



Hydrogen generation from biogas reforming using a gliding arc plasma-catalyst reformer

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ARTICLE INFO

Article history:

Available online 24 October 2009

Keywords:

Gliding arc plasma
Ni catalyst
Hydrogen generation
Biogas
Reforming

ABSTRACT

Biogas generated from landfills, wastewater disposal plants, wastes of livestock houses, etc., causing global warming when released into the air. This study developed a GAPCR (Gliding Arc Plasma-Catalyst Reformer) which convert the biogas into higher percentage of hydrogen as low pollution recycling energy, reduced the global warming and environmental problems. This study also conducted tests for the different variables that affect the biogas reforming efficiency of the GAPCR, and presented the optimum operating conditions for high percentage of hydrogen generation.

The parametric studies were carried out according to the change of steam to carbon ratio, catalyst bed temperature, total gas flow rate, input electric power, and biogas component ratio, i.e., CH₄:CO₂. The hydrogen concentration increased up to specific limit, and then maintained almost constant values for the same steam to carbon ratio and catalyst bed temperature. Hydrogen percentage decreased with the increase in total gas flow rate but little bit increases with the increase in electric power. In terms of biogas component ratio, hydrogen concentration decreased with the increase of CO₂ amount.

The optimum operating conditions showed the concentrations of 62% H₂, 8% CO, 27% CO₂, and 0.0% CH₄ on the basis of steam to carbon ratio of 3, catalyst bed temperature of 700 °C, total gas flow rate of 16 L/min, input electric power of 2.4 kW, and biogas component ratio of 6:4 (CH₄:CO₂). At this condition, H₂ yield and H₂ selectivity were same values of 59%. Energy efficiency and specific energy requirement were 53% and 289 kJ/mol, respectively.

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1. Introduction

The problem of energy insufficiency is being come due to the limitation of fossil fuel reserves from which energy utilization continuously increases. Global warming occurs due to increase of CO₂ and other gases in air that released from the burning of fossil fuels and from other working process, etc. This needs an urgent evolution in clean and environmental friendly alternative energies and their making technologies.

Now, more and more attention focused on the technology of using biogas, which released from landfills, wastewater treatment plants, wastes of livestock houses, etc., as alternative energy. Concentrations of such biogases are different from each other, depending on regional characteristics and material production methods. Generally, each biogas is composed of 55–70% methane, 27–44% carbon dioxide, 1% or less hydrogen, 3% or less H₂S, etc. It is possible to reprocess the released methane as energy for combustion, but it is difficult to provide continuous supply of

heat to boiler avoiding fluctuation when using as heat. The released CO₂ from biogas plays a vital role for global warming, having an adverse influence on environment. After converting biogas into high percentage of hydrogen, it is possible to use the biogas as a more stable, energy effectiveness, and environmental friendly fuel [1–3]. The reduction in greenhouse gas encourages us using biogas as an alternative energy against the imposition of carbon tax. Today, more attention diverted in building an environmental friendly modern system for producing hydrogen-using biogas (waste gas).

Now plasma technology applied for the production of hydrogen or synthesis gas as a new method. Generally, plasma classified in two kinds: thermal plasma called equilibrium plasma and non-thermal plasma called non-equilibrium plasma [4].

For the thermal plasma, the electrical power injected in the discharge is high (higher than 1 kW) and the neutral species and the electrons have then the same temperature (around 5000–10,000 K). The temperature in the reactor and the energy consumption are very high. The cooling of the electrodes is generally useful to reduce their thermal erosion [5,6]. The use of this technology is not relevant for an efficient production of hydrogen in terms of energy consumption.

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In non-thermal plasma, the electrical power is very low (few hundred watts) and temperature of neutral species does not change whereas the temperature of electrons is very high (up to 5000 K). In this case, plasma not only provides energy to the system but also generate radical and excited species, allowing, initiating and enhancing the chemical reactions. The advantages of non-thermal plasma relate to the low temperature that will result in less energy consumption and minimum electrode erosion therefore size and weight of the non-thermal plasma reactors are relatively less and attractive for mobile applications.

There is two types of general plasma discharges and it is impossible to keep a high level of non-equilibrium, high electron temperature and high electron density simultaneously, whereas most prospective plasma-chemical applications require high power for high reactor productivity and a high degree of non-equilibrium at once to support selective chemical processes. These parameters are somewhat practicable in gliding arc. The gliding arc occurs when the plasma is generated between two or more diverging electrodes placed in a gas flow. The gliding arc discharge has strong point of simple response control, high-energy efficiency, and environmental friendliness that developed in a new modern technology [7,8].

