Analysis of overall heat balance in self-heated proton-exchange-membrane fuel cells for temperature predictions☆

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Abstract

The effect of self-heating and cooling by natural convection on a sustainable temperature of PEM fuel cell stacks was studied. Overall mass and heat balance equations are combined to predict self-heated temperatures at various operating conditions. Analyses show that the effect of a heat loss coefficient is more important than other variables such as air flow rate and surrounding temperature. The stack design variables such as active cell area and number of cells also have significant influence on self-controlled temperature. A lower Ohmic resistance of cells is expected to allow a wider range of current load applications. The proposed model can also be used to evaluate heat loss coefficient from measured stack performance and temperature data. Experiments performed on a seven-cell stack of $50 \, cm^2$ active area were used to provide data for the validation of the model.
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1. Introduction

Proton-exchange-membrane fuel cells (PEMFC) have a narrow range of operating temperatures due to the need of water-containing electrolyte and the limit of low temperature for proton conductivity. Temperature control or thermal management is a practical issue in general for the design of fuel cell systems [1]. Unlike the high temperature fuel cells, a PEMFC works at a relatively low temperature and it does not have to rely heavily on the use of external thermal management systems for heating or cooling. At a temperature below $100 \, ^\circ C$, a self-controlled temperature is conceivable by balancing heat released from the membrane-electrode-assembly (MEA) and heat loss through end-plates to surrounding atmosphere. Self-heating makes the system simpler, just as self-humidification does. While self-humidification has been mostly a material design issue, self-heating and cooling is an engineering design issue.

Both the heat release and heat removal rates are dependent on many variables. The objectives of this study are: (1) to identify these variables and introduce a model that accounts for those aspects of self-heating and cooling, (2) to demonstrate how such a model can be applied to the design of stacks. An overall mass and energy balance model is developed in this work. Although we focused on using the model to predict temperature of PEM fuel cells in the absence of external heating and cooling devices in this work, the model developed here can be applied to any kind of fuel cells in general.

2. Overall heat and mass balance

We propose a model based on a combination of the steady-state overall heat and mass balance equations. With hydrogen

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coming in through anode and air through cathode, both of humidified, the overall mass balances for each species are expressed in mass or molar flow rates (n_j) and the reaction rate (r_{H_2O}) by Faraday's law.

$$n_{H_2,in(A)} - r_{H_2O} = n_{H_2,out(A)} \tag{1}$$

$$n_{H_2O,in(A)} = n_{H_2O,out(A)} \tag{2}$$

$$n_{O_2,in(C)} - 0.5r_{H_2O} = n_{O_2,out(C)} \tag{3}$$

$$n_{N_2,in(C)} = n_{N_2,out(C)} \tag{4}$$

$$n_{H_2O,in(C)} + r_{H_2O} = n_{H_2O,out(C)} \tag{5}$$

Water is assumed to be present in the saturated vapor phase only, for simplification of the model. A saturated water vapor pressure, P_{sw}, can be calculated from the saturation temperature, T_{sw}, using the thermodynamic equation (Antoine equation):

$$\ln P_{sw} = \frac{18.3036 - 3816.44}{T_{sw} - 46.13} \tag{6}$$

where the unites of P_{sw} is in mmHg and T_{sw} is in Kelvin.

Humidified gas feed rates are calculated with the added saturated water vapor as:

$$\frac{P_{sw}}{P_{system}} = y_{H_2O} \tag{7}$$

$$\frac{n_{H_2O(A)}}{n_{H_2(A)}} = \frac{y_{H_2O(A)}}{1 - y_{H_2O(A)}} \tag{8}$$

$$\frac{n_{H_2O(C)}}{n_{O_2(C)} + n_{N_2(C)}} = \frac{y_{H_2O(C)}}{1 - y_{H_2O(C)}} \tag{9}$$

