Development of Microwave-Matrix Partial Oxidation Reformer

Young Nam Chun*[‡], June An*

* Department of Environmental Engineering, Chosun University, #309, Pilmundaero, Dong-gu, Gwangju, 61452,

Rep. of Korea

(ynchun@chosun.ac.kr, soyobi@naver.com)

[‡]Corresponding Author; Young Nam Chun, Department of Environmental Engineering, Chosun University, #309, Pilmundaero, Dong-gu, Gwangju, 61452, Rep. of Korea, Tel: +82-62-230-7156, ynchun@chosun.ac.kr

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Abstract- This study proposes a novel microwave-matrix reformer (MMR) to convert biogas, a greenhouse gas that causes climate change, into high-quality fuel energy. To show its performance potential, it has identified the characteristics of biogas conversion and product gas for O_2/C ratio, steam feed, reformed gas recirculation, and water supply in the recirculation pipe, which affect CH₄ and CO₂ conversions. The reforming characteristics for different variables were achieved and the results are as follows. The optimal conditions of the MMR were 100% recirculation rate and 10 mL/min water feed rate when O_2/C ratio and total gas supply were 1 and 30L/min, respectively. Under these conditions, CH₄ conversion was 68.6%, CO₂ conversion was 37.9%, H₂ selectivity was 88.1%, and H₂/CO was 1.54, which were resonable for applying to stack for Solid Oxide Fuel Cell.

Keywords Partial oxidation reforming, biogas conversion, hydrogen production, SOFC stack, greenhouse gas.

1. Introduction

Methane and carbon dioxide are the components of a biogas from bioreactor. The both gases are known main greenhouse gas affected world climate change. Technologies for converting the biogas into useful fuel energy include thermal conversion, electro-chemical conversion, and so on. [1-5 3 5] Some of the thermal conversion and electro-chemical conversions have been applied to the field.

Thermal conversion technology includes steam reforming [6], dry reforming [7,8], and partial oxidation reforming [9-11 **11**]. Although the steam reforming has already been put to use for industrial applications, dry reforming is recognized as relatively more attractive. The reason is why the dry reforming could be reduced the carbon dioxide emission, a major greenhouse gas, and enables more efficient energy conversion. Unlike steam reforming and dry reforming, which are endothermic reactions, partial oxidation reforming is an exothermic reaction thus involves low energy consumption. [12,13]

Auto-thermal reforming combines catalytic partial oxidation reforming with steam reforming (or carbon dioxide reforming). This has the advantage of being able to produce large volumes of hydrogen, but involves matters such as catalyst poisoning and the cost of steam production. In order to solve these problems, a more advanced new technology needs to be developed.

The research tried to develop a new type of reformer that operates with microwave heating and thermal regeneration. Matrix combustion is the technology that uses radiant heat generated from high-temperature pores cavity to heat incoming gas mixture to increase the combustion stability. The premixed fuel-air mixture in-flowing in through the porous matrix burns while passing through the reacting zone, and some of the heat energy is accumulated in matrix to enable stable combustion in high-energy environment. [14-16] This type of reformer in which matrix surface combustion and the thermal regeneration in the microwave-heating receptors are combined maintains a porous matrix shape with a threedimensional volume in the reformer chamber, and the microwave irradiated into it heats the receptors, thus allowing stable and efficient partial oxidative combustion reforming.

The study achieved the reforming characteristics for O_2/C ratio, steam supply, reformed gas recirculation rate, and recirculated water supply when the biogas is supplied. It further suggests the optimal operating conditions for the novel microwave-matrix reformer based on the reforming performance for each variable.

2. Experimental Setup

Fig. 1 shows the configuration of a Microwave-Matrix Reformer (MMR) for producing hydrogen, which can apply an SOFC stack and uses biogas as its fuel.

Microwave-matrix reformer consists of a threedimensional rectangular porous material matrix in a rectangular chamber, and a silicon carbide (SiC) porous body, which is a microwave receptor, is installed at the center. The microwave-radiated magnetron is installed on the chamber surface in alignment with the center surface of the receptor. In addition, a recirculation pipe through which some gas from the reformer outlet is re-supplied to the inlet is installed, and the recirculation amount is controlled by a valve.