Three-phase gliding arc plasma-catalyst reformer (GAPCR) studied and designed. The new developed form combines a catalyst reactor with a gliding arc plasma reactor. The combined reformer has a quick starting characteristics and response time. It is open for testing of various kinds of fuel and biogas, including high molecular hydrocarbon, has a high conversion rate, and maintains optimum operating status for gas property. Therefore, GAPCR produces high percentage of hydrogen than others [9–11].

In addition, the study grasped characteristics of hydrogen generated from biogas by using the GAPCR. Lastly, tests were conducted on the changes of steam feed amount, i.e., steam to carbon ratio, catalyst bed temperature, total gas flow rate, input electric power, and biogas component ratio, i.e., $\text{CH}_4:\text{CO}_2$.

2. Experimental apparatus and methods

2.1. Experimental apparatus

Fig. 1 shows an experimental setup used for plasma reforming tests. The setup consists of a plasma-catalyst reformer, power supply, gas/steam supply line, measurement/analysis line, and control/monitoring system.

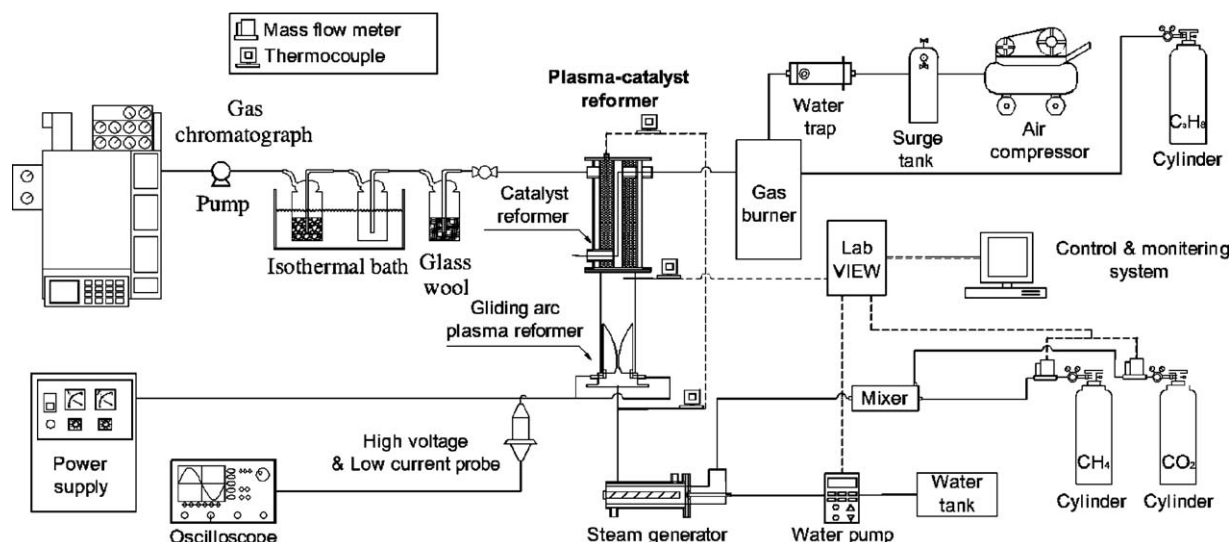


Fig. 1. Schematic of the GAPCR setup.

The plasma-catalyst reformer combined with a gliding arc plasma reformer and a catalyst reformer. The gliding arc plasma reformer had three fan-shape electrodes located at 120° in a quartz tube and fixed on the opposite side at a gap of 4 mm on a ceramic ring part. The quartz tube is used for the purpose of insulation and internal inspection of the plasma reactor. The jet nozzle for gas installed with a diameter of 3 mm at the upper part of electrodes. The catalyst reformer designed in a triple co-axial tube to heat catalysts evenly in the reactor.

The manufactured $\text{Ni}/\gamma\text{-Al}_2\text{O}_3$ catalyst (Süd-chemie, FCR-4, Japan) using the impregnation method. The catalyst consist of 12% Nickel oxide (NiO) with gray spherical $\gamma\text{-Al}_2\text{O}_3$ as catalyst supports. Bulk density and diameter are 1.22 kg/L and 2 mm, respectively.

The power supply (Unicorn Tech, UAP-15KIA, Korea) used to stabilize plasma within the plasma reactor, and has maximum capacity of 15 kW (voltage: 15 kV; AC: 1 A). The gas/steam supply line supplied gases into the plasma reactor precisely using the methane and CO_2 mass flow controllers (LINETECH, M3030V, Korea) and (BRONKHIST, F201AC-FAC-22-V, Netherlands), respectively. The quantitative pump (KNF, STEPPOS 03, Switzerland) supplied water into the steam generator.

