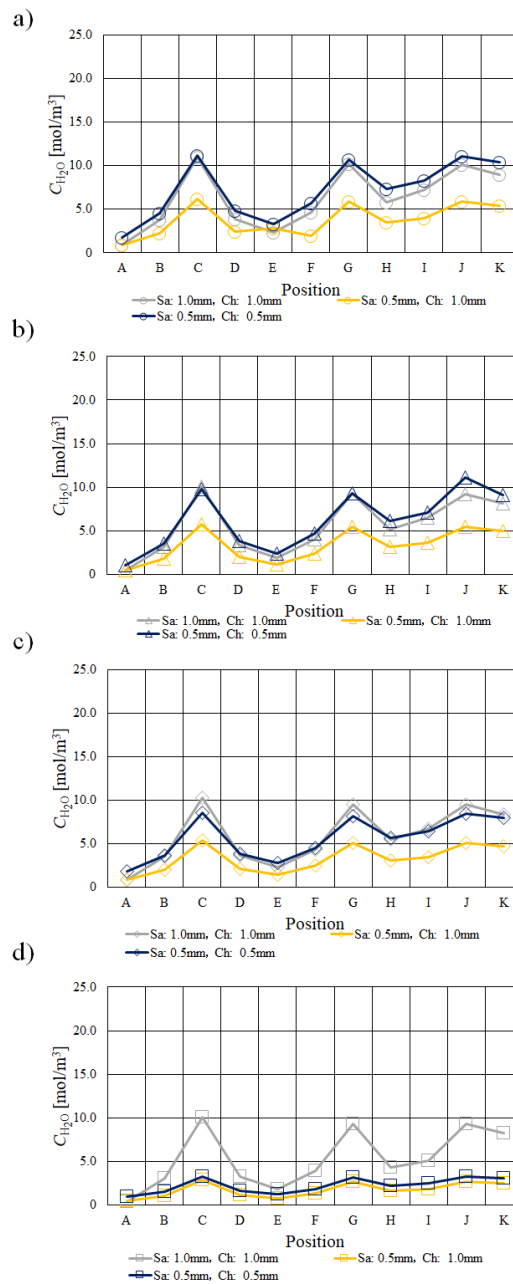


**Figure 10.** Comparison investigation on  $H_2O$  profile among different separator thickness at  $T_{ini} = 353$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.

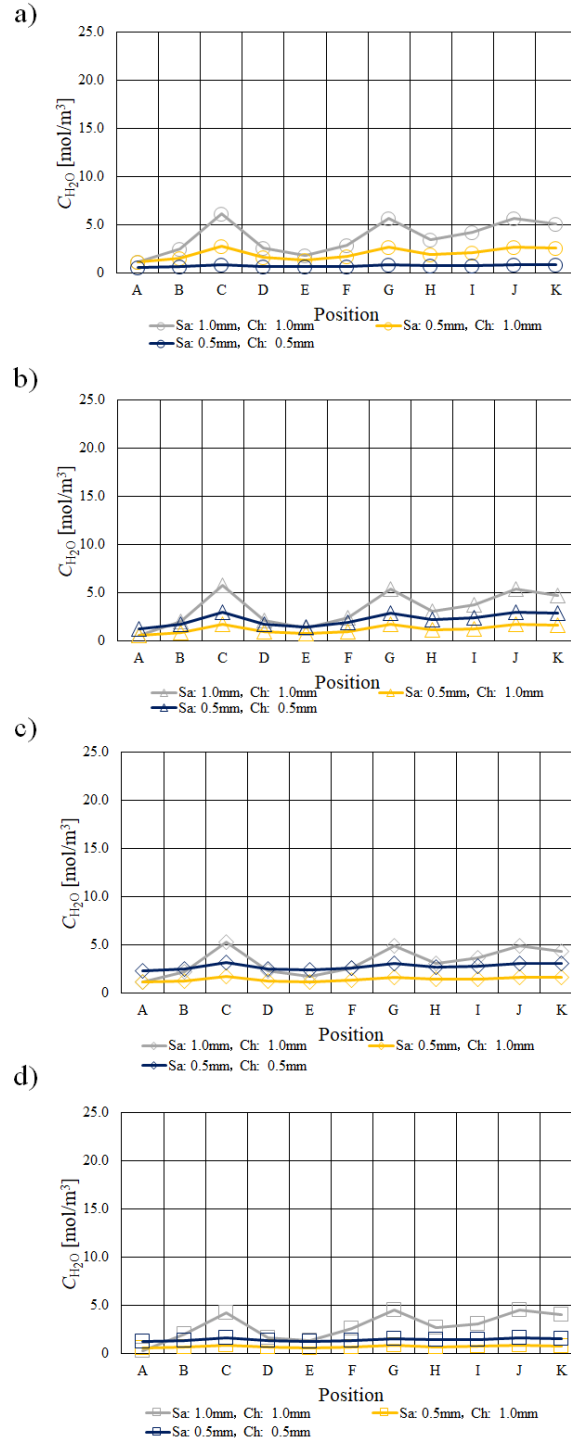
As to the impact of separator thickness, it is known from **Figures 10–12** that the molar concentration of  $H_2O$  increases at analysis positions of C, G and J when the separator thickness is 2.0 mm, especially at  $T_{ini} = 353$  K and for A80%RH-C80%RH, which matches approximately the points of the temperature drop shown in **Figures 4–6**. The heat capacity of the separator thickness of 2.0 mm is the biggest among the separators investigated in this study, resulting that the dry up of PEM and catalyst layer would be lower compared to the

