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Short communication

Operation results of a 100 kW class reformer for molten carbonate fuel cell

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Abstract

The performance and operation results of the reformer that supplies synthesis gases to a 100 kW class molten carbonate fuel cell are reported. A CH_4 conversion ratio of 95.6%, a CO conversion ratio of 31.2%, a reforming reaction temperature of 745 °C and a produced hydrogen rate of 70.7 $Nm^3\,h^{-1}$ are obtained from a comparison of post gas analysis and theoretical estimation of thermodynamics at 87.6 h. To calculate the efficiency, the Cycle-Tempo 5.0 program is used. The thermal efficiency of the designed system is 61.1% and the real thermal efficiency of the system is 44.5% at 108 h. The low thermal efficiency is mainly attributed to supplying excess fuel to meet the outlet temperature of the reforming reactors. In the present system, the outlet temperature of the reforming reactors appears to be below the temperature required at the stack of molten carbonate fuel cell (MCFC), that is, over 580 °C. In order to maintain the outlet temperature of the reforming reactor over 580 °C, it is necessary to heat the reformed gases at the convection zone of combustion gases. For higher thermal efficiency, the combustion space and the excess fuel should be reduced until reaching minimum temperature of the surface of reforming tube, at which point has no influence on the CH_4 conversion ratio.
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Keywords: Tubular type reformer; Molten carbonate fuel cell (MCFC); *S/C* ratio (steam to carbon ratio); CH_4 conversion ratio; CO conversion ratio; Efficiency of reformer

1. Introduction

The fuel cell is a promising alternative power system because it can generate electricity with less fuel and emits fewer pollutants than the conventional power systems. Among fuel cells, the molten carbonate fuel cell (MCFC) has the most potential to scale up it for the use as power system because it can use various types of fuel. Natural gas (NG) was verified as a good fuel in the MCFC system, and additional gases such as coal gas and several hydrocarbon gases have been studied.

In this paper, we study hydrogen production from NG for 100 kW class MCFC. NG is converted into hydrogen rich gases through a reforming reactor, and the produced gases enter the anode inlet of the MCFC. The MCFC process does not need to refine synthesis gases, including CO and CO_2, because CO can react with carbonate ion, putting electrons out, and CO_2 can be recycled to the cathode inlet to produce carbonate ion. Therefore, the synthesis gases are directly supplied to the anode inlet of the MCFC stack.

The catalysts used here are steam reforming catalysts supplied by Haldor Topsoe. The company says that the base material of the steam reforming catalyst is magnesium aluminate, which is a less acidic oxide than pure alumina as a major constituent of other commercially available steam reforming catalysts. The activity and non-acidic nature are sufficient to ensure carbon free operation when operating on light higher hydrocarbons [1].

The compositions of domestic NG are shown in Table 1. Table 1 shows that NG includes about 10% slightly higher hydrocarbons, with the exception of methane. In many cases of reforming reactions of fossil fuels, carbon formation is considered

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Nomenclature

Cp	molar heat capacity (J mol^{-1} K^{-1})
E	thermal efficiency on the base of inlet and outlet of reformer
E'	thermal efficiency when using non-reacted CH_4
F_{CH_4}	flow rate of non-reacted methane among produced synthesis gas (kg s^{-1})
F_{H_2}	flow rate of hydrogen among produced synthesis gas (kg s^{-1})
$F_{NG\text{-}feed}$	flow rate of NG for reforming reaction (kg s^{-1})
$F_{NG\text{-}fuel}$	flow rate of NG for burner (kg s^{-1})
ΔG	Gibbs free energy change (J mol^{-1})
HHV_{CH_4}	higher heating value of methane among produced synthesis gas (55558.9 kJ kg^{-1})
HHV_{H_2}	higher heating value of hydrogen among produced synthesis gas (141802.0 kJ kg^{-1})
HHV_{NG}	higher heating value of natural gas (54467.4 kJ kg^{-1})
ΔH	enthalpy change (J mol^{-1})
K_1, K_2, K_3, K_4, K_5	chemical equilibrium constants of reforming reactions of hydrocarbons
K_{10}	chemical equilibrium constant of CO shift reaction (3)
P	partial pressure of element
R	gas constant (8.314 J mol^{-1} K^{-1})
s	steam to carbon ratio, S/C ratio
T	temperature (K)
x	NG feed rate (Nm^3 h^{-1})
y	mole fraction of element
ε_1	CH_4 conversion ratio of CH_4 reforming reaction (1)
ε_2	CO conversion ratio of CO shift reaction (3)

as a serious by-product. With a methane content of about 90%, the formation of solid carbon can be avoided by increasing the reactor temperature and/or the steam to carbon (S/C) ratio [2]. Some papers of [3,4] note that higher hydrocarbons, similar to ethane and propane, are susceptible to non-catalytic thermal cracking, thereby producing olefins, which are precursors for coke formation. However, the catalysts used in this study are reported as being effective in suppressing carbon deposition, as noted above. In another paper [5], it is noted that propane dehydrogenation actually already occurs near 800 K, as shown in the equation below, and the conversion of produced propene and steam grows steadily above 550 K giving rise to CO, CO_2 and hydrogen as the largely predominant products.