The measurement/analysis line was operational for temperature and gas analysis. Temperature measured by K-type thermocouple while H_2 , CO and hydrocarbon (C_nH_m) gases were sampled and analyzed at the same time using two gas chromatographs (SHIMADZU, GC-14B, Japan; VARIAN, CP-4900, Netherlands). The LabVIEW instrument (National Instrument, LabVIEW 8.6, USA) control flow of gases, water pump and records the change of temperature and other conditions automatically.

2.2. Experimental methods

The plasma-catalyst reformer kept the temperatures of 580°C in the gliding arc reactor and 700°C in the catalyst reactor. Since steam-reforming reaction of Eq. (10) is highly endothermic, an external heat source (gas burner) is required. For the complete conversion of methane, high temperature in the catalyst reformer is critical. Therefore, the catalyst reformer provided gas burner to maintain 700°C for the best activation of Ni catalyst.

Tests at optimum conditions conducted after injecting steam of 12 L/min, methane of 2.4 L/min and CO_2 of 1.6 L/min in a stable state at which plasma discharge power was 2.4 kW. CH_4 and CO_2 injected with controlled amount through mass flow controller and the steam entered from the steam generator together with these

Table 1

The experimental range for parametric test studies.

Experimental variables	Steam/carbon ratio	Catalyst bed temperature (°C)	Total gas flow rate (L/min)	Input electric power (kW)	Biogas component ratio (CH ₄ :CO ₂)
Range	1–5.5	506–777	8–2.4	2.4–3.5	6:4–4:6
Residence time (s)	Gas space hourly velocity (h ^{−1})			Catalyst loading (g/cm ²)	
4.32	2529			8.5	

gases, and injected into the plasma reactor as a mixed gas. The micro-control pump controlled water amount and calculated the converted steam amount.

The reformed gas collected at the sampling port installed at the exit of the plasma-catalyst reactor and analyzed continuously by gas chromatographs in dry basis after passing through the glass wool and isothermal bath to remove moisture and soot in the gas. TCD used as a GC detector. The analysis columns used as Molecular Sieve 5A for H₂, CO, CH₄ and Porapak Q for CO₂.

The parametric studies carried out according to the change of steam feed amount i.e., steam to carbon ratio, catalyst bed temperature, total gas flow rate, input electric power, and biogas component ratio, i.e., CH₄:CO₂. Table 1 shows the experimental range for various parameters. The test conducted at optimum operating condition that is highest hydrogen generation for each variable.

2.3. Data analysis

(1) H₂ yield [12] and H₂ selectivity [13]

The amounts mentioned in different units: mol, mol percentage, yield or selectivity. The definition of the last two magnitude amounts is more clear and therefore taken in all calculations as followed:

$$\text{H}_2 \text{ yield (\%)} = \frac{[\text{H}_2]_{\text{syngas}}}{[\text{H}_2]_{\text{feed gas}}} \times 100 \quad (1)$$

where [H₂]_{syngas} is hydrogen amount (L/min) within reforming products, and [H₂]_{feed gas} is maximum hydrogen amount (L/min) from feed gases:

$$\begin{aligned} \text{H}_2 \text{ selectivity (\%)} \\ = \frac{[\text{H}_2]_{\text{syngas}}}{2([\text{CH}_4]_{\text{input}} - [\text{CH}_4]_{\text{output}}) + ([\text{H}_2\text{O}]_{\text{input}} - [\text{H}_2\text{O}]_{\text{output}})} \times 100 \end{aligned} \quad (2)$$

where [H₂]_{syngas} is a hydrogen amount (L/min) within reforming products and [CH₄]_{input} and [H₂O]_{input} are methane and water amounts (L/min), [CH₄]_{output} and [H₂O]_{output} are methane and water output amounts (L/min).

(2) CH₄ conversion rate [14]

In order to produce hydrogen, hydrocarbon molecule (CH₄) has to be “cracked” or to break into C–H links. The performance of this operation evaluated by using the CH₄ conversion rate that the ratio of CH₄ contained in reforming products to the CH₄ contained in the feed gases:

$$\text{CH}_4 \text{ conversion rate (\%)} = \frac{[\text{CH}_4]_{\text{input}} - [\text{CH}_4]_{\text{output}}}{[\text{CH}_4]_{\text{input}}} \times 100 \quad (3)$$

where [CH₄]_{input} is a methane input amount (L/min) and [CH₄]_{output} is a output amount (L/min).