The heat balance is obtained from the enthalpies of the anode and cathode gas streams at inlet and outlet with the reference temperature (T_0, usually 25 °C). Self-heating is calculated from heat generated during the fuel cell reaction (Q_{cell}). Self-cooling is calculated from heat loss to an atmosphere (Q_{loss}) with a surrounding temperature (T_{surr}) through a total external surface area of exposure (A_{ext}). External heating or cooling is not considered in this model. The heat balance equations are:

$$\text{input enthalpy} = n_{in(A)}C_{p,in(A)}(T_{in(A)} - T_0)$$
$$+ n_{in(C)}C_{p,in(C)}(T_{in(C)} - T_0) \tag{10}$$

$$\text{output enthalpy} = n_{out(A)}C_{p,out(A)}(T_{out(A)} - T_0)$$
$$+ n_{out(C)}C_{p,out(C)}(T_{out(C)} - T_0) \tag{11}$$

$$\text{heat generated} = Q_{cell} = (-\Delta H)_{H_2O}r_{H_2O} - iA_{cell}\sum V_{cell} \tag{12}$$

$$\text{heat loss} = Q_{loss} = h_{loss}A_{ext}(T_{cell} - T_{surr}) \tag{13}$$

The heat capacity of gas mixture is easily obtained from those of pure gases, and the enthalpy change of fuel cell reaction is shown in thermodynamic tables as a function of temperature [2].

$$C_{p,mix}(y_j, T) = \sum_j y_j C_{p,j}(T) \quad \text{and} \quad \Delta H = f(\Delta H_0, T) \tag{14}$$

There is a temperature distribution inside a fuel cell stack. In the present simplified model, the variation of temperature is neglected and a single average temperature (T_{cell}) is assumed, with a further assumption that outlet gas temperature is equal to this average cell temperature.

$$T_{cell} = T_{out(A)} = T_{out(C)} \tag{15}$$

Combining Eq. (10) through (15) and the assumptions made, we obtain the following simple equation for temperature predictions of self-heated fuel cells:

$$n_{in(A)}C_{p,in(A)}(T_{in(A)} - T_0)$$
$$+ n_{in(C)}C_{p,in(C)}(T_{in(C)} - T_0) + Q_{cell} - Q_{loss}$$
$$= (n_{out(A)}C_{p,out(A)} + n_{out(C)}C_{p,out(C)})(T_{cell} - T_0) \tag{16}$$

The overall balance model and parameters are schematically shown in Fig. 1. S_A and S_C are reactant stoichiometry (defined here is a ratio of the reactant gas flow rate divided by the amount required for 100% conversion to generate electrical current applied) of anode and cathode gases for a given current load. Thermodynamic properties (heat capacity and enthalpy change of the reaction) are dependent on temperature. The enthalpy change of fuel cell reaction ($H_2 + 1/2O_2 \rightarrow H_2O$) is represented by a linear correlation presented in Table 1. There is only a 0.2% variation of the enthalpy change for a 50 °C temperature change. Therefore, we assume a constant enthalpy change over the temperature range of interest. Such a negligible dependence on temperature is also observed in heat capacities. Table 1 shows linear correlations of enthalpy and heat capacities and their constant values at the PEM FC temperature range.

With these further simplifications, the input variables in the overall balance equation are all constants except the cell voltage (V_{cell}). Cell or stack voltage is dependent on current load, humidity, temperature, pressure, water flooding, and so on. Fuel cell models are often very comprehensive and account for all these effects. As a result of this comprehensiveness, the models proposed are very diverse as reported

Table 1
Linear correlation ($A + BT$) and average values of thermodynamic properties