$$C_3H_8 \leftrightarrow C_3H_6 + H_2$$

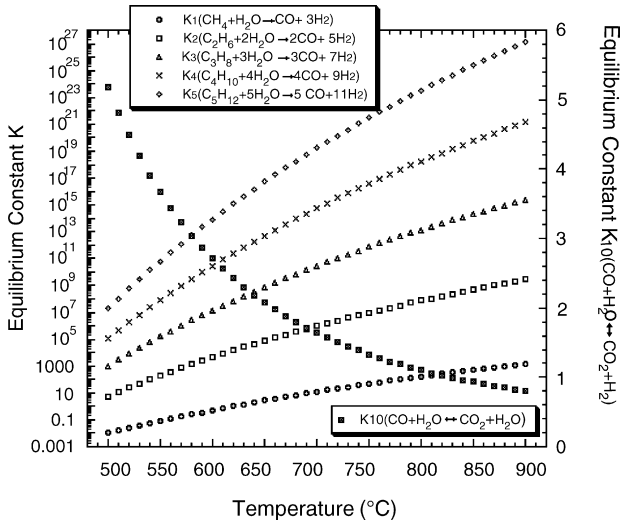

Fig. 1. Estimation of chemical equilibrium constants of reforming reactions of natural gas over temperature.

$$C_3H_6 + 3H_2O \leftrightarrow 3CO + 6H_2$$

$$3CO + 3H_2O \leftrightarrow 3CO_2 + 3H_2$$

Therefore, we consider that higher hydrocarbons are ultimately converted to hydrogen rich gases through olefins without taking coke formation into account.

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \quad (1)$$

$$C_nH_m + nH_2O \leftrightarrow nCO + (n + m/2)H_2 \quad (2)$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \quad (3)$$

Reaction (2) is assumed to be predominant because the chemical equilibrium constants in the reforming reactions of higher hydrocarbons are higher than those of CH_4, as shown in Fig. 1. Therefore, it is assumed that the reforming reactions of NG might be similar to Eqs. (1) and (2) above. After the reforming reactions of (1) and (2), the CO shift reaction (3) occurs.

In this paper, the amounts of gases produced from actual operation and the theoretical values by thermodynamic calculation are compared. Also, the thermal efficiencies of the operating system and the designed system are compared. For the calculation of efficiency, the Cycle-Tempo 5.0 program supplied by Delft university of Technology in the Netherlands is used.

2. The theoretical formula of the NG reforming reaction

In order to verify the performance of the reformer, it is necessary to compare the operation results with the theoretical results in the thermodynamics. The hydrogen amount that is required

Table 1
Composition of natural gas

Element	CH_4	C_2H_6	C_3H_8	n-C_4H_{10}	i-C_4H_{10}	N_2	i-C_5H_{12}	n-C_5H_{12}
Mole%	90.53	5.97	2.26	0.53	0.46	0.22	0.03	0.01

theoretically is calculated based on the chemical equilibrium constants over temperature; the conversion ratios of CH_4 and CO are calculated from the chemical equilibrium formulation. The chemical equilibrium constants are also obtained from the sum of heat capacity constants, the enthalpy and Gibbs energy. The following Eq. (4)–(7) are used [6]

$$\frac{Cp}{R} = A + BT + CT^2 + DT^{-2} \tag{4}$$

$$\mathrm{d}H = Cp\,\mathrm{d}T \tag{5}$$

$$\mathrm{d}\left(\frac{\Delta G}{RT}\right) = -\frac{\Delta H}{RT^2}\,\mathrm{d}T \tag{6}$$

$$\ln K = -\frac{\Delta G}{RT} \tag{7}$$

where A, B, C and D are constants of heat capacity of elements [6], and the sum of heat capacities and the change values of enthalpy and Gibbs free energy of the elements involved in reforming and CO shift reactions are calculated by using the constants A, B, C and D.