(3) Energy efficiency [15]

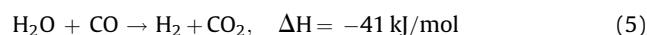
Hydrocarbon reacts with oxygen or water to produce hydrogen, whose heating value is higher than any other

hydrocarbon. Therefore, the efficiency of a reforming system is the lower heating value (LHV) of hydrogen produced divided by the input energy that is the summation of the electrical energy of the plasma and the LHV of the hydrocarbon injected:

$$\eta (\%) = \frac{[\text{H}_2 + \text{CO}]_{\text{produced}} \times \text{LHV}(\text{H}_2)}{\text{Input Plasma Energy} + \text{FUEL injected} \times \text{LHV}(\text{CH}_4)} \quad (4)$$

where [H₂ + CO]_{produced} is a generation amount of synthetic gas (L/min), LHV(H₂) is the lower heating value of hydrogen (kJ/Nm³), “Input Plasma Energy” is plasma input electric power (W), “FUEL injected” is methane amount in feed gas (L/min), and LHV(CH₄) is the lower heating value of methane (kJ/Nm³).

We consider that the whole CO produced then converted into H₂ by water gas shift (WGS) reaction Eq. (4). Therefore, we take into account the CO produced.



(4) Specific energy requirement [15]

This value is the input electric power used by the plasma that is required for producing 1 mol of H₂. Still considering the CO produced, the specific energy requirement is

$$\text{SER (kJ/mol)} = \frac{\text{Input plasma power}}{[\text{H}_2 + \text{CO}]_{\text{produced}}} \quad (6)$$

where input plasma power is input electric power (W) supplied to a gliding arc plasma reactor, and [H₂ + CO]_{produced} is a generation amount of synthetic gas (L/min).

Notice that this study focused on the “energy efficiency” and “specific energy requirement” of the plasma reactor and not for the overall system. Therefore, these calculations do not take into consideration the energy spent in the preheating of the reactant species.

(5) Residence time

$$\tau = \frac{V_R \rho_g}{F \rho_o} \quad (7)$$

τ is the residence time (s), V_R (m³) is the volume of the reactor without catalyst (the volume of the reactor minus the volume of catalyst in it). ρ_g is the density of reforming gas at reaction conditions (kg/m³), ρ_o is the density of reforming gas (kg/m³) and F is the flow rate of feedstock at temperature of 293 K and atmospheric pressure (m³/s).

(6) Gas space hourly velocity (GSHV) and catalyst loading (CL):

$$\text{GSHV} = \frac{F}{V} \quad (8)$$

where F is the ration between the mixture reaction volumes introduced per unit time (m³/h), and V is the catalyst volume (m³).

$$\text{CL} = \frac{m}{A} \quad (9)$$

where m the catalyst weight (g) and A is the catalyst bed cross section (cm²).

Table 2

Optimum operating conditions and their results.

Conditions				Steam/carbon ratio	Catalyst bed temperature (°C)	Total gas flow rate (L/min)	Input electric power (kW)	Biogas component ratio (CH ₄ :CO ₂)
Value				3	700	16	2.4	6:4
Syngas concentrations (dry vol.%)				CH ₄ conv. rate (%)	H ₂ selectivity (%)	H ₂ yield (%)	Energy efficiency (%)	Specific energy requirement (kJ/mol)
H ₂	CO	CO ₂	CH ₄					
62	8	27	0	100	59	59	53	289

3. Results and discussions

This most up-to-date study developed and set up a GAPCR (Gliding Arc Plasma-Catalyst Reformer) for the conversion of biogas into low-pollution recycling energy. It tries to solve the world energy crises and settle down the global warming environmental problem. This study conducted and test performs for the optimum operating conditions that generate high percentage of hydrogen by using the GAPCR. The respective conditions and test results of the optimum situation shown in Table 2.

The results indicates 100% CH₄ conversion rate, which proves that the GAPCR developed in this study was well designed as compared to previous study [16] having the 95% CH₄ conversion rate and specific energy requirement of 1270 kJ/mol.

The parametric studies carried out according to the changes of steam feed amount, i.e., steam to carbon ratio, catalyst bed temperature, total gas flow rate, input electric power, and biogas component ratio, i.e., CH₄:CO₂. Each test achieved by changing its value, while fixed other variables. The feed gas used by the simulated biogas that consists of CH₄ and CO₂.

3.1. Effect of steam feed amount

Fig. 2 shows experimental results for different steam to carbon ratio. The other variables like catalyst bed temperature, total gas flow rate, and input electric power are set at 700 °C, 16 L/min, and 2.4 kW respectively that are on optimum operating conditions in Table 2.

In the test rig, carbon black was formed in the plasma arc reactor at the steam to carbon ratio of 1 or less but not more than this limit and have maximum production capacity of steam to carbon ratio is 5.5. Therefore, tests conducted with the steam to carbon ratio of 1–5.5.