In the gas-air supply line, methane and carbon dioxide, which are biomass gases, and air or steam, which are oxidizing agents, were respectively supplied. They were first mixed in a venturi mixer, and then supplied to the MMR. Methane and carbon dioxide were supplied at a discharge pressure of $2kg_{f}/cm^{2}$ from each cylinder, and steam was supplied after the water supplied from the water pump was converted into steam in the steam generator. The compressor supplies compressed air (7kg_{f}/cm^{2}), which first passes through a surge tank, and

through a water trap. The flow rates of process gas and combustion air were each controlled by MFC. As for the water supply among the recirculation generated gas, its micro flow rate was adjusted in the water pump before the water was supplied to top of the recirculation pipe.

The electric power was supplied by power supply (model: LUP1200Q, Korea), and a voltage probe (model: P6015, Tektronix, USA), a current probe (model: A6303, Tektronix, USA), an amplifier (model TM502A, Tektronix, USA) measured electrical properties.

The measurement and analysis lines are each equipped with sampling ports (S_i , S_o) to collect the inflowing mixture and converted gas at the inlet and the exit of the MMR. The product gas passes through glass wool and cooler before it is sampled by a suction pump. As for the sampled gas, CO₂, CH₄, H₂, CO, etc., are analyzed by GC-TCD (model CP-4900, Varian, Netherland), an analysis device. The gas temperature in the MMR was measured by thermocouple and data logger (model: FLUKE 2625A HYDRA, Japan).



Fig. 1. Schematic of an experimental setup for microwave-matrix reformer.

3. The Experimental Method

To develop a microwave-matrix reformer, a partial oxidation combustion reformer that operates with microwave heating and thermal regeneration was designed and fabricated, and an experimental study was conducted. The experimental range for each variable is shown in Table 1, and the reference condition is the standard condition for each experimental parameter.

To describe the experimental method, the biogas, a reforming fuel, and air, a partial oxidizer, were each adjusted to a certain air-fuel ratio by the MFC, mixed in a venturi gas

mixer, and then, supplied to the MMR while the total gas feed rate was fixed at 30 L/min. As for the reformed gas recirculation experiment, it was conducted by regulating the ratio of the actual supply amount to the maximum amount supplied for recirculation with a value. The amount of steam supplied was already converted into steam in the steam generator after the calculated amount of water was adjusted by the water pump. The power supply to the microwave was constant at 1 kW.

GC (CP-4900, Varian, the Netherlands) was used for product gas analysis. H₂, CO, O₂, N₂, and CH₄ were analyzed with Molecular Sieve capillary column (model: MS 5A, Varian, the Netherlands), and CO₂, C₂H₄, C₂H₂, C₂H₆, and C₃H₈ were analyzed with PoraPLOT capillary column (model: PPQ, Varian, the Netherlands).

Table 1. Experimental condition and range of test variables

Variables	O ₂ /C ratio	Steam feed rate (L/min)	Recirculation rate (%)	Water feed rate (mL/min)
Test ranges	0.8-1.2	5, 10, 15, 20 ¹⁾	50, 100	5, 10, 15, 20
Reference conditions	1.1	10 ¹⁾	100	10

¹⁾ These amounts indicate the water supply which is not calculated into the steam for comparison with the water feed rate.

 CH_4 conversion and CO_2 conversion was calculated by equation (1). [17]

$$CE(\%) = \frac{[PG]_{input} - [PG]_{output}}{[PG]_{input}} \times 100$$
(1)

where, $[PG]_{input}$ is the feed rate (L/min) of the injected CH₄ and CO₂, and $[PG]_{output}$ is the feed rate (L/min) of the discharged CH₄ and CO₂.

