CHAPTER VI

COMBINED PLASMA REFORMING OF CO_2-CONTAINING NATRUAL GAS WITH STEAM AND PARTIAL OXIDATION IN A MULTISTAGE GLIDING ARC SYSTEM

6.1 Abstract

For the production of synthesis gas from methane reforming, conventional catalytic processes require both high temperature and high pressure, resulting in high energy consumption and catalyst deactivation. Non-thermal plasma is considered a promising alternative technology for synthesis gas production because it can be operated in ambient conditions. In this research, the effect of stage number of multistage gliding arc discharge system on the process performance of combined steam reforming and partial oxidation of simulated natural gas was investigated. The simulated natural gas contained 70% methane, 5% ethane, 5% propane and 20% carbon dioxide. The experiments were carried out to investigate reactant conversions, product selectivities and yields, and power consumption by varying either residence time or stage number of plasma reactors, feed flow rate, hydrocarbons-to-O_2 feed molar ratio and input voltage. An increase in stage number from 1 to 3 stages at a constant feed flow rate enhanced the reactant conversions, and H_2 yield with a reduction of energy consumption. The lowest energy consumption of 1.99×10^{24} eV per mole of reactants converted or 1.24×10^{24} eV per mole of hydrogen produced was obtained from 3 stages of plasma reactors at a residence time and feed flow rate of 4.11 s, total feed flow rate of 100 cm^3/min, HCs-to-O2 feed molar ratio of 2/1, and input voltage of 15 kV, an input frequency of 300 Hz, and an electrode gap distance of 6 mm.

Keywords: Gliding arc discharge; Plasma; Steam reforming; Natural gas

6.2 Introduction

Synthesis gas (a mixture of hydrogen and carbon monoxide), is an important intermediate for synthesis of various chemicals, such as ammonia, methanol, acetic acid and dimethyl ether [1]. Regarding downstream synthesis, each desired product requires a different H_2/CO ratio of synthesis gas [2]. For instance, a H_2/CO ratio of 1/1 is needed to synthesize acetic acid or acetyloxide, while 2/1 is required to synthesize methanol or ethanol.

In general, coal, petroleum coke, natural gas, or biomass can be used to produce synthesis gas whereas, the major source for synthesis gas is produced from natural gas, since it provides the lowest cost worth, convenient in operation, and environmentally friendly to use. There are several commercial technologies for synthesis gas production: (1) steam reforming of CH_4, (2) partial oxidation of CH_4, and (3) CO_2 reforming of CH_4, which contribute to various H_2/CO ratios. The most popular technique to produce synthesis gas is steam CH_4 reforming (Equation 6.1) because it can provide a high H_2:CO raito. However, steam CH_4 reforming reaction requires a numerous amount of energy due to its highly endothermic reaction.

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \Delta H^0_{298} = +206 \text{ kJ/mol} \qquad (6.1)$$

The CH_4 reforming with CO_2 becomes increasing interest due to the fact that it does not only lessen methane consumption but also utilizes carbon dioxide. Nevertheless, the CH_4/CO_2 reforming reaction is also an endothermic reaction (Equation 6.2), and so, appropriate methods are required to achieve a considerable reaction rate in order to meet industrial requirements [3].

$$CH_4 + CO_2 \rightarrow 2CO + 2H_2 \qquad \Delta H^0_{298} = +247 \text{ kJ/mol} \qquad (6.2)$$

Non-thermal plasma has been proposed by several studies as an alternative technique to convert natural gas to more valuable products including synthesis gas [4-11], because it provides highly energetic electrons to initiate active species, leading to subsequential reactions. Corona discharge one of non-thermal plasma has was tested with combination of catalysts for CH_4/CO_2 reforming by Chang-jun Liu et al. [12-13]. It was pointed out that, corona discharge is characterized of localized breakdown to provide a small reaction volume and so, it is difficult to scale up for a large production capacity.

In our previous work [14-15], the challenging concept of the direct utilization of raw natural gas with a high CO_2 content was explored by using an AC low-temperature gilding arc discharge system, where the effects of each gas component in a simulated natural gas, operational parameters, and oxygen addition in feed were investigated. The results interestingly showed that the addition of a small amount of oxygen effectively minimized the carbon deposit on the electrode surface and inside the reactor wall, and also enhanced the performance of CO_2-containing natural gas reforming in terms of reactant conversion, desired product selectivity, desired product yield, and power consumption. However, to our knowledge, the combined plasma reforming of CO_2-containing natural gas with steam and partial oxidation has never been investigated. Therefore, this present work aimed, for the first time, to examine the effects of stage number of plasma reactors on both fixed residence time and total feed flow rate, hydrocarbons-to-oxygen (HCs-to-O_2) feed molar ratio, and input voltage on reactant conversion, product selectivity, product yield, and power consumption.