thinner separator thicknesses<sup>[12]</sup>. In addition, the humidification of PEM and catalyst layer is larger for A80%RH-C80%RH. Consequently, the  $O_2$  reduction reaction generating  $H_2O$  is improved with the increase in the separator thickness and RH of supply gas. The analysis points of C and G are located at the corner parts of the serpentine separator. Therefore, it is thought  $H_2O$  may accumulate there<sup>[49,50]</sup>. Additionally, we can claim that  $H_2O$  remaining in gas flowing through the gas channel accumulates near the outlet of cell<sup>[9,51]</sup>, i.e., the analysis points of J and K. Consequently, the molar concentration of  $H_2O$  rises at the analysis points of C, G and J. As a result, the  $O_2$  diffusion is inhibited, the  $O_2$  reduction reaction is not carried out well there. Consequently, the heat generated by  $O_2$  reduction reaction decreases at the analysis points of C, G and J, causing the temperature drop. The impacts of separator thickness as discussed above become larger under higher temperature and lower RH conditions, which are thought to be easy dehydration conditions.

From this study, the saturation is below 1.0 under the investigated conditions, resulting that it can be thought the phase condition of  $H_2O$  is vapor. Therefore, the assumption that  $H_2O$  is a vapor is valid in this study.



**Figure 11.** Comparison investigation on  $H_2O$  profile among different separator thickness at  $T_{ini} = 363$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.



**Figure 12.** Comparison investigation on H<sub>2</sub>O profile among different separator thickness at  $T_{ini} = 373$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.

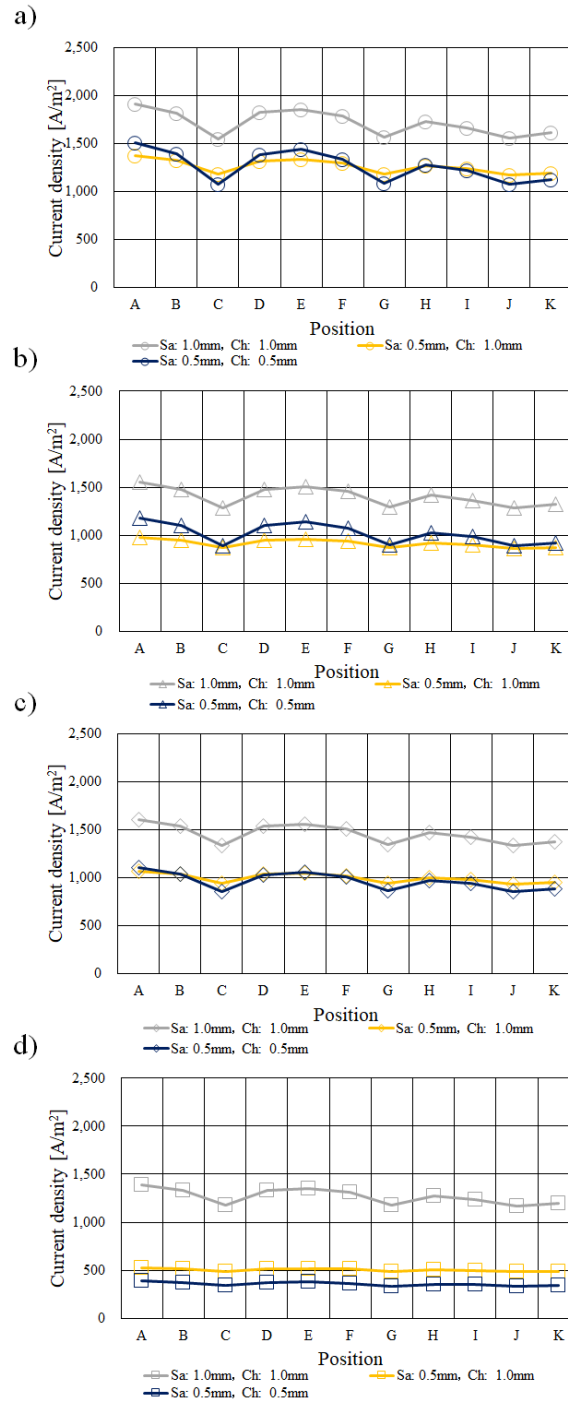
### 3.4. Comparison of current density profile