The estimations of chemical equilibrium constants of reactions relating to NG are shown in Fig. 1, where K_1–K_5 and K_{10} are the chemical equilibrium constants of each reaction, respectively. Fig. 1 shows that the hydrocarbon compounds above C_2 are easily converted into hydrogen and CO.

The gas compositions at the equilibrium state have to be calculated from the equilibrium constants. The estimations of gas flow rates are shown in Tables 2 and 3, where x is the flow rate of NG, s the steam to carbon ratio, ε_1 the CH_4 conversion ratio and ε_2 is the CO conversion ratio. Using the gas flow rate of Tables 2 and 3, the mole fraction of each element, i.e. y, is

Table 2
Estimation of gas flow rate at the equilibrium state after reforming reaction

Element	Initial flow	Flow at equilibrium
CH_4	0.9053χ	$0.9053\chi(1-\varepsilon_1)$
C_2H_6	0.0597χ	–
C_3H_8	0.0226χ	–
C_4H_{10}	0.0099χ	–
H_2O	$s\chi$	$s\chi-\chi(0.9053\varepsilon_1+Z)$
CO	–	$\chi(0.9053\varepsilon_1+Z)$
H_2	–	$\chi(3\times0.9053\varepsilon_1+Y)$
C_5H_{12}	0.0003χ	–

where $Z = 2\times0.0597+3\times0.0226+4\times0.0099+5\times0.0003$; $Y = 5\times0.0597+7\times0.0226+9\times0.0099+11\times0.0003$.

Table 3
Estimation of gas flow rate at the equilibrium state after CO shift reaction

Element	Initial flow	Flow at equilibrium
CO	$\chi(0.9053\varepsilon_1+Z)$	$\chi(0.9053\varepsilon_1+Z)(1-\varepsilon_2)$
H_2O	$s\chi-\chi(0.9053\varepsilon_1+Z)$	$s\chi-\chi(0.9053\varepsilon_1+Z)(1+\varepsilon_2)$
CO_2	0	$\chi(0.9053\varepsilon_1+Z)\varepsilon_2$
CH_4	0.9053χ (1-ε_1)	$0.9053\chi(1-\varepsilon_1)$
H_2	$\chi(3\times0.9053\varepsilon_1+Y)$	$\chi(3\times0.9053\varepsilon_1+Y+(0.9053\varepsilon_1+Z)\varepsilon_2)$