6.3 Experimental

6.3.1 Reactant Gases

The simulated natural gas used in this work consisted with a CH_4:C_2H_6:C_3H_8:CO_2 molar ratio of 70:5:5:20, and oxygen (O_2) were specially manufactured by Thai Industry Gas (Public) Co., Ltd.

6.3.2 AC Gliding Arc Discharge System

The schematic of a low-temperature multistage gliding arc system used in this work is shown in Figure 6.1. The 4 gliding arc reactors connected in series were made of a glass tube with an outer diameter of 9 cm and an inside diameter of 8.5 cm and had two diverging knife-shaped electrodes, which were fabricated from stainless steel sheets with a 1.2 cm width. The gap distance between the pair of electrodes was fixed at 6 mm. The steam fed into the system was achieved by vaporizing water in a mixing chamber at a controlled temperature of 120 °C by using a heating tape. The water flow rate to generate steam was controlled by a syringe pump. The flow rates of the simulated natural gas and O_2 were controlled by using mass flow controllers (98% accuracy) with transducers. A 7-μm stainless steel filter was placed upstream of each mass flow controller in order to trap any solid particles in the reactant gases. The check valve was also placed downstream of each mass flow controller to prevent any backflow. The simulated natural gas, oxygen, and steam were homogenously mixed in the mixing chamber before being fed upward into the first plasma reactor of the multistage gliding arc system at atmospheric pressure. To prevent the water condensation in the feed line and plasma reactors, the temperatures of the stainless tube from the syringe pump to the gliding arc reactor, as well as the plasma reactors, were maintained at 120 °C by using heating tapes. The compositions of the feed gas mixture and the outlet gas were quantitatively analyzed by an on-line gas chromatograph (HP, 5890) equipped with two separate columns, i.e. a Carboxen 1000 packed column and a PLOT Al_2O_3 “s” capillary column, which are capable to analyzing all hydrocarbons, CO, CO_2, and H_2.

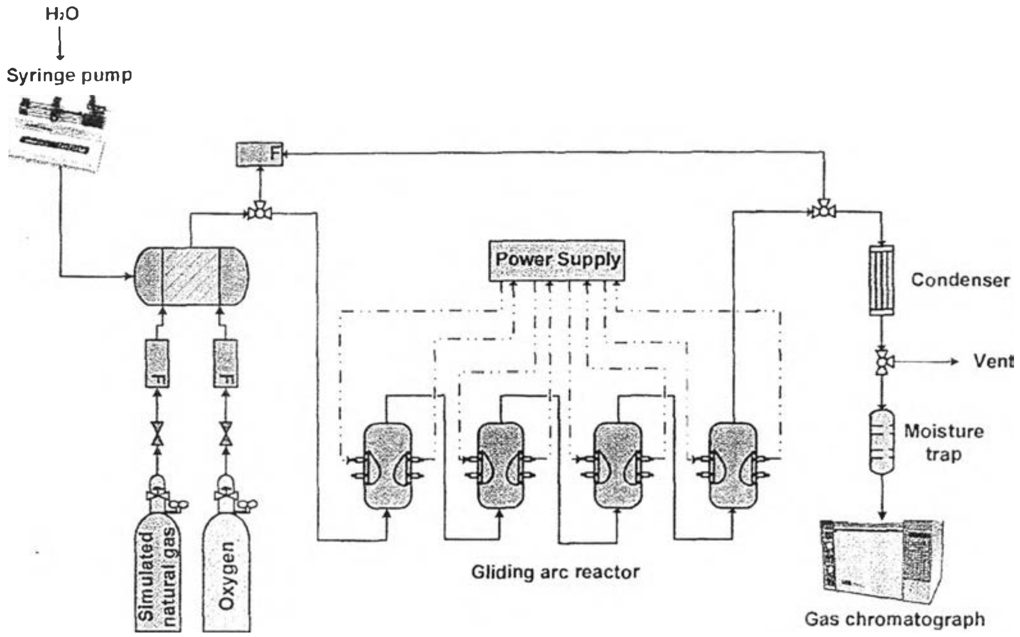

Figure 6.1 Schematic of gliding arc discharge system.