The plasma reformer produces the carbon black by means of plasma cracking reaction [14] Eqs. (10)–(12) at the situation of without steam or lower steam (like steam to carbon < 1). However the produced carbon black may be removed by steam to carbon gasification reaction [17] Eq. (13) like this test condition (steam to carbon > 1).



Carbon black is oxidized to CO, CO₂ and H₂ by hydroxyl radical (OH) formed through the radiolysis of water in plasma reformer ($\text{H}_2\text{O} \rightarrow \text{H}^+ + \text{OH}^* + \text{e}^-$) [18,19]. The reactions shown in Eqs. (14) and (15).



Fig. 2(a) shows the concentrations of hydrogen and carbon monoxide as a function of steam to carbon ratio. Tests conducted with or without catalysts in the catalyst reformer. With catalyst in the catalyst reactor, hydrogen concentration increases as steam to carbon ratio increase until reaching the optimum value of 62% at 3. The reason of increasing hydrogen concentration at the region of low steam to carbon ratio is that water gas shift (WGS) reaction of Eq. (5) prevailed over. Therefore, hydrogen concentration almost maintained because the excessive injection of steam little bit influence the hydrogen generation.

The catalyst in the catalyst reactor improves the efficiency of reforming at optimum conditions increasing hydrogen concentra-

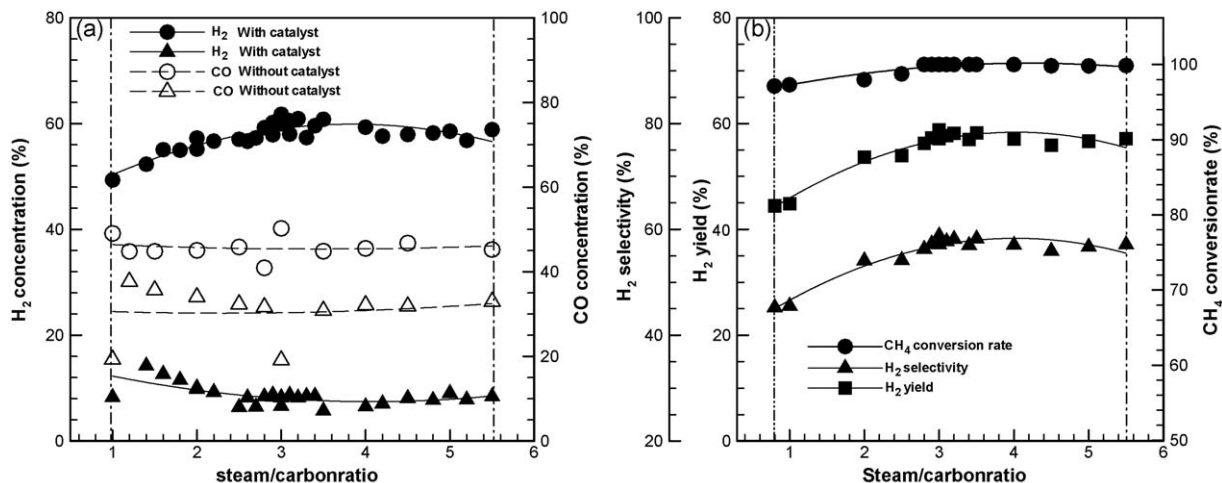


Fig. 2. The effect of various steam to carbon ratios.