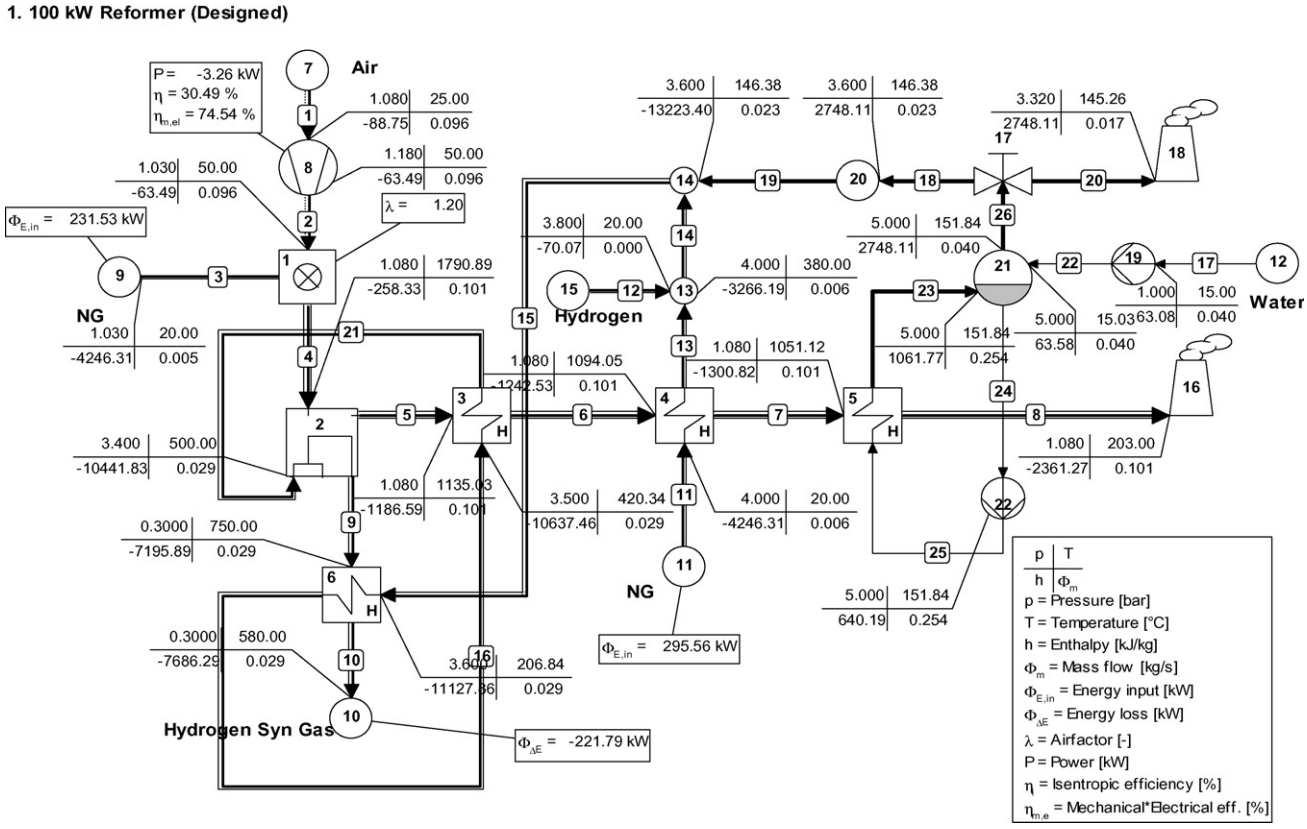

Fig. 2. 100 kW class reformer (designed), apparatus 1: combustor; 2: reformer; 3, 4, 5, 6: heat-exchanger; 8: air blower; 13: desulfurizer; 19: pump feeding water; 21: steam drum; 22: pump recycling water.

obtained and then inserted in the relating Eqs. (8) and (9). ε_1, ε_2, gas flow rate and mole fractions of each element are then obtained.

$$K_1 = \frac{P_{CO} \times P_{H_2}^3}{P_{CH_4} \times P_{H_2O}} = P^2 \frac{y_{CO} \times y_{H_2}^3}{y_{CH_4} \times y_{H_2O}} \quad (8)$$

$$K_{10} = \frac{P_{H_2} \times P_{CO_2}}{P_{CO} \times P_{H_2O}} = \frac{y_{H_2} \times y_{CO_2}}{y_{CO} \times y_{H_2O}} \quad (9)$$

3. Configuration of the reforming system

As (1) and (2) reactions above occur favorably at temperatures in excess of 700 °C, external heat is necessary for the reforming reaction. In this study, the external heat is provided from a central burner that uses the same fuel of NG. For the operation of the reformer, liquefied natural gas with a pressure of 50 kg cm^{-2} is used. For the process line, the pressure is lowered to 3 kg cm^{-2} which is the operating pressure of the designed MCFC system, and to 0.5 kg cm^{-2} for the burner line. The amount of hydrogen required for a 100 kW class MCFC is about 100 Nm^3 h^{-1}.

The reformer connected to the MCFC stack is a conventional type and is comprised of a top-fired burner in the center and 16 tubular reactors surrounding the central burner at equal distance, including the steam reforming catalysts.

NG consists of more than 90% of CH_4, more or less higher hydrocarbons and sulfur compounds as odorants. Because the sulfur among sulfur compounds deactivates reforming catalysts, a desulfurizer is necessary to eliminate sulfur before the reforming reactor. As the additional units, there are heat-exchangers, a drum and driving devices. The heat-exchangers play roles in heating cold NG and water that is fed to reforming reactors. The drum is used in storing the excess heat. The configuration and flow diagram are shown in Fig. 2. The gases produced from the reforming reactor are analyzed through gas chromatography DS6200 supplied by Donam Instruments Inc. in Korea.

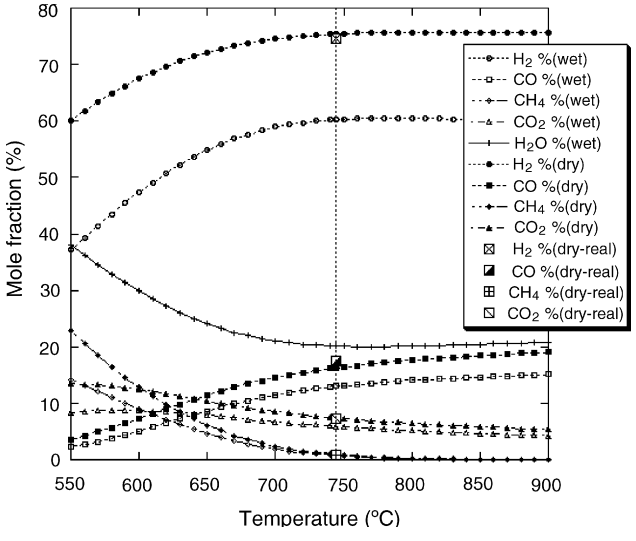

Fig. 3. Post gas analysis (large dots) at operation time of 87.6 h and theoretical gas composition (%) (small dots and lines) at NG feed 20.3 Nm^3 h^{-1}, *S/C* 2.6 and pressure 1.65 kg cm^{-2}.