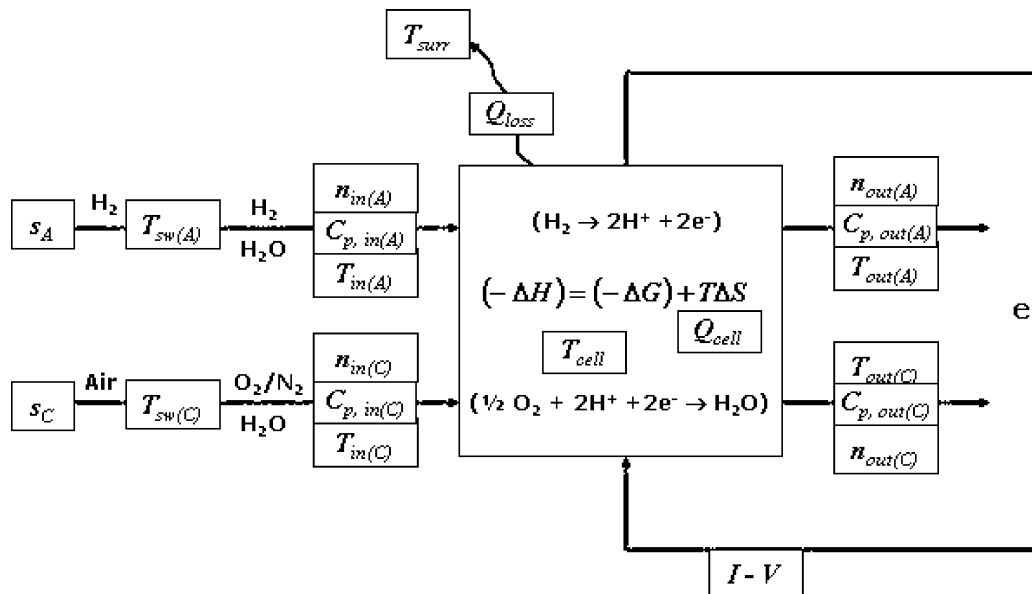| Parameter	A	B	Avg. (300–400 K)
$-\Delta H(H_2O)$	238870.3	9.92008	242346 J mol^{-1}
$C_p(H_2)$	28.209	0.0022	29.02 J mol^{-1} K^{-1}
$C_p(O_2)$	26.847	0.00839	29.75 J mol^{-1} K^{-1}
$C_p(N_2)$	28.451	0.00218	29.19 J mol^{-1} K^{-1}
$C_p(H_2O)$	31.171	0.00801	33.93 J mol^{-1} K^{-1}

Fig. 1. Overall heat balance model at steady-state.

in the literature [3]. Because so many variables affect cell performance in very different patterns and also because there is no single equation to account for such a relationship, to avoid unnecessary complication, we choose the well-known model using a Tafel slope (b) and Ohmic resistance (R_{Ohm}) to calculate the cell voltage:

$$V_{\text{cell}} = E_0 - b \ln i - i R_{\text{Ohm}} \tag{17}$$

3. Experimental setup

A commercially available small PEM fuel cell stack was used to provide experimental data for model validation. The stack has seven MEAs (Nafion-115, $1\,\text{mg cm}^{-2}$, 20 wt.% Pt/C) with an active area of $50\,\text{cm}^2$ (EFC-50-ST, Electrochem Inc., Woburn, MA). The six graphite bipolar plates have a dimension of 4 in. × 4 in. × 1/4 in., and two graphite end-plates have a dimension of 4 in. × 4 in. × 1/2 in. with the two end frames of 5 in. × 5 in. × 1/2 in. The total external surface area is $820\,\text{cm}^2$. We tested this stack in a test system equipped with bubble humidifiers and heated gas transfer lines (FCTS-HTK and FCTS-HB, Lynntech Industries Ltd., College Station, TX). Several tests were done at various current loads and humidifying conditions, as shown in Table 2. In each test, current load increased from zero to a certain value, and the temperature rise and the change of stack voltage were observed until they reach steady-state values.

4. Results and discussion

The variables in the model schematics shown in Fig. 1 are classified into three categories: controllable, dependent, and noise variables. Controllable or input variables are those we can control independently, and they are reactant stoichiometries (S_A and S_C), saturated water temperatures (T_{sw}), heating line temperatures (T_{in}), etc. Active cell area (A_{cell}), number of cells for stack (N_{cells}), and external area of exposure (A_{ext}) are also controllable at the design stage, though not during operation.