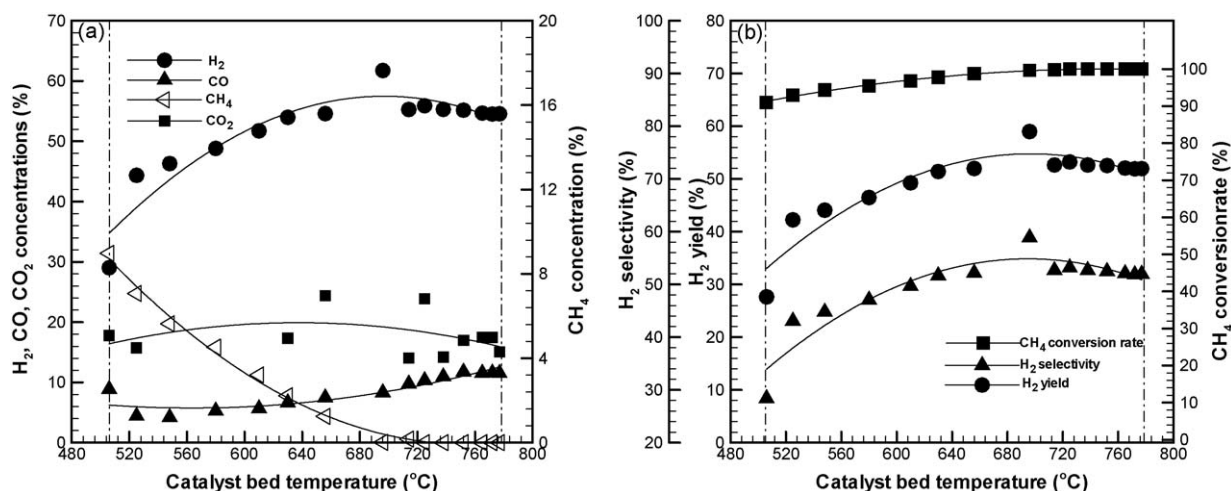


Fig. 3. The effect of the various catalyst bed temperatures.

tion from 40% to 62%, as compared to without catalyst. On the other hand, percentage of carbon monoxide decreases from 19% to 8%.

Fig. 2(b) shows the H₂ yield, H₂ selectivity and CH₄ by changing steam injection in case of with catalyst, calculated by Eqs. (1)–(3).

H₂ yield increases with the increase in steam to carbon ratio, amounting to maximum 59% at the steam to carbon ratio of 3 at this point H₂ selectivity and CH₄ conversion rate are 59% and 100%, respectively, showing that CH₄ gas was completely converted into syngas. The maximum conversion rate maintained at constant value while increasing steam to carbon ratio.

3.2. Effect of catalyst bed temperature

Fig. 3 presents the reforming characteristics of catalyst bed temperatures. Tests were conducted with changing catalyst bed temperatures from 506 to 777 °C, while fixing the steam to carbon ratio, total gas flow rate, and input electric power at 3, 16 L/min, and 2.4 kW, respectively.

Fig. 3(a) shows the concentrations of selected reforming products with variation of catalyst bed temperature. At the catalyst bed temperature of 700 °C, the hydrogen concentration is 62%, showing the maximum value, but above 700 °C, the concentration does not change nearly. The carbon monoxide concentration somewhat increases with the increase of catalyst bed temperature, but does not vary greatly. Methane concentration shows 9% at the initial catalyst temperature of 506 °C and

decreases gradually up to 0.0% at the temperature of 700 °C at the optimum condition. This means that even if methane mostly converted into hydrogen and carbon monoxide in the plasma reactor others also completely converted by catalyst surface reaction.

Fig. 3(b) shows H₂ yield, H₂ selectivity, and CH₄ conversion rate by the variation of catalyst bed temperatures. H₂ yield and H₂ selectivity increase on the same values of 59% each at 700 °C which are the optimum operating condition. CH₄ conversion rate is 100% at 700 °C or higher, showing that the injected methane is converted into syngas through the reforming reaction.

3.3. Effect of total feed gas amount

Fig. 4 presents the test results of total gas flow rate. Supply gas was injected with the total gas flow rates of 8–24 L/min after fixing steam to carbon ratio, catalyst bed temperature and input electric power at 3, 700 °C and 2.4 kW, respectively. Thus, tests conducted from 8 L/min to 24 L/min with the optimum conditions set at 16 L/min.

Fig. 4(a) shows that the increase of total gas flow rate causes decrease of hydrogen generation from 63% to 55%, increase of CO concentration from 2% to 17%. CH₄ and CO₂ increases with increasing the total gas flow rate.

By increasing total gas flow rate leads to decrease the residual time in the plasma-catalyst reactor. Therefore, supply of CH₄ and