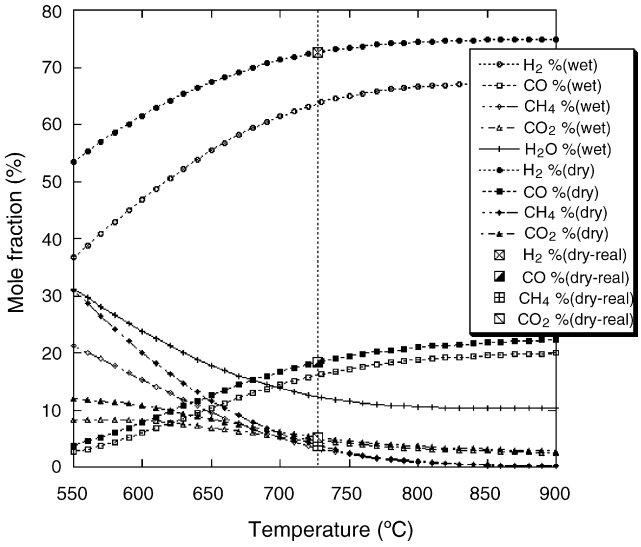

Fig. 4. Post gas analysis (large dots) at operation time of 108 h and theoretical gas composition (%) (small dots and lines) at NG feed 28.9 Nm^3 h^{-1}, *S/C* 1.78 and pressure 1.97 kg cm^{-2}.

4. The operation results of NG reforming

Comparisons of gas compositions analyzed and theoretical values in the operation conditions at operation times of 87.6 and 108 h are presented in Figs. 3 and 4, respectively. The drawing lines of theoretical gas compositions in Figs. 3 and 4 are graphs of the results obtained from Eqs. (8) and (9) at any condition of NG feed rate, *S/C* ratio and pressure according to the variation of temperature.

Figs. 3 and 4 include the results of gas compositions with and without water. The result of real gas analysis is shown as large dots. The temperature of the reactor is obtained when the large dots are the closest to the theoretical graphs. At the operation time of Fig. 3, the temperature of reactor is regarded as 745 °C, the CH_4 conversion ratio 95.6%, the CO conversion ratio 31.2% and the amount of produced hydrogen is calculated as 70.7 Nm^3 h^{-1}. At the time of Fig. 4, the temperature is regarded as 727 °C, the CH_4 conversion ratio 82.6%, the CO conversion ratio 22.0% and hydrogen rate 87.0 Nm^3 h^{-1}.

5. Thermal efficiencies of the designed system and a real operating system

The 100 kW class reformer was not operated as a full load, but at about 87%. Due to improper calibration of the steam flow meter, sufficient steam greater than a *S/C* ratio of 3 was not supplied, and hence a smaller amount of gas was produced than the theoretical value of *S/C* of 3.

In order to calculate the thermal efficiency of system, the Cycle-Tempo 5.0 program is used. The definition of the thermal efficiency is given as follows [7].

$$\text{Efficiency}\% = \frac{F_{H_2} \times \text{HHV}_{H_2}}{(F_{NG-feed} + F_{NG-fuel}) \times \text{HHV}_{NG}} \times 100 \quad (10)$$

The 100 kW class reforming system is configured as shown in Fig. 2. The configuration, the conditions for operation, the

Table 4
Gas composition of each pipe in Fig. 2

Gas	#pipe							
	1,2	3,11,13	4,5,6,7,8	9,10	12	14	15,16,21	19
N_2	0.7729	0.0015	0.7128	0.0002		0.0014	0.0003	
O_2	0.2075		0.0332					
H_2O	0.0101		0.1626	0.3153			0.7791	1.0000
Ar	0.0092		0.0085					
CO_2	0.0003		0.0811	0.0646				
CH_4		0.8998		0.0045		0.8314	0.1836	
C_2H_6		0.0629				0.0581	0.0128	
C_3H_8		0.0254				0.0235	0.0052	
C_4H_{10}		0.0101				0.0093	0.0021	
C_5H_{12}		0.0003				0.0003	0.0001	
H_2			0.0005	0.5239	1.0000	0.0760	0.0168	
CO			0.0013	0.0915				
Average mole	28.85	18.10	27.90	12.22	2.02	16.87	17.76	18.02
LHV (kJ kg^{-1})	0.00	49260.67	35.10	12785.15	119979.50	49903.04	10471.77	0.00
HHV (kJ kg^{-1})	0.00	54467.58	35.94	14703.68	141801.00	55260.87	11596.07	0.00