To examine the effects of these variables on self-maintained temperature in PEM fuel cells, calculations were done for the seven-cell stack (described in Section 3) at a bubble humidifier temperature of 75 °C, a surrounding temperature of 20 °C, and a heat loss coefficient of $10.0\,\text{W m}^{-2}\,\text{K}^{-1}$. The relationship between current density and cell performance (voltage) needs empirical parameters, and we chose the reference data for the empirical parameters related to the Tafel equation from the literature [4]: $E_0 = 0.942\,\text{V}$; $b = 0.061\,\text{V}$; and $R_{\text{Ohm}} = 0.39\,\Omega\,\text{cm}^2$. These are for Nafion 115 at 50 °C.

4.1. Effect of operating condition and design variables

Fig. 2 shows the effect of one of the controllable variables, air stoichiometry. Self-heated temperature was calculated by changing air stoichiometry from 1.0 to 10.0 while other variables are unchanged. The fuel (hydrogen) stoichiometry was also kept constant at 1.5. In fuel cells, the air stream to cathode is frequently used for the purpose of cooling, especially in large-scale high-temperature fuel cells [5]. In PEM fuel cells, a high flow rate of air is also useful to minimize water accumulation in the cathode channel.

Fig. 2 shows that the high flow rate of air (with a higher stoichiometry) has some cooling effect at a relatively high

Table 2
Experimental conditions for the stack tests to measure self-heated temperature

No.	Stoichiometry, S_A/S_C	T_{sw} (°C), H_2/air	I (A)	V_{stack} (V)	T_{avg} (°C)	h_{loss} (W m^{-2} K^{-1})
1	1.20/1.20	0/0	1.0	1.988	28.06	13.64
2	1.20/1.20	0/0	2.0	1.346	29.58	23.86
3	1.20/1.20	0/0	3.0	0.711	36.29	20.54
4	2.82/2.96	0/0	4.0	0.694	43.65	17.75
5	2.82/2.96	50/50	4.0	1.09	47.06	15.08
6	2.82/2.96	22/50	4.0	0.813	46.98	15.50
7	2.25/2.37	22/22	5.0	0.735	50.77	16.57
8	2.25/2.37	50/22	5.0	1.176	51.69	15.27
9	3.01/3.16	70/60	5.0	2.324	57.6	11.24
10	3.76/4.73	53/53	2.5	2.0	43.0	10.09

current load. At a relatively low current load of 0.25 A cm^{-2}, the air cooling has almost no influence on temperature. At such a low current density, the consumption of fuel is very small and the rate of heat generation is too low to heat the fuel cell stack. The temperature at 0.25 A cm^{-2} is even slightly increased as air stoichiometry increases. This is because the gas inlet temperature (75 °C) is higher than the fuel cell temperature. The increased air stream carries more heat from the humidified line to the fuel cell stack than that produced from the stack itself, as the heat generation rate is very low. If the humidifying condition changes to a lower temperature, we will see the air cooling effect by an increased air stoichiometry, though not significant at a low current load.

