Cooling Technology for MCFC Stack*

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This research is intended to study relationship between cooling design of molten carbonate fuel cell (MCFC) stack and stack/system performance quantitatively. Firstly energy balance and temperature distribution are studied in 10 kW internal reforming (IR) MCFC stack. Based on the analysis 57% of waste heat is removed by endothermic reforming reaction in the IR-MCFC. The rest of the waste heat is removed by cathode gas. Cooling by internal reforming reaction (IR-cooling) reduces flow rate of recycled cathode gas necessary for stack cooling, which improves stack voltage and system performance. As for the temperature distribution, IR-cooling establishes more flat distribution, which increases average stack temperature and improves stack voltage. The present IR-MCFC has not fully taken advantage of the above profits of the IR-cooling design. By increasing capacity of IR-cooling, further improvement of stack/system performance can be achieved.

Key Words: Energy Conversion, Chemical Reaction, Fuel Cell, MCFC, Cooling Design, Internal Reforming

1. Introduction

Most of MCFC stacks are in general cooled by process-gas (PG) cooling, utilizing sensible heat of process-gas (typically, oxidant gas). The PG-cooling has an advantage of a simple plant design because of the minimum additional equipment for stack cooling. But the PG-cooling has disadvantages in performance: difficulty in achieving uniform temperature distribution and decrease of stack/system performance due to large flow rate of recycled reactant gas⁽¹⁾.

As for cooling performance, the IR-MCFC is considered to have a good performance⁽²⁾. In the IR-MCFC stack, reforming reaction and fuel cell reaction coexist, which enables efficient combination of endothermic (reforming) reaction and exothermic (fuel cell) reaction in one stack. Better cooling performance generally improves stack/system performance:

higher stack voltage, higher system efficiency, more stable stack performance, etc.

In this paper performance of the IR-cooling is evaluated, focusing on effects of cooling design on temperature distribution, on recycled gas flow rate/stack voltage, and on parasitic power for gas recycle. The above analyses are done based on two 10 kW stack tests. Considering the above analyses, possibility of further improvement of the present IR-MCFC is discussed.

2. Experiment

Two kinds of experiments are performed using the two 10 kW IR-MCFC stacks in order to measure energy balance/temperature distribution in the stack and dependency of stack voltage on cathode gas condition. Specifications of the both stacks are shown in Table 1. Stack (A) is the stack tested for measuring energy balance and temperature distribution. Stack (A) has 1 m² effective cell area and 12 cells. Stack (B) is the stack tested for measuring stack voltage dependency of cathode gas operating condition. Stack (B) has 1 m² effective cell area and 8 cells. In both stacks reforming plates (R.'s) are

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Table 1 Specifications of stack (A) and (B)

Specifications	Stack (A)	Stack (B)
Cell Size / Number	1 m ² / 12 cells	1 m ² / 8 cells
Number of Reforming Plate (R)	2 R's (3-R-6-R-3)	1 R (4-R-4)
Gas Flow	Counter Flow (Oxidant vs. Fuel) Parallel Flow (Oxidant vs. Reforming Gas)	
Cooling Design	IR + Cathode Gas Cooling	
Reformer Design	Internal Reforming (Plate + Direct Reformers)	

Table 2 Test conditions of stack tests

Condition	Energy Balance Test	Cathode Gas Condition Test
Stack No.	Stack (A)	Stack (B)
U _{O₂}	46%	22 ~ 60%
U _{CO₂}	41%	38 ~ 76%
Pressure	0.1 MPa	0.1, 0.5 MPa
Current	150 mA/cm ²	
Fuel	City Gas + Steam	
Oxidant	Air + CO ₂ + N ₂	
U _f	80%	

inserted in the stack (laminated body): Stack (A) has 2 reforming plates in the stack (3 cells/R./6 cells/R./3 cells) and Stack (B) has 1 reforming plate (4 cells/R./4 cells). The reforming plate works as a pre-reformer, receiving raw fuel gas from the outside, reforming the fuel gas into hydrogen-rich gas, and sending the reformed fuel gas to the anode gas channel of the fuel cell. The residual methane in the fuel gas is further reformed at the anode gas channel by the help of the reforming catalyst loaded there (direct reformer). In the reforming process the reforming plate works as a cooling plate, receiving excess heat from the neighboring fuel cells. In the reforming plate reforming catalyst is loaded in such selected area that uniform temperature distribution is to be obtained⁽³⁾. As for the direct reformer, reforming catalyst is loaded evenly in all the effective cell area.