estimated temperatures and pressures of the designed system are shown in Fig. 2 and the gas compositions of each pipe are shown in Table 4. To calculate the thermal efficiency of the designed and operating systems, the values of the inserted NG, produced H_2 and non-reacted methane are required, as indicated in Table 5. The flow rates of produced hydrogen and non-reacted methane are obtained from Eqs. (8) and (9). The thermal efficiency of the system is obtained using Eq. (10). The thermal efficiency of the designed system is calculated as 61.1%, the efficiency at the operation time of 87.6 h is 42.6% and at 108 h, 44.5%.

$$E_{\text{designed}} = \frac{0.00251 \times 141802.0}{(0.0047 + 0.006) \times 54467.4} \times 100 = 61.1\% \quad (11)$$

If non-reacted methane is used again as a fuel of the burner, the thermal efficiencies become higher as follows [7].

$$\text{Efficiency}_{\text{using-nonreacted CH}_4} = \frac{F_{H_2} \times \text{HHV}_{H_2}}{(F_{\text{NG-feed}} + F_{\text{NG-fuel}}) \times \text{HHV}_{\text{NG}} - F_{CH_4} \times \text{HHV}_{CH_4}} \times 100 \quad (12)$$

$$E'_{\text{designed}} = \frac{0.00251 \times 141802.0}{(0.0047 + 0.006) \times 54467.4 - 0.00017 \times 55558.9} \times 100 = 62.1\% \quad (13)$$

Table 5
NG feed and fuel rate, product H_2, CH_4 rate on the designed and operating systems

	Designed	At 87.6 h	At 108 h
NG feed rate (kg s^{-1})	0.0060	0.00455	0.00649
NG fuel rate (kg s^{-1})	0.0047	0.006103	0.006046
Product H_2 rate (kg s^{-1})	0.00251	0.001742	0.002143
Non-reacted CH_4 rate (kg s^{-1})	0.00017	0.000208	0.001148
E (%)	61.1	42.6	44.5
E' (%)	62.1	43.4	49.1

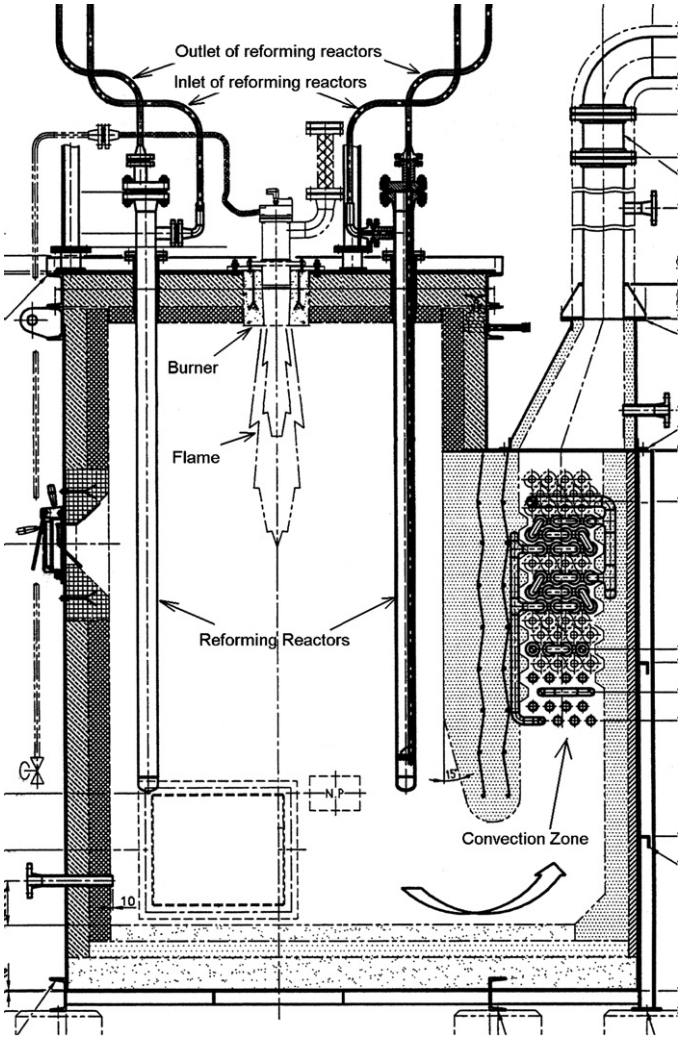

Fig. 5. The assembly of burner and reforming reactors.