Figs. 3 and 4 show the effects of stack size in terms of active area and number of cells. In single-cells, there is almost certainly no effect of self-heating on fuel cell temperature. Single-cells are mostly used for fundamental research purposes in laboratories. In laboratory tests, cell temperature usually needs to be controlled at a desired set point. Therefore, single tests need external heating, even at a high current load. Larger-scale stacks, on the other hand, have a distinct self-heating effect resulting in a significant temperature rise. More often, they require cooling than heating unless
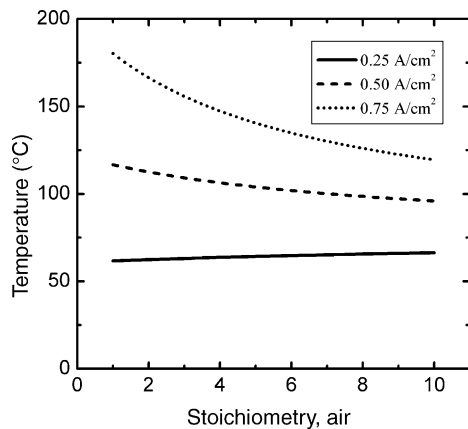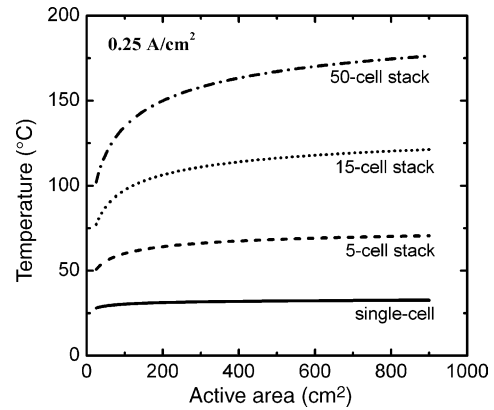

Fig. 3. Effect of active area on self-heated temperature at a current density of 0.25 A cm^{-2}.

the current load is extremely low. As we compare the cases of Figs. 3 and 4, the five-cell stack cannot maintain temperature below 100 °C at a current density of 0.5 A cm^{-2}. This indicates a self-heating stack can be used only for low current applications.

Fig. 5 shows a contour plot of temperature on the plane coordinate of active area and number of cells. This plot il-
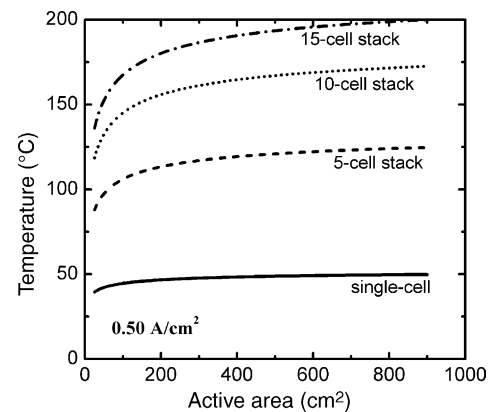

Fig. 2. Effect of air flow rate (stoichiometry ratio) on self-heated temperature.

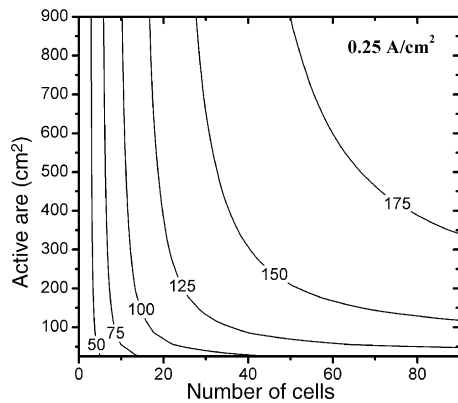Fig. 4. Effect of active area on self-heated temperature at a current density of 0.50 A cm^{-2}.

Fig. 5. Temperature contours (°C) for the effect of stack size (active area and number of cells) on the self-heating PEM fuel cell stack.

Fig. 7. Self-heated temperature as a function of current density at various Ohmic resistances.

lustrates a design issue of stack dimension. Even at a low current application, a self-heating stack is to be designed to have at least one stack dimension limited: either the number of cells should be small or the active area should be small.