The H_2 selectivity and H_2 /CO ratio were calculated using equations (2) and (3). [17]

$$S_{H_2}(\%) = \frac{H_2 \operatorname{Produced}(\operatorname{mol}/s)}{2 \times CH_4 \operatorname{converted}(\operatorname{mol}/s)} \times 100$$
(2)

$$\frac{H_2}{CO} = \frac{H_2 \text{ produced (mol/s)}}{CO \text{ produced (mol/s)}}$$
(3)

The energy conversion efficiency (ECE) was calculated as in the following equation (4). [18]

$$ECE(\%) = \frac{n_{H2} \times LHV(H_2) + n_{co} \times LHV(CO)}{MWP + LHV(fuel) \times fuel(injected)} \times 100$$
(4)

where, n_i refers to the moles of the reformed gas i(mol); fuel(injected) the moles of CH₄ converted (mol); MWP the input power in kW; LHV the lower heating value of product gases in kJ/mol.

4. Results and Discussions

To develop a microwave-matrix reformer, a study was conducted on each of the O_2/C ratio, steam supply amount, recirculation rate, and water supply amount in the recirculation gas, which are major factors influencing biogas reforming. The results are as follows.

Biogas is a mixed gas generated from anaerobic digestion of wastewater sludge, food waste, livestock manure, and so on. These gas components contain 45-75% CH₄, 25-55% CO₂, 0-25% N₂, 0.01-5% O₂, and small amounts of H₂S and NH₃.[19] As such, biogas has a ratio of CH₄ and CO₂ that varies depending on its sources and contains various other components. For the purpose of testing the application of reforming biogas, the study applied simulated gas with fixed methane and carbon dioxide ratios of 65% and 35%, respectively to the newly proposed microwave-matrix reformer.

4.1. The Influence of the O2/C Ratio

Fig. 2 shows test results when the main component of the biogas fuel is supplied as a mixture of CH₄, CO₂, and air, which is an oxidizing agent, the total gas supply is 30 L/min, and the O₂/C ratio is changed from 0.8 to 1.2. As experimental variables (see Table 1), steam supply and recirculation are not performed.

As shown in Fig. 2(a), as O_2/C ratio increased, CH_4 conversion and CO_2 conversion increased. When the O_2/C ratio was 1.1, the maximum values were 79.1% and 43.6%, respectively, and then they decreased again. The CH_4 conversion increases as the air supply increases because the methane partial oxidation reforming reaction (equation 5) is activated to decrease CH_4 and increase H_2 and CO_2 . The increase in CO_2 conversion is due to the reduction of CO_2 by the dry reforming reaction (equation 6). That is, as the O_2 concentration increased, the methane oxidation reaction increased. And when the reformer temperature rose, the reactivity of the dry reforming reaction increased.

 $CH_4 + 1/2O_2 \rightarrow CO + 2H_2 \quad \Delta H_{298} = -38kJ/mol$ (5)

$$CH_4 + CO_2 \rightarrow 2CO + 2H_2 \quad \Delta H_{298} = +260.5 \text{kJ/mol}$$
(6)

The reason that the CH₄ conversion decreased after reaching the maximum value is because the CH₄ and CO₂ conversions decreased as the drying reaction (equation 6) decreased in the oxidizing environment. In the case of CO₂, which is also increased by the oxidation reaction of already converted CO (equation 7), the CO₂ conversion seems to have a slightly larger decrease than the CH₄ conversion.

$$\text{CO} + 1/2\text{O}_2 \rightarrow \text{CO}_2 \qquad \triangle \text{H}_{298} = -283 \text{ kJ/mol.}$$
(7)

The H₂/CO also closely followed the pattern shown in the

 CH_4 and CO_2 conversions, decreasing after registering the maximum value of 1.42 at the O_2/C ratio of 1.1. The H_2/CO increases because the methane partial oxidation reforming (equation 5) is enhanced in a reduction environment. The H_2/CO decreased after showing the maximum value because the amount of H_2 decreased by the hydrogen oxidation reaction (equation 8) below as the amount of oxygen increased.