Operating condition for both tests is summarized in Table 2. Temperature distribution exists in the stacks and temperature is usually in the range of 570 – 670°C. In order to compensate for heat loss from the stack, electrical heater-plates are set at the top/bot-

Fig. 1 Operating condition of energy balance test

tom ends of the stacks. In the energy balance test heater power is specially adjusted so that temperature of the end cells becomes almost same as the one of the center cell. In such adjustment electrical input to the heater is almost same as the heat loss estimated from the stack design for thermal insulation.

3. Energy Balance

Energy balance and temperature distribution in the IR-MCFC stack are studied using Stack (A). Reactant gas flows/temperatures at the test condition are shown in Fig. 1. The figure shows the anode-side surface of the bipolar plate. Manifolds are set on each corner of the plate. The hexagonal area surrounded by the four manifolds is the effective cell area (1 m²). The tube on the upper side of the plate in the figure is the feed tube, which connects to the plate reformer neighboring to the bipolar plate. The raw fuel (CH₄) is fed to the reforming plate through the gas feed tube. The raw fuel is reformed in the reforming plate and fed out to the anode-in manifold. The reformed fuel is redistributed in the manifold to the anode gas channel of each fuel cell. The shaded area in the figure corresponds to the catalyst-loaded area in the reforming plate. The reforming catalyst is loaded at the latter half of the effective cell area in the direction of the cathode gas flow.

Cathode gas enters the stack at 559°C and leaves at 669°C. The stack is cooled by the cathode gas by the difference of these two temperatures. The raw fuel (city gas) is reformed at first at the reforming plate up to 86% and simultaneously cools the stack. The residual 14% of the raw fuel is reformed in the anode gas channel (direct reformer). As a whole fuel

Table 3 Energy balance of stack (A)

Energy Input (kW)	Energy Output (kW)
ΔH (enthalpy change) of H_2 Oxidation 22.1	DC Output 12.7
	IR Cooling 5.3
	Cath. Gas Cooling 4.0
	Fuel Gas Cooling 0.1
Total 22.1	Total 22.1

gas enters the stack at 570°C and leaves at 585°C. In the present counter-flow design, anode gas leaves the stack at relatively low temperature close to the cathode inlet temperature, because flow rate of the anode gas is as small as 1/13 of the cathode flow rate. Therefore contribution of the fuel gas to the stack cooling is negligible as far as sensible heat of it is concerned.

Energy input/output is calculated based on the operating parameters of the stack: stack voltage, current, temperature, gas composition, and gas flow rate. Energy (excess heat) removed by IR-cooling is calculated as difference of enthalpy of anode inlet gas vs. anode exit gas. Gas composition of reactants is measured by gas chromatography. The calculated results are summarized in Table 3. As shown in the table good energy-balance is obtained.

As for cooling load of the stack, the difference between the enthalpy change of H_2 oxidation (ΔH) and DC output is the heat to be removed. 57% of the cooling load is removed by the IR cooling and 42% of it is by the cathode gas cooling. Contribution of the fuel gas (sensible heat) is small.

Advantage of the IR-MCFC is an effective cooling utilizing endothermic reforming reaction. In the example mentioned above about half of the cooling load is removed by the IR cooling, which reduces cooling load of the cathode gas cooling. The cathode gas cooling has such disadvantages as difficulty in attaining uniform temperature distribution and dilution of reactant gas due to high flow rate of recycled cathode gas. In the standard PG-cooled MCFC system, required flow rate of cathode gas for stack cooling is usually supplied by recycling a part of cathode exhaust gas to cathode inlet. Reduction of cooling load of cathode gas improves stack/system performance by suppressing the above disadvantages.