**Figures 13–15** show current density profiles calculated using 3D numerical simulation model at  $T_{ini} = 353$  K, 363 K and 373 K, respectively.

It can be found from **Figures 13–15** that the current density drops with the increase in  $T_{ini}$  and the decrease in RH of supply gas irrespective of separator thickness. It is known that the saturation pressure of H<sub>2</sub>O increases with the temperature exponentially<sup>[46]</sup> as described above, resulting in easy dehydration of PEM at higher temperature than usual. The proton conductivity of PEM reduces under higher temperature and low RH conditions since PEM is dehydrated<sup>[14]</sup>. Therefore, the ohmic over-potential becomes larger. On the other hand,



the ionomer in the catalyst layer at the cathode side is not easy to be humidified by  $H_2O$  migrated through PEM from the anode side to the cathode side, which is significant for the performance of  $O_2$  reduction reaction at the cathode side<sup>[12,46]</sup>. The big ohmic over-potential is provided due to ionic and electronic resistances. The ionic resistance is related with the resistance of PEM as well as the ionomer of catalyst layer<sup>[27]</sup>. Therefore, the current density reduces with the increase in  $T_{ini}$  and the decrease in RH of supply gas due to lower humidification.



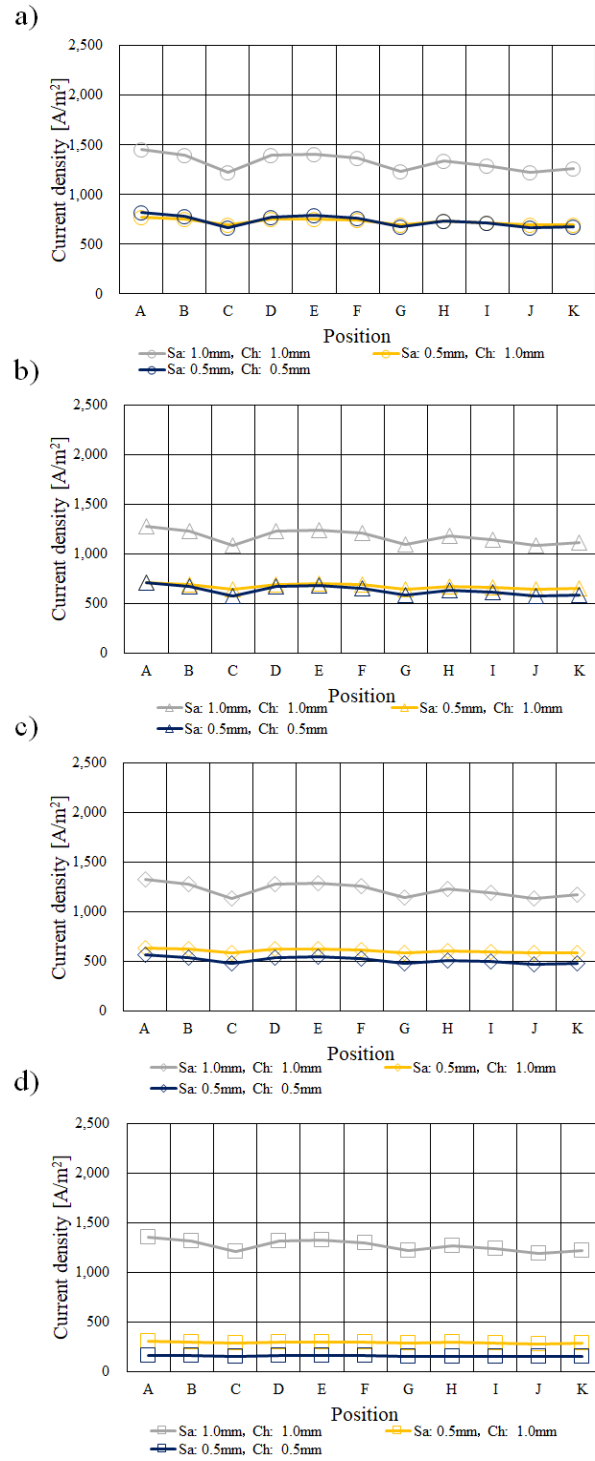
**Figure 13.** Comparison investigation on current density profile among different separator thickness at  $T_{ini} = 353$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.