$$H_2 + 1/2O_2 \rightarrow H_2O$$
 $\triangle H_{298} = -285.8 \text{ kJ/mol}$ (8)

Fig. 2(b) shows the H₂ selectivity and lower heating value (LHV). The H₂ selectivity showed a similar pattern as predicted from H₂/CO pattern, and when O_2/C ratio was 1.1, the maximum value was 58.6. The lower heating value showed a larger value when the O_2/C ratio value was smaller. This is because CH₄, the feed fuel, remained in the product gas without being converted, and the amounts of the converted combustible components of CO, H₂, CH₄, and THCs (C₂H₂, C₂H₄, C₂H₆) were small. However, at the moment when O_2/C ratio is 1.1 and CH₄ and CO₂ conversions are maximized, the heating value is slightly increased due to the increase of combustible gas.



Fig. 2. Effect of O₂/C ratio. (a) CH₄, CO₂ conversions and H₂/CO (b) H₂ selectivity and HHV(Lower Heating Value).

4.2. The Effects of Steam Supply

Fig. 3 depicts how steam generated by steam generator blends with incoming methane and air in a venturi mixer before it enters the microwave-matrix reformer. At this time, steam was supplied in an amount of 5, 10, 15, and 20 mL/min, respectively, and otherwise, the test conditions to the standard conditions as shown in Table 1.

Fig. 3(a) shows the CH_4 and CO_2 conversions, the H_2/CO , and H_2 selectivity.

 CH_4 conversion and CO_2 conversion decreased after showing maximum values of 66.6% and 35.4%, respectively, at 10 mL/min as the steam supply increased. CH_4 conversion and CO_2 conversion increased as the steam supply increased because the methane steam reforming reaction (equation 9) increased along with the dry reforming reaction (equation 6). However, when the steam feed amount is 15 mL/min or more, the reformer temperature falls, the reactivity mentioned above decreases, and the water-gas shift reaction (equation 10) increases. [20] The H₂ selectivity and H₂/CO showed the maximum values of 88.2% and 1.53, respectively, when the steam feed rate was 10 mL/min, when CH₄ conversion and CO₂ conversion were at their maximum.

$$CH_4 + H_2O \rightarrow CO + 3H_2 \tag{9}$$

$$CO + H_2O \rightarrow CO_2 + H_2 \tag{10}$$

Fig. 3(b) shows the concentration and the lower heating value of the reformed gas. When the steam feed rate for maximum CH_4 conversion and CO_2 conversion was 10 mL/min, the maximum values were shown for H_2 , CO, and THCs, which were the combustible matters of the product gases, and CO_2 and CH_4 , the main components of the biogas, were converted to the maximum and showed the minimum concentration value. This is because the already mentioned steam reforming reaction (equation 10) was excellent. However, the heating value of the product gases was almost

similar in the range of 551-553 MJ/Nm³ except when the

steam supply was 15 mL/min or more.



Fig. 3. Effects of steam feed rate; (a) CH₄ conversion, H₂/CO and H₂ selectivity (b) gas yield.

4.3. The Effects of the Recirculation of the Reformed Gases

Fig. 4 shows test results when the reformed gas is recirculated under the condition that the total gas supply, which is the standard condition of biogas, is 30 L/min and the O_2/C ratio is 1.1.

As shown in Fig. 4(a), the CH₄ conversion and CO₂ conversion decreased in the case of as the recirculation rate increased. As shown in Fig. 4(b), it is not that the amount of biogas components converted to CH₄ and CO₂ production gas decreased, but the unconverted CH₄ and CO₂ contained in the recirculated gas were re-introduced into the reformer and thus their concentrations increased. The H₂ selectivity and the H₂/CO were 63.8% and 1.42 when the recirculation was not performed, and gradually increased as the recirculation proceeded, and the values increased to 87.4% and 1.46,

respectively, when the recirculation rate was 100%. As confirmed by the concentrations of H_2 and CO as product gases in Fig. 4(b), the concentrations were 14.9% and 10.5%, respectively, when not recirculated, the value increased as the recirculation rate increased, and when the recirculation rate was 100%, the values increased to 16.2% and 11.1%, respectively.