The power supply unit consisted of three steps. For the first step, a domestic AC input of 220 V and 50 Hz was converted to a DC output of 70 V by a DC power supply converter. For the second step, a 500 W power amplifier with a function generator was used to transform the DC into AC current with a sinusoidal waveform and different frequencies. The third step, was to step up voltage by using a high voltage transformer. Both output voltage and frequency were varied by the function generator. The voltage and current at the low voltage side were measured instead of those at the high voltage side by using a power analyzer since the plasma generated is non-equilibrium in nature. The high side voltage and current were, thereby, calculated by multiplying and dividing by a factor of 130, respectively [14-17].

The feed gas mixture was first introduced into the gliding arc reactors without turning on the power supply unit for any studied conditions. After the compositions of outlet gas became invariant, the power supply unit was turned on. The flow rate of the outlet gas was also measured by using a bubble flow meter. The outlet gas was then analyzed every 30 min by the on-line GC. After the plasma system reached a steady state with invariant outlet gas concentrations, the outlet gas was then taken for analysis at least a few times every hour. The average data were used to assess the process performance of the gliding arc discharge system.

6.3.3 Reaction Performance Calculation

The plasma system performance was evaluated from reactant conversions, product selectivities, H_2, CO, and C_2 yields, and power consumptions. The reactant conversion is defined as:

$$\%\ \text{Reactant conversion} = \frac{(\text{Moles of reactant in} - \text{Moles of reactant out})\times(100)}{\text{Moles of reactant in}} \quad (6.1)$$

The selectivity of any C-containing product is defined on the basis of the amount of C-containing reactants converted to any specified product, as stated in Equation 6.2. The percentage of coke formed can be calculated from the difference between the total amount of reactant conversions and total C-containing products, as given in Equation 6.3. In the case of the H_2 product, its selectivity is calculated based on the amount of H-containing reactants converted, as stated in Equation 6.4:

$$\%\ \text{Selectivity for any hydrocarbon product} = \frac{[P]\,C_p 100}{\Sigma[R_i](C_{Ri})} \quad (6.2)$$

$$\%\ \text{Coke} = \frac{\Sigma\,[R_i]\,C_{Ri} - \Sigma\,[P_i]\,C_{Pi}}{\Sigma[P_i]\,C_{pi}} \quad (6.3)$$

$$\%\ \text{Selectivity for hydrogen} = \frac{[H]\,H_P\,100}{\Sigma\,[R_i]\,H_{Ri}} \quad (6.4)$$

where $[P]$ = moles of product in the outlet gas stream

$[R_i]$ = mole of each reactant in the feed stream to be converted

$[H]$ = mole of hydrogen in the outlet gas stream

C_P = number of carbon atoms in a product molecule

C_R = number of carbon atom in each reactant molecule

H_P = number of H_2 atoms in a product molecule

H_R = number of H_2 atoms in each reactant molecule

The yields of various products are calculated using the following equations:

% C_2 hydrocarbon yield =

$$\frac{[\Sigma(\%\ CH_4,\ \%\ C_2H_6,\ \%\ C_3H_8\ ,\ \%\ CO_2\ \text{conversions})][\Sigma(\%\ C_2H_2,\ \%\ C_2H_4\ \text{selectivities})]}{(100)} \quad (6.5)$$

$$\%\ H_2\ \text{yield} = \frac{[\Sigma(\%\ CH_4,\ \%\ C_2H_6,\ \%\ C_3H_8\ \text{conversions})][\%\ H_2\ \text{selectivity}]}{(100)} \quad (6.6)$$

$$\%\ CO\ \text{yield} = \frac{[\Sigma(\%\ CH_4,\ \%\ C_2H_6,\ \%\ C_3H_8\ ,\ \%\ CO_2\ \text{conversions})][\%\ CO\ \text{selectivity}]}{(100)} \quad 6.7)$$

The power consumption is calculated in a unit of Ws per C-containing reactant molecule converted and Ws per H_2 molecule produced using the following equation:

$$\text{Power consumption} = \frac{P\times 60}{N\times M} \quad (6.8)$$

where P = power measured at the low voltage side of the power supply unit (W)

N = Avogadro's number (6.02×10^{23} molecule g mole^{-1})

M = rate of converted carbon in the rate of produced H_2 molecules (g mole min^{-1})

The residence time was calculated based on the reaction volume of gilding arc reactor divided by the feed flow rate. The reaction volume was estimated from the appearance of generated plasma.

6.4 Results and Discussion

It should be noted here that all possibilities of all plasma chemical reactions under the studied conditions were well described in our previous works [14-18].