According to **Figures 13–15**, the current density drops from the inlet of cell to the outlet of cell.  $H_2$  and  $O_2$  are consumed along with the gas channel, resulting that the driving force for the diffusion toward the catalyst layer reduces along with the gas channel. As a result, the current density reduces from the inlet of cell

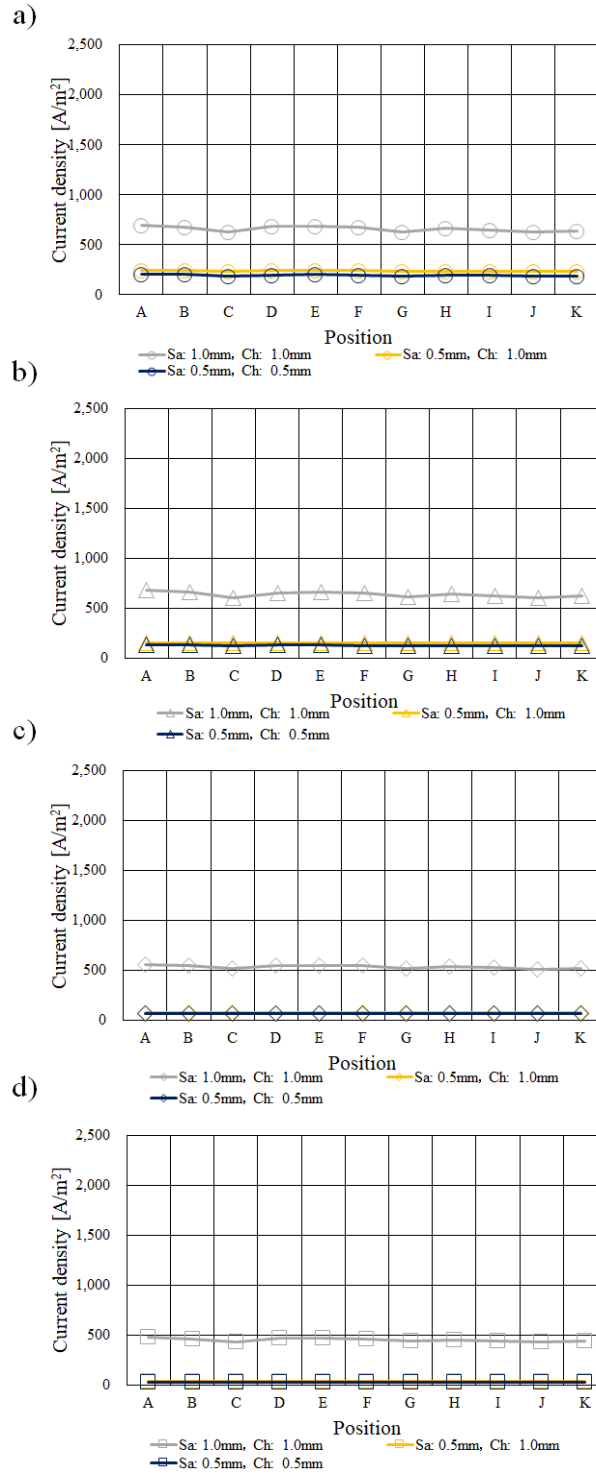
to the outlet of cell.

Regarding the impact of separator thickness, it can be found from **Figures 13–15** that the current density drops at analysis positions of C, G and J when the separator thickness is 2.0 mm, especially at  $T_{\text{ini}} = 353 \text{ K}$  and for A80%RH-C80%RH, which matches approximately the points of the temperature drop shown in **Figures 4–6**. The heat capacity of the separator thickness of 2.0 mm is the biggest among the separators investigated in this study. Therefore, the humidification of PEM and catalyst layer would be lower compared to the thinner separator thicknesses<sup>[12]</sup>. Additionally, the humidification of PEM and catalyst layer is larger for A80%RH-C80%RH. Consequently, the  $\text{O}_2$  reduction, which generates  $\text{H}_2\text{O}$ , is improved with the increase in the separator thickness and RH of supply gas. The analysis points of C and G are located at the corner parts of the serpentine separator. Therefore,  $\text{H}_2\text{O}$  may accumulate there<sup>[49,50]</sup>. Moreover, it is considered that  $\text{H}_2\text{O}$  remaining in gas flowing through the gas channel accumulates near the outlet of cell<sup>[9,51]</sup>, i.e., the analysis points of J and K, resulting in the increase in the molar concentration of  $\text{H}_2\text{O}$  at the analysis points of C, G and J. As a result, the  $\text{O}_2$  diffusion is inhibited, causing that the  $\text{O}_2$  reduction reaction is not carried out well there. We can also claim that the concentration over-potential is larger there. Consequently, the current density drops at the analysis points of C, G and J, causing the temperature drops shown in **Figures 4–6**. The impacts of separator thickness as discussed above become larger under higher temperature and lower RH conditions, which are thought to be easy dehydration conditions.