As can be seen from the above, the amount of H_2 and CO, both combustible gases, increased during reformed gas recirculation. In particular, it can be seen that the H_2 selectivity improved because of the relatively large increase in H_2 , and at this time, the lower heating value (552 MJ/Nm³ at 100% recirculation rate) was high enough to enable the production of the high-quality gases.



Fig. 4. Effects of reformed gas recirculation; (a) CH₄ conversion, H₂/CO and H₂ selectivity (b) gas yield.

4.4. The Effects of Water Supply to the Recirculated Gas

Fig. 5 shows the results when water is injected to the top of the recirculation pipe at 10, 20, or 30 mL/min with the reformed gas recirculation rate at 100%.

CH₄ conversion and CO₂ conversion increased as the water supply increased, and decreased after registering the maximum values of 68.6% and 37.9% at the water feed rate of 10 mL/min. This result is almost similar to the case where steam is supplied in the injection gas (see Fig. 3). The H₂ selectivity and H₂/CO showed the maximum values of 88.1%

and 1.54, respectively, at the same water feed rate.

As a result, the above variables increased as the water feed rate increased up to 10 mL/min because as water converted into steam in the recirculation pipe while water was supplied, its amount increased, so methane steam reforming reaction (Equation 10), etc. took place. However, when the water feed rate was 15 mL/min or more, the above-mentioned variable values decreased because the amount of water converted into steam was reduced due to a decrease in the temperature of the recirculated gas from an increase in the water feed rate.



Fig. 5. Effect of water feed in the recirculated gas; (a) CH₄ conversion, H₂/CO and H₂ selectivity (b) gas yield.

Experiments were conducted in four cases to understand the biogas reforming characteristics found with the new type of microwave-matrix reformer as proposed in this study. "Case 1" applies the standard conditions as in Table 1, and "Case 2" supplies steam to the gas inlet under the standard conditions of "Case 1". And "Case 3" gets part of the reformed gas recirculated under the standard conditions, and "Case 4" supplies water to the top of the recirculation pipe as in "Case 3".

In Table 2, the H_2 selectivity indicates the ratio of the amount converted to hydrogen in the gas produced by reforming biogas. Supplying steam (Case 2) or supplying water to the top of the recirculation pipe (Case 4) showed a large value of 88% or more. Energy conversion efficiency

represents the ratio of the energy supplied as feed fuel and electricity to the energy generated as H₂ and CO. "Case 1" was the smallest, whereas "Case 4" was the largest at 48.6%. The power required to generate $1m^3$ of product gases (H₂+CO) was greatest when steam was supplied (Case 2) and smallest when reformed gas was recirculated (Case 3). The lower heating value of the product gas was the smallest in "Case 1" and the largest in "Case 3" at 5.52 MJ/Nm³.

Judging from the above results, when some of the product gases is recirculated (Case 3) in a concept newly proposed in this study, or when water is supplied to the top of the recirculation pipe (Case 4), it performed excellently in the hydrogen production amount and energy conversion cost, and the heating value of product gases.

	Case Study		Case 1	Case 2	Case 3	Case 4
	Air	L/min	23.2	23.2	23.2	23.2
Flow rate input	CO_2	L/min	2.4	2.4	2.4	2.4
	CH_4	L/min	4.4	4.4	4.4	4.4
	H_2O	mL/min	-	10	-	10
O/C atomic ratio @ input		mol/mol	1.1	1.1	1.1	1.1
GlidArc power		KW	1	1	1	1
		CO_2	5.37	5.08	5.38	4.88
		CH_4	4.42	4.92	5.48	4.62
		H_2	12.10	17.31	16.16	17.81
		СО	8.95	11.30	11.09	11.60
Output gas conc	c. (vol.%)	C_2H_2	0.28	0.44	0.42	0.45
		C_2H_4	0.10	0.20	0.18	0.20
		C_2H_6	0.03	0.14	0.12	0.13
		N_2	68.35	60.64	60.71	59.44
		O ₂	0.62	0.82	0.76	0.72
H ₂ Selectivity(%)			51.40	88.22	87.35	88.08
Energy Conversion Efficiency(%)			25.2	44.2	40.8	48.6
Output gas flow rate in m ³ /h			1.62	1.62	1.62	1.62
kWh to produce $1 \text{ m}^3 \text{ of } H_2 + CO$			0.513	0.624	0.495	0.53
Output LHV from Syngas MJ/Nm ³		MJ/Nm ³	4.253	5.51	5.524	5.493