There are two reasons accounting for why the thermal efficiency of the operating system is lower than the designed value. One minor reason is that sufficient steam is not supplied due to the improper calibration of the steam flow meter. Thus, very little synthesis gas is produced, which affects the efficiency. At an operating time of 87.6 h, the *S*/*C* ratio was 2.6. However, if the steam had been supplied at a *S*/*C* of 3.8, the value in the designed system, the thermal efficiency would have been 47.7%, compared to an efficiency of 42.6%.

The more significant reason is that excessive fuel is used to meet the necessary temperature of the outlet of the reforming reactors. The actual temperature measured at the outlet of reforming reactors was less than 500 °C, contrasting with to the design value of 750 °C. Thus, more fuel than that of the design was used, thereby affecting the thermal efficiency.

But in this system, the outlet temperature of reformed gas does not appear to reach 750 °C. At the real operation time, the inlet temperature of the reforming reactors was roughly 390 °C and the inlet gases were supplied at the region outside of the combustion space, as shown in Fig. 5. Although the outlet temperature of the reformed gases is higher than 700 °C, the temperature is mitigated through exchange of heat with inlet gases into the reforming reactors and reaches an intermediate temperature value.

In our system connected with a MCFC stack, the outlet temperature of reformed gases is required to be at least 580 °C, even though the designed value is 750 °C. This is because the reformed gas is supplied into the stack of the MCFC. In order to heighten the outlet temperature of reformed gases, re-design of the system is required.

6. The necessity of re-designing surrounding units and burner

In our system, the fraction of absorbed energy to combustion energy was about 21% and considerable energy, except the absorbed energy, was exhausted via the convection zone. Therefore, in order to heighten the outlet temperature of the reformed gases, it is reasonable to use the energy of the convection zone. That is, pipe 9 and heat-exchanger 6 in Fig. 2 should be considered to be located at the convection zone. In Fig. 6, the re-designed flowchart is shown. Heat-exchangers 6, 3, 4 and 5 are located at the convection zone in Fig. 5.

We supplied the fuel in excess of the necessary amount. As the fuel of combustion space is reduced, the heat transmitted to the reforming reactors becomes low and so, the CH_4 conversion of reforming reaction can be affected. But in our operations, the fuel was supplied more than the amount to have no influence on the CH_4 conversion. Therefore, if the amount of fuel is decreased to a point where the CH_4 conversion is not influenced, the thermal efficiency will be increased.

However, the authors still consider the designed efficiency to be low. Two methods to enhance the efficiency can be considered; reducing the combustion space and using the present combustion space more efficiently by using a swirl burner and less fuel.