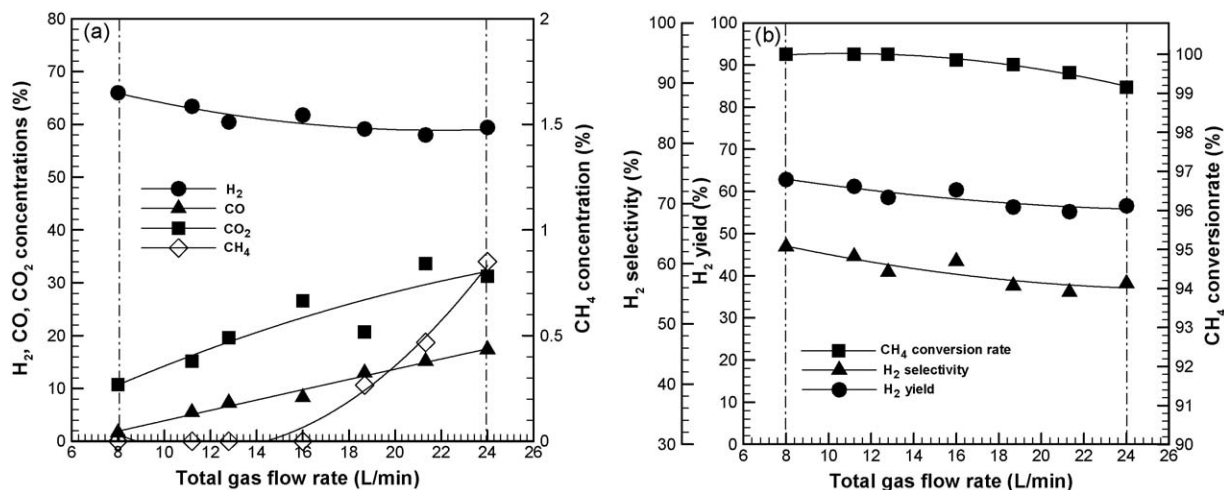


Fig. 4. The effect of total gas flow rate.

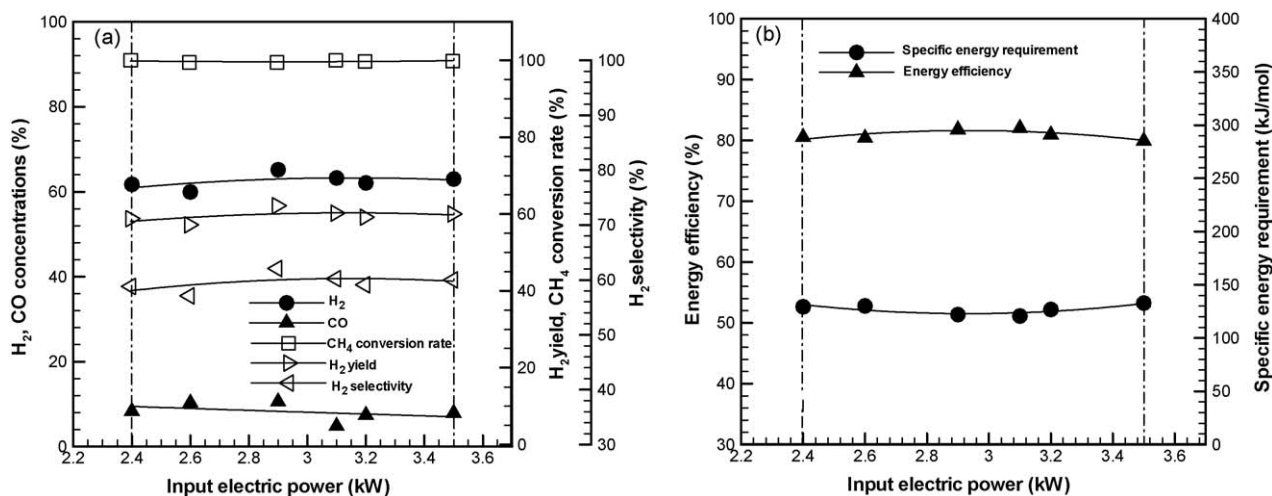


Fig. 5. The effect of the various input electric powers.

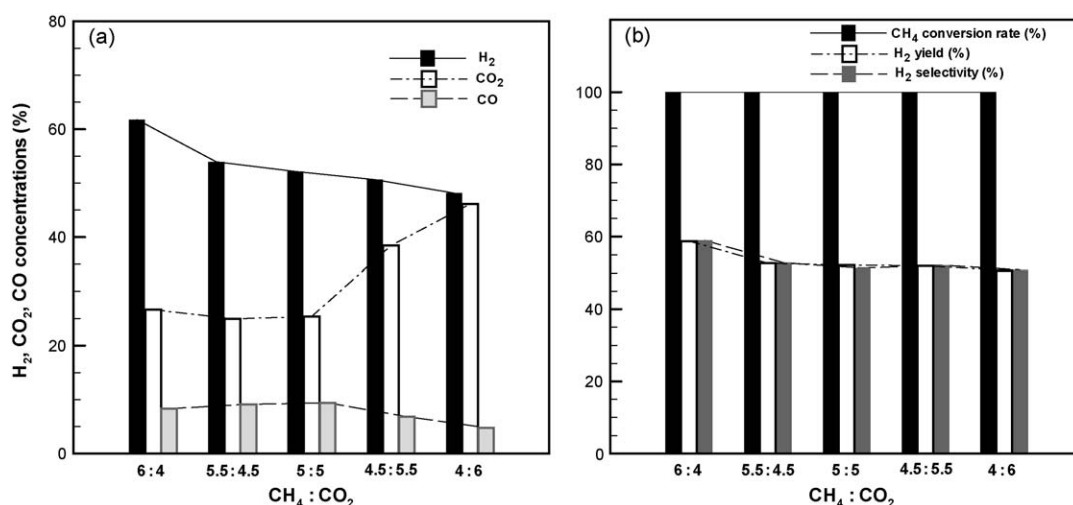


Fig. 6. The effect of the various biogas producing from CH₄:CO₂ ratios.