The temperature distributions of #6 cell (center cell) and #9 cell (neighboring cell to the reforming plate) along the cathode gas flow is shown in Fig. 2. The catalyst-loaded area in the reforming plate is shown in the figure. Temperature distribution in the

Fig. 2 Temperature distribution along cathode gas

first half of the cathode gas flow where reforming reaction does not occur is explained as below. In this region waste heat is removed by the cathode gas only. The oxidant increases its temperature as it deprives the heat of the stack. In MCFC stack it is believed that heat transfer between gas and stack hardware is relatively fast comparing to production rate of waste heat and that gas temperature is almost same as the one of neighboring hardware (ex. $< \sim 10^\circ\text{C}^{(4),(5)}$). By assuming that temperature increase of the stack along cathode gas flow is same as the one of cathode gas, and by applying energy balance to this region, average current density of this region is calculated as 116 mA/cm². This result seems reasonable because the average current density of this region should be less than the total average current density (150 mA/cm²). It is due to the lower operating temperature (cathode inlet) and the lower H_2 concentration (fuel outlet).

In the second half of the cathode flow, temperature increase almost stops especially at the catalyst-loaded region. Moreover, temperature difference in the stacking direction is obvious in this region (#6 vs. #9 cell). The difference of temperature is observed as 14°C. These two characteristics are caused by the cooling design of this region: reaction cooling as the main cooling in this area by the two plate reformers inserted in the stack. In principle reaction cooling does not require increase of temperature of coolant while it absorbs heat, because the heat removed is stored as chemical energy, not as sensible heat. By looking at the temperature distribution in the figure closely, temperature in the two catalyst-loaded regions is almost constant, while it increases between the two catalyst-loaded regions. These phenomena agree with the characteristics of the two different cooling methods: reaction cooling and sensible heat cooling.

4. Stack and System Performance

The relationship between cooling design and stack/system performance is summarized as below ;

1. Cooling design can affect stack performance through temperature distribution in a stack.

2. Cathode gas cooling needs more cathode gas than usually needed only for cathode reaction (fuel cell reaction). The extra gas for gas cooling is usually supplied by cathode gas circulation, which dilutes O_2 and CO_2 concentration of cathode inlet gas. Thus cooling design effects stack voltage.

3. Parasitic power required for cathode gas recycle decreases system efficiency.

Effects of cooling design are discussed, comparing PG-cooled stack with IR-cooled stack.

4.1 Cooling design and stack temperature

Typical expected distribution of stack temperature along cathode gas flow is shown in Fig. 3. The operating condition is assumed as 0.8 of fuel utilization, $559^\circ C$ cathode gas-in, and $110^\circ C$ of ΔT (cathode gas in/out). Distribution of the IR-cooled stack with cathode gas recycle is the one for the present IR-stack. The distribution is based on the test result of Stack (A). Effect of temperature distribution on stack performance can be evaluated by average stack temperature⁽⁶⁾. Average stack temperature of the present IR-cooled stack is $632^\circ C$. The reason why the average stack temperature is higher than the average of cathode in/out gas temperatures (i.e. $614^\circ C$) is the existence of flat temperature distribution at higher temperature range in the IR-cooled stack. This feature comes from a character of the IR-cooling.

On the other hand almost linear (slightly convex shape) distribution of stack temperature along cathode gas flow is expected at the PG-cooled stack of co-flow geometry^{(7),(4)}. At the same condition of Fig. 3, average stack temperature of the PG-cooled stack

Fig. 3 Expected temperature distribution

will be $618 - 624^\circ C$ ($4 - 10^\circ C$ higher than the average of linear distribution). Based on the experimental relationship between the average stack temperature and the stack voltage⁽⁶⁾, the present IR-cooled stack is to have higher cell voltage than the PG-cooled stack by $10 - 18$ mV.