4.2. Effect of fuel cell performance variables

Dependent variables are usually output variables such as cell voltage, which is strongly dependent upon materials (membrane, catalysts, and gas diffusion layers), operating conditions, and hardware such as cell frames. A decrease of cell voltage means a lower efficiency in fuel conversion to electrical energy which then means more thermal energy produced. The effects of two cell performance parameters (Tafel slope and Ohmic resistance) were studied. In Fig. 6, the Tafel slope varied from 0.01 to 0.09 and the self-heated temperature was calculated. Fig. 7 shows the effect of Ohmic resistance when it changed from 0.05 to 3.0 Ω cm^2. The Tafel slope is a measure of activation overpotential which is dominant at relatively low current load. Due to the relatively low amount
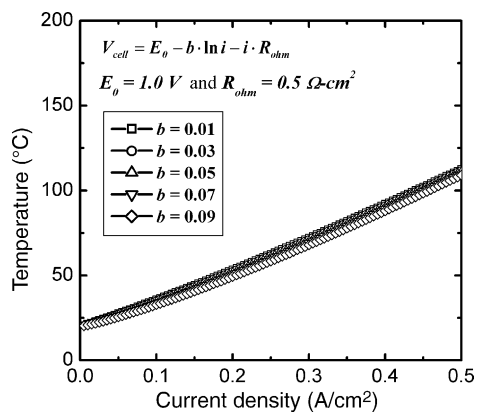
of heat produced at a low current load, the effect of Tafel slope is not significant on self-heated temperature, as shown in Fig. 6. The effect of Ohmic resistance is more significant because it is the dominant factor for cell performance at an increased current load. Obviously, sustainable fuel cell temperature is obtained for a broader range of current load when MEA materials have lower Ohmic resistances, as shown in Fig. 7.

4.3. Effect of noise variables

Noise variables are those which have significant influences on the performance variables but it is difficult to control. This is a term usually encountered in statistical analysis or quality engineering. In self-heating/cooling PEM fuel cells and stacks, heat loss to an atmosphere is generally difficult to predict, due to a relatively large variance of surrounding temperature (T_{surr}) and uncertainty in a heat loss coefficient (h_{loss}). For this reason, they are considered noise variables. We can reasonably presume that the surrounding atmosphere temperature varies from place to place in the range of 0–40 °C. The heat loss coefficient is approximately in the range of 3–12 W m^{-2} K^{-1} for natural convection through the surrounding air and in the range of 12–85 W m^{-2} K^{-1} for forced convection through air [6]. A total external area of cells/stacks exposed (A_{ext}) can be a noise factor, too, because it is often difficult to define this area in different shapes and configuration of cell/stack frames and end-plates.

Fig. 8 shows that a high current load operation requires forced convection to maintain temperature below 100 °C. With only natural convection near room temperature, stack operation is limited up to 0.5 A cm^{-2} in this specific example. Forced convection usually needs a device with an external supply of energy such as a motor-driven fan. For practical applications, the energy to drive such an external cooling device must be provided from the fuel cell stack. Consider a case when a fuel cell device needs to run at a current density of 0.75 A cm^{-2} for a given stack design and performance: if we use the result of Fig. 8 for further design, self-heating will raise the stack temperature to near 150 °C with natu-

Fig. 6. Self-heated temperature as a function of current density at various Tafel slopes.
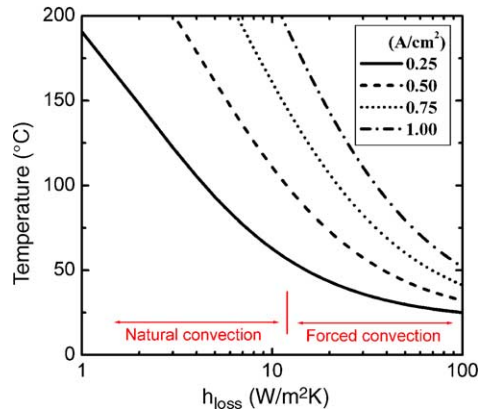
Fig. 8. Effect of the heat loss coefficient on self-heated fuel cell temperature at various current loads with constant stoichiometry ratios of fuel ($S_A = 1.5$) and oxidant ($S_C = 2.5$) gases.

ral convection cooling (assuming $h_{loss} = 10\,\mathrm{W\,m^{-2}\,K^{-1}}$). To maintain temperature below 80 °C, the stack needs a cooling fan that can increase the heat loss coefficient to about $30\,\mathrm{W\,m^{-2}\,K^{-1}}$. It will take a couple of experimental tests to evaluate how much power is needed to achieve such a heat loss coefficient. If the expected power requirement is a small portion of the power produced from the fuel cell stack, the design of this self-heated stack with an auxiliary cooling fan is feasible with respect to overall heat balance and power consumption. This case illustrates how the proposed model can be quickly and effectively used for design of fuel cell application systems.