CO₂ increased with lower steam reforming and dry CO₂ reforming reactions in Eqs. (16) and (17).

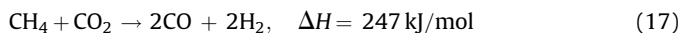
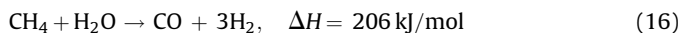


Fig. 4(b) shows H₂ yield, H₂ selectivity, and CH₄ conversion rate. The increase of total gas flow rate causes H₂ yield to decrease from 63% to 55%, and H₂ selectivity to decrease from 63% to 55%. The increase of total gas flow rate causes CH₄ conversion rate to decrease from 100% to 99%.

3.4. Effect of electric input energy

Fig. 5 shows the influence of input electric power. The changes in input electric power made after fixing steam to carbon ratio, catalyst bed temperature, and total gas flow rate at 3, 700 °C, and 16 L/min, respectively. Tests were conducted, with the input electric power set at 2.4–3.5 kW.

Fig. 5(a) presents influence of electric power on H₂ yield, H₂ selectivity, CO concentration and CH₄ conversion rate. The increase of input electric power keeps H₂ concentration and CO concentration almost constant at about 62% and 8%, respectively. H₂ yield

and H₂ selectivity are same values of 59% each, showing no great change and CH₄ conversion rate was 100%, showing that methane mostly converted into syngas.

Fig. 5(b) shows the energy efficiency and specific energy requirement caused by the variation of input electric power. The energy efficiency is almost same value about 52% with increasing the input electric power. The increase of input electric power causes increase of specific energy requirement from 289 to 297 kJ/mol.

As a result, when discussing CH₄ conversion rate and energy efficiency into consideration, it seems appropriate to apply the minimum input electric power of 2.4 kW as the optimum operation conditions in this study.

3.5. Characteristics of different biogas

Fig. 6 shows the influences caused by the changes of biogas component ratio. Steam to carbon ratio, catalyst bed temperature, and total gas flow rate were fix on 3, 700 °C, and 16 L/min, respectively. As the biogases generated from landfill varies from region to region, tests were conducted, with the gas component ratio set at 6:4–4:6 based on the ratios of CH₄ and CO₂.

Fig. 6(a) shows that the decrease of methane ratio in the simulated biogas causes the decrease of hydrogen concentration

from 62% to 48% and decreases CO concentration from 8% to 4.8%. On the other hand, the increase of methane ratios causes the increase of CO₂ from 25% to 46%.

Fig. 6(b) shows that the decrease of the methane ratio effect on H₂ yields from 58% to 51%, and H₂ selectivity from 58% to 51%. CH₄ conversion rate is approximately 100%, showing that conversion almost made uniformly.

As a result, CO₂ concentration, H₂ yield, and H₂ selectivity influenced by the change of biogas component ratio, but the conversion into syngas has no great change in case of CH₄ conversion rate.

4. Conclusions

The gliding arc plasma-catalyst reformer (GAPCR) designed and made to investigate about hydrogen generation from waste biogas CH₄ and CO₂. The parametric studies were carried out according to the change of steam feed ratio (steam to carbon), catalyst bed temperature, total gas flow rate, input electric power and biogas component ratio (CH₄:CO₂), which mainly affect hydrogen yield.

The percentage of hydrogen increases up to specific limit and then maintained almost constantly for the same steam to carbon ratio and catalyst bed temperature. The increase of gas flow rate caused the reduction of residual time in the plasma-catalyst reformer thus decreasing H₂ percentage, H₂ yield and CH₄ conversion rate. Hydrogen percentage somewhat increases with the increase in electric power. In terms of biogas component ratio, hydrogen concentration decreases with the increase of CO₂ amount in the biogas.

Through above parametric studies, optimum operating conditions obtained. The optimum conditions were taken as steam to

carbon ratio of 3, catalyst bed temperature of 700 °C, total gas flow rate of 16 L/min, input electric power of 2.4 kW, and biogas component ratio (6:4). The concentration in the syngas was 62% H₂, 8% CO, 27% CO₂, and 0.0% CH₄. H₂ yield and H₂ selectivity were 59% and 59% respectively and CH₄ conversion rate was 100% showing a high level. Energy efficiency was 53% and specific energy requirement was 289 kJ/mol.

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