By extending the above discussion, further improvement of the IR-MCFC is possible. In the present IR-MCFC stack, only 57% of the waste heat is removed by the IR cooling. If a stack can be cooled mainly by the IR cooling, more uniform temperature distribution and as a result higher average stack temperature can be obtained. By assuming that cathode gas flow rate is determined as the minimum flow rate for cell reaction only (no cathode gas recycle for stack cooling) and that major part of the cooling load is removed by the IR cooling, the distribution shown in Fig. 3 (no cathode recycle) is expected. In this case average stack temperature is $643^\circ C$ and 11 mV/cell gain over the present IR-MCFC stack is possible.

4.2 Cooling design and gas composition

The standard cooling operation of the present MCFC is to recycle a part of cathode outlet gas to inlet so that total flow rate of cathode inlet reaches enough flow rate to satisfy energy balance of the stack. The rest part of cathode outlet gas is exhausted from the recycle loop. To keep steady-state operation, fresh make-up air and CO_2 is added to this loop at cathode inlet. From stack voltage point of view, circulation of dilute outlet gas to inlet decreases stack voltage. This trend is true for both the IR-cooled stack and the PG-cooled stack. But this defect is not significant for the IR-cooled stack because cooling demand for cathode gas is about 42% of the one for the PG-cooled stack. Figure 4 shows the relationship between stack voltage and flow rate of cathode inlet gas (0.1 MPa, 0.5 MPa). In this test average stack temperature is kept constant for each run (about $629^\circ C$ for the run at 0.1 MPa and about $635^\circ C$ for the run at 0.5 MPa). Cell voltages obtained are at unsteady-state condition. Throughout the test, fresh make-up gas supply (O_2 , CO_2) to the recycle loop is kept constant: U_{O_2} (fresh) = 0.6, U_{CO_2} (fresh) = 0.76. The fresh CO_2 utilization of 0.76 is the one expected at a MCFC plant, where fuel utilization is set 0.8 and fresh CO_2 gas to the cathode inlet is fed by transferring CO_2 of anode exit gas through a burner. The fresh CO_2 gas in this test is simulated mixed gas of burner exhaust ($CO_2/N_2 = 60/40$ mol%). The flow rate of cathode inlet gas is changed by adjusting recycle rate in the recycle loop.

The required flow rate of cathode gas which satisfies energy balance of the stack of each cooling design is shown by the arrow with the name of the

Fig. 4 Cell voltage and cathode gas flow rate

stack design in the figure (at the condition of ΔT (cathode gas in/out) = 110°C). The operating voltage of the stack is given at the intersecting point of the corresponding arrow and the curve of cell voltage. From the test result, the stack voltage of the IR-MCFC (with cathode recycle) is to be higher than the one of the PG-cooled stack: 16 mV/cell at 0.1 MPa and 18 mV/cell at 0.5 MPa.

The test result in the figure tells that more voltage gain could be obtained if the stack could be operated without cathode gas circulation; at such condition cathode gas flow is 29.7 Nm³/h in Fig. 4. In order to realize such operating condition, new idea on stack cooling should be introduced to allow cathode gas flow to be decreased. In such an IR system without cathode gas recycle, voltage gain over the conventional IR-MCFC (with cathode gas recycle) is 40 mV/cell at 0.1 MPa and 24 mV/cell at 0.5 MPa.

4.3 Parasitic power for gas recycle

Parasitic power for cathode gas circulation is calculated by the standard equation of estimating blower-power based on adiabatic compressive work⁽⁹⁾. The calculated condition is that ΔP in the blower for the circulation is 0.01 MPa at 0.1 MPa operation and 0.02 MPa at 0.5 MPa. The calculated results are summarized in Table 4. The number in the brackets in the table is the virtual voltage loss of one 1 m²-cell equivalent to the parasitic power loss per one cell. At the atmospheric operation, parasitic power loss for

Table 4 Parasitic power for gas circulation

Operating Pressure	Conventional IR-MCFC	PG-cooled MCFC
0.1 MPa	0.058 kW/kWDC (41 mV loss)	0.167 kW/kWDC (116 mV loss)
0.5 MPa	0.0066 kW/kWDC (5 mV loss)	0.0344 kW/kWDC (27 mV loss)