Fig. 9 shows the effect of another noise parameter, surrounding temperature, on self-heating. The surrounding temperature in the range of 0–40 °C does not have a very significant influence compared to the other parameters.

4.4. Evaluation of heat loss coefficient

Table 2 summarizes ten experimental tests for measurement of self-heated temperature at various conditions. This
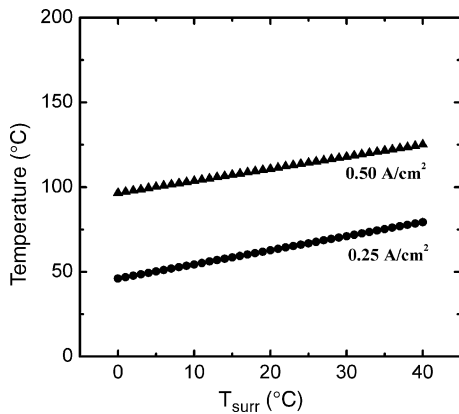

Fig. 9. Effect of surrounding temperature on self-heated fuel cell temperature at various current loads with constant stoichiometry ratios of fuel ($S_A = 1.5$) and oxidant ($S_C = 2.5$) gases.

seven-cell PEM FC stack showed a relatively poor overall performance, and it was difficult to apply a load current higher than 5 A (0.1 A cm^{-2}) due to a low stack voltage. Stack voltage was unstable during the constant current load, and it was more unstable in the test with dry fuel and air gases ($T_{sw} = 0$ °C). Temperatures were measured at three different points (two end-plates and one bipolar plate), and they were averaged. There was, in fact, a small degree of temperature variation. The inlet end-plate temperature was lower than the outlet end-plate temperature, and the center bipolar plate temperature was the highest. In the overall balance model, however, an average temperature is sufficient to evaluate a heat loss coefficient from Eqs. (10) to (16). The resulted heat loss coefficient ranged from 10.09 to 23.86 W m^{-2} K^{-1}.

The results in Table 2 show that the heat loss coefficient becomes high with dry gases, and also it becomes low when self-heated temperature increases. Overall, however, the heat loss coefficient does not make a reasonable correlation with any variables, so we believe this heat loss coefficient is intrinsic to the system. It will vary from one stack to another, depending on stack size and materials, but it is an intrinsic value of a specific stack; therefore, particular attention is required to the heat loss coefficient in the process of fuel cell stack design in order for self-heating to be effecting.

5. Conclusion

The proposed overall balance model allows us to study the effects of various parameters on self-heated fuel cell temperature. The model is straightforward, and can provide an average temperature that is sufficient for the purpose of a basic design of specific fuel cell application systems without external heating. From our case studies, we can draw several findings to conclude: (1) self-heating fuel cells can be effective for low current loads in general; (2) the design of stack size is important and at least one of the stack dimensions (area or number of cells) should be small for self-heating to be effective; (3) the range of allowable current load increases with lower Ohmic resistance materials; (4) high current load applications of self-heating stack can be made possible by increasing the heat loss coefficient to the range of forced convection; (5) heat loss coefficient has a strong effect on self-heating effectiveness. We also found from experimental measurement that the heat loss coefficient by natural convection is mostly in the range of 10–20 W m^{-2} K^{-1}, which agrees with the typical range of heat transfer coefficient in atmosphere known from the literature.

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