Table 2. Comparison of reforming characteristics in different case.

Case 1 – standard conditions in Table 1 without recirculation; Case 2 – When steam is supplied to the inlet under standard conditions; Case 3 – When reformed gas is recirculated under the standard conditions; and Case 4 – When water is supplied to the recirculation gas line

Table 3 below shows the optimal operating conditions and results for MMR reforming through partial oxidation of methane as revealed through the above-mentioned study with the different variables. When the O_2/C ratio was 1.1, the total

gas feed rate was 30 L/min, the recirculation rate was 100%, and water feed rate was 10 mL/min, the CH_4 conversion was 68.6%, the CO_2 conversion was 37.9%, the H_2 selectivity was 88.1%, and the H_2/CO was 1.54. The results of the study are thought to be applicable to a SOFC stack, which is a fuel cell for a residential power generator.

Standard	Standard Conditions O ₂ /C Total Gas Feed Rate		Water Feed Rate		Microwave Power			
Va	lues	1.1	30 L/	ímin	10 mL/min		0.8 kW	
Product Gas ¹⁾ (%)			CH ₄ conversion	CO ₂ conversion	H ₂ selectivity	H ₂ /CO		
H ₂	СО	CH ₄	CO_2	THCs ²⁾	68.6 %	37.9 %	88.1 %	1.54
44.87	29.23	11.64	12.30	1.96				

Table 3. Optimal operating conditions and their results (Case 4)

¹⁾ Product gas is reformed gas concentration excluding N₂ and O₂; ²⁾ THCs is total amount of hydrocarbons (C₂H₂, C₂H₄, C₂H₆).

5. Conclusion

This study has proposed a new type of microwave-matrix reformer, and identified the reforming characteristics of O_2/C , steam supply, reformed gas recirculation, and water supply in the recirculated gas involved in supplying methane. It has further presented the operating conditions of optimal conversion for each case based on the reforming conversion performance for different variables.

In the biogas reforming through partial oxidation combustion, the O2/C ratio has an optimal value while a reducing environment is maintained. However, the MMR applied in this study was 1.1, and the conversions, H_2 selectivity, the H_2/CO , and the heating value (H_2+CO) all had the maximum values. When steam was supplied to the inlet, H₂ production was not only increased due to steam reforming in addition to the existing partial oxidation and dry reforming until water feed rate was 10 mL/min, and the H₂ selectivity was increased as well and all the indicators show the maximum values. The amount of H₂ and CO, which were combustible gases, slightly increased during reformed gas recirculation. In particular, it was found that the H₂ selectivity improved because the H₂ increase was relatively large, and at this time, the heating value was high, so that high-quality gases could be produced. The water supply to the recirculated gas shows a pattern almost similar to that of steam, but it is considered to be a more useful method than steam injection because of the advantages of recirculation and less energy consumption required to make steam.

Through the above-mentioned study by variable, the water supply in the recirculation gas method has turned out advantageous for biogas reforming in MMR through partial oxidation. Here, as far as optimal operating conditions and main performance values are concerned, when the O_2/C ratio was 1.1, the total gas feed rate was 30 L/min, the recirculation rate was 100%, and the water feed rate was 10 mL/min, the CH₄ conversion was 68.6%, the CO₂ conversion was 37.9%, the H₂ selectivity was 88.1%, and the H₂/CO was 1.54. These results are deemed applicable to a SOFC stack for RPG.

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