Unit: mol/s						
NO.	1	2	3	4	5	6
CH ₄	1.0	0.218	0.516	0.002	2.667	1.067
H ₂ O	3.5	1.428	1.910	4.919	11.537	11.537
H ₂	0	1.353	0.275	0.578	4.200	1.000
CO	0	0.116	0.001	0.215	1.113	1.113
CO ₂	0	0.252	0.068	3.983	—	—
T (°C)	570	640	350	600	570	670
P (MPa)	0.51	0.50	0.47	0.49	0.50	0.49

Fig. 5 IR-MCFC system without cathode gas recycle

the PG-cooled stack is significant. Even at 0.5 MPa condition, the IR-MCFC stack has the advantage of 22 mV/cell over the PG-cooled stack.

If cathode gas circulation becomes unnecessary, further improvement of the system efficiency is expected comparing to the conventional IR-MCFC system (as the best case, 41 mV/cell gain at 0.1 MPa and 5 mV/cell gain at 0.5 MPa as virtual voltage loss).

5. MCFC System without Cathode Gas Recycle

From system efficiency point of view, MCFC system without cathode gas recycle is desired^{(1),(9),(10)}. For such a purpose one approach is to increase IR-cooling capacity. One idea to realize such a system is the system shown in Fig. 5⁽⁹⁾. In the system part of the reformed gas at the exit of the reforming plate is recycled to its inlet through the methanator on the circulation line. In the methanator H₂/CO₂ in the reformed gas is methanated via exothermic reaction, where the reformed gas is regenerated as a coolant. One example of operating condition around the stack is shown in the figure based on 1 CH₄-mol/s input to the system. As shown in the figure, about 0.5 CH₄

mol/s is recovered from the circulation loop and thus IR-cooling capacity is increased. According to this design cathode gas circulation for cooling the stack is not necessary. In this system waste heat generated around 650°C at the fuel cell is released at the methanator at 350 - 580°C. This waste heat is for example utilized to generate steam to be consumed in the system.

In this system IR stack can be operated mainly by the IR cooling. Therefore average stack temperature becomes higher and O₂ and CO₂ concentrations at the cathode inlet increase. As a result in this system IR stack can be operated at a higher voltage than the one in the present system (with cathode gas circulation): by 51 mV/cell at 0.1 MPa and by 35 mV/cell at 0.5 MPa higher based on the test results (Figs. 3 and 4).

As for the parasitic power for the gas circulation, the parasitic power for the reforming gas is necessary in the system of Fig. 5. Heat capacity (reaction heat + sensible heat) of the reforming gas as a coolant is 7 times higher than the one of the oxidant gas (sensible heat only). Moreover, the inlet temperature of the reformed gas at the blower is relatively low (350°C). Due to the above reasons parasitic power for the circulation of reforming gas in the new system is much smaller than the one for the cathode gas circulation in the conventional IR-MCFC system. In the new system (Fig. 5), parasitic power is estimated as 0.019 kW/kWDC ($\Delta P=0.04$ MPa) at 0.1 MPa and 0.0017 kW/kWDC ($\Delta P=0.04$ MPa) at 0.5 MPa; as virtual voltage loss for one 1-m² cell, the loss is equivalent to 14 mV at 0.1 MPa and 1.3 mV at 0.5 MPa.

6. Conclusion

Looking at good cooling performance of the IR-MCFC, energy balance and temperature distribution of the IR-MCFC is evaluated. In parallel the relationship between cooling design and stack/system performance is quantitatively studied. Based on the test results, the following conclusions are obtained;

1. In the present IR stack 57% of the waste heat is removed by the IR-cooling and thus the cooling load of the cathode-gas-cooling is decreased. As for the temperature distribution the IR-cooling establishes relatively flat distribution and increases average stack temperature.

2. The IR-cooling improves stack/system performance by higher average temperature, higher oxidant gas concentration, and less parasitic power.

3. By an appropriate system design, IR-MCFC

system without cathode gas circulation is possible. In such a system further improvement of stack/system performance is to be obtained.

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