From the current study, the optimum separator thickness is 2.0 mm to realize higher power generation performance among the investigated separator thicknesses. The separator with the thickness of 2.0 mm is the commercial and normal type and the authors have customized the separator thickness of 1.5 mm and 1.0 mm to investigate the impact of channel height and saddle thickness on mass and heat transfer phenomena as well as power generation characteristics in this study. This study had set that the separator thickness of 2.0 mm is the base case and the other separator thicknesses are comparing cases. To decrease the thickness of separator keeping the strength of it, the separator thickness of 1.5 mm (saddle thickness = 0.5 mm, channel height = 1.0 mm) and 1.0 mm (saddle thickness = 0.5 mm, channel height = 0.5 mm) were selected in this study. If we select the separator thickness which is larger than 2.0 mm, it is expected that the power generation performance is improved due to preventing the dry up of PEM and catalyst more. Since the heat capacity is larger with the increase in the separator thickness, the dehydration of PEM and catalyst due to temperature rise is prevented. As a result, it is expected that the performance of  $\text{O}_2$  reduction reaction would be improved and the amount of  $\text{H}_2\text{O}$  produced by  $\text{O}_2$  reduction reaction increases. However, the optimum separator thickness for HT-PEMFC depends on the thermal design. The thinner thickness of separator is better to improve the volumetric power density of the stack. If we develop the separator whose heat capacity is larger than the separator thickness of 2.0 mm, it is expected that the damage of PEM and catalyst due to heat up would be prevented and the power generation performance could be improved due to preventing the dry up of PEM and catalyst. For example, a porous metal separator may be a good candidate since the larger heat capacity can be obtained even if the thickness is smaller than 2.0 mm. This is the future work in this study. If a separator could be designed, which could remove the generated heat smoothly, the separator thickness would be thinner. Since the weight ratio of separator to that of total cell is approximately 80%<sup>[21]</sup>, the thinner separator is desirable.



**Figure 14.** Comparison investigation on current density profile among different separator thickness at  $T_{ini} = 363$  K; **(a)** A80%RH-C80%RH; **(b)** A80%RH-C40%RH; **(c)** A40%RH-C80%RH; **(d)** A40%RH-C40%RH.



**Figure 15.** Comparison investigation on current density profile among different separator thickness at  $T_{ini} = 373$  K; (a) A80%RH-C80%RH; (b) A80%RH-C40%RH; (c) A40%RH-C80%RH; (d) A40%RH-C40%RH.

## 4. Conclusion

The impact of separator thickness on the relationship between the temperature profile and not only the current density profile but also the profiles of gases, e.g.,  $O_2$  and  $H_2O$  were examined in this study by numerical simulation using CFD software COMSOL Multiphysics. In the study, the operation temperature was set at 353 K, 363 K and 373 K respectively to compare the characteristics of HT-PEMFC with that of a general PEMFC. The following conclusions drawn from the study include:

- 1) The temperature change from the inlet of cell to the outlet of cell in the case of  $T_{ini} = 353$  K or 363 K was larger for the separator thickness of 2.0 mm compared to other separator thicknesses.

- 2) The decrease in the molar concentration of  $O_2$  and the increase in the molar concentration of  $H_2O$  from the inlet to the outlet of cell was smaller with the increase in  $T_{ini}$  and the decrease in RH of supply gas, respectively.
- 3) The current density decreased with the increase in  $T_{ini}$  and the decrease in RH of supply gas irrespective of separator thickness.
- 4) The optimum separator thickness was 2.0 mm to realize higher power generation performance among the investigated three separator thicknesses. If a separator which could remove the generated heat smoothly could be made, the separator thickness could be thinner.

## Author contributions

Conceptualization and writing—original draft preparation, AN; methodology and software, DM; data curation, SI; methodology TK; writing—review and editing, EH. All authors have read and agreed to the published version of the manuscript.

## Conflict of interest

The authors declare no conflict of interest.

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