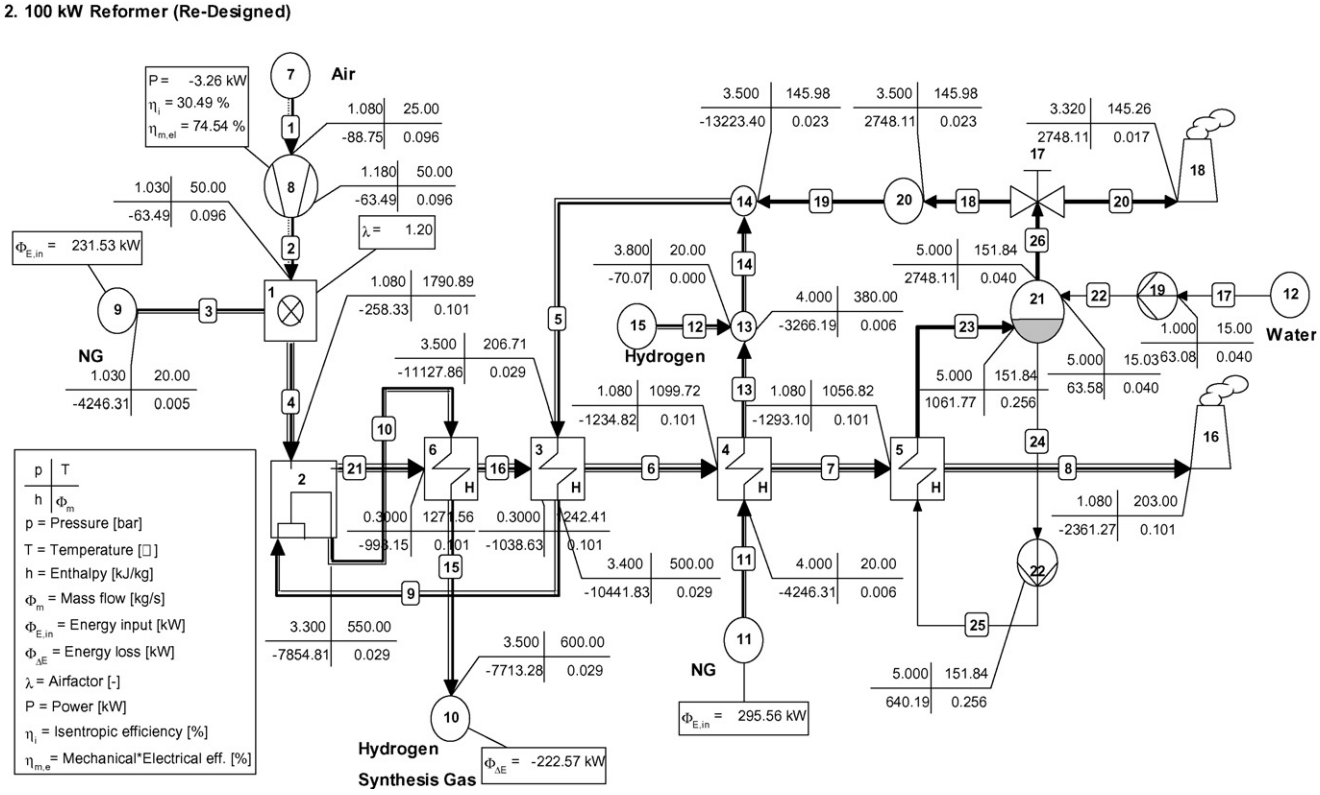

Fig. 6. 100 kW class reformer (re-designing), apparatus 1: combustor; 2: reformer; 3, 4, 5, 6: heat-exchanger; 8: air blower; 13: desulfurizer; 19: pump feeding water; 21: steam drum; 22: pump recycling water.

The burner vertically spouts flame downward in our reformer system, as shown in Fig. 5. The purpose of vertical flames is to avoid the transmission of heat with very high temperatures to the reaction tubes, even though the combustion space is not used sufficiently. The larger the combustion space, the less heat is transmitted at the surfaces of the reforming tubes. Therefore, when the combustion space is large, in order to supply the heat required at the surfaces of the reforming tubes, a large amount of fuel must be supplied. However, for the thermal efficiency, the combustion space and the fuel should be reduced until the minimum temperature of the surface of the reforming tube is obtained, at which point has no influence on the CH_4 conversion ratio.

In the case of using a swirl burner, the present combustion space can be used. If the swirl angle of the burner is enlarged, the radius of the flame will be large and the vertical length of the flame short. By adjusting the swirl angle of the burner, the surface temperature of the reforming tube can reach the required temperature, despite that less fuel is supplied. It is necessary to consider whether the temperatures of the surface of the reforming tube and the end tip of the reforming tube are within the limit of the temperatures required.

Re-designing the combustion space involves several considerations such as temperatures of the surface and the end tip of the reforming reactor and the calorie required for heat-exchange. To investigate these aspects, the results of computation and real operations should be mutually considered. These works will be carried out in the future.

7. Conclusions

As a characteristic of the MCFC, it is not necessary to eliminate CO and CO_2 from the reformed gas, and thus the reformed gas is directly supplied to the anode inlet of the MCFC. The 100 kW class reformer consists of a central top-fired burner and 16 tubular reforming reactors surrounding the burner at the same distance. Thermodynamically, the theoretical amount is compared with the result of post gas analysis, and the CH_4 and CO conversion ratios and the temperature of the reactor are estimated at the conditions of the operation time. At an operation time of 87.6 h, CH_4 conversion is 95.6%, CO conversion 31.2%, and the temperature of the reactor is estimated as 745 °C.

To calculate the thermal efficiency of the system, Cycle-Tempo 5.0 program is used, and the thermal efficiency of the designed system is revealed to be as 61.1% on the basis of HHV while that of the system at the operation time of 108 h is 44.5%. The thermal efficiency of the operating system is lower than the designed efficiency partially because of improper calibration of the steam flow meter and largely due to excessive supply of fuel.

In the present system, the outlet temperature of the reforming reactors appears to be below the temperature required at the stack of the MCFC, that is, greater than 580 °C. In order to maintain the outlet temperature of the reforming reactor beyond 580 °C, the reformed gases should be heated at the convection zone of the combustion gases.

For higher thermal efficiency, the combustion space and the excess fuel should be reduced until the minimum temperature of the surface of the reforming tube is obtained, at which point has no influence on the CH_4 conversion ratio.

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