Effects of Stage Number of Plasma Reactors

In general, for any given input voltage and frequency, the probability of collisions between reactant molecules and highly energetic electrons in plasma system is governed by both residence time and stage number of plasma reactors [16]. In order to identify the effect of stage number of plasma reactors, two sets of experiments were conducted. First, the effect of stage number of plasma reactors at a constant total feed flow rate was initially investigated to evaluate how it affected the process performance. The experiments were performed at a constant total feed flow rate of 100 cm^3/min, while the other operating parameters were controlled at the base conditions (a steam content of 10 mol%, an input voltage of 14.5 kV, an input frequency of 300 Hz, and an electrode gap distance of 6 mm) [17]. The residence times of the single stage, 2, 3, and 4 stages were 1.37, 2.71, 4.11, and 5.48 s, respectively. Secondly, to investigate the effect of stage number of plasma reactors, the experiments were carried out at constant residence time of 4.11 s, whereas the other operating parameters were controlled at the base conditions as mentioned above. Hence, the total feed flow rate was controlled at 33.3, 66.6, 100, and 133.3 cm^3/min for the single stage, 2, 3, and 4 stages, respectively.

6.4.1 Effect of Stage Number of Plasma Reactors on Reactant Conversion and Product Yield

Figure 6.2a illustrates the influence of the effects of stage number of plasma reactors on the reactant conversions and product yields at a constant total feed flow rate. The results show that all reactant conversions tended to remarkably increase with increasing stage number of plasma reactors, especially in C_3H_8 conversion. The results can be explained by the fact that, for a fixed total feed flow rate, the increase in stage number of plasma reactors directly increased the residence or reaction time in the system. The higher the residence time, the higher the opportunity for all reactants to be collide with highly energetic electrons in the plasma reaction zone, leading to increases in all reactant conversions. In addition, the highest C_3H_8 conversion as compared with the other reactants results from its lowest dissociation energy [19]. Surprisingly, the CO_2 conversion was found to reach to a positive value when the system was operated at 3 or 4 stages, which was different from previous work of the combined reforming and partial oxidation of CO_2-containing natural gas using an AC multistage gliding arc system [16]. This can be explained by the fact that the combined plasma reforming with steam and partial oxidation play an important role in enhancement of the CO_2 conversion.

The CO yield dramatically increased with increasing stage number of plasma reactors from 1 to 4 stage number of plasma reactors, whereas the C_2 yield remained almost unchanged (Figure 6.2a). In the meantime, the H_2 yield initially increased with increasing stage number of plasma reactors from 1 to 3, and then remained almost unchanged with further increasing stage number of plasma reactors from 3 to 4. The results suggest that under the studied conditions, an increase in stage number of plasma reactors at a fixed total feed flow rate or an increase in residence time by increasing stage number of plasma reactors affects predomently both of the dehydrogenation reactions to produce hydrogen and the partial oxidation to produce CO.

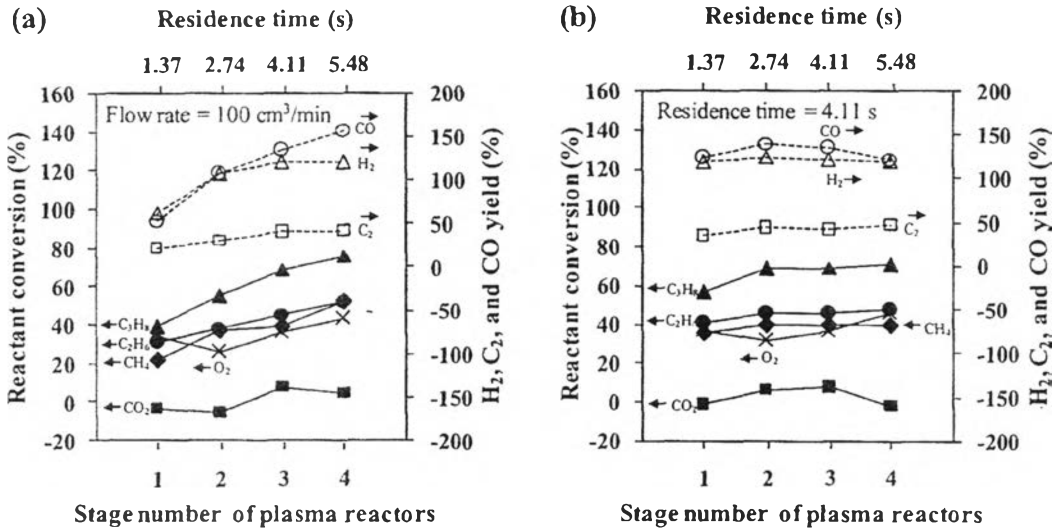

Fig. 6.2 Effect of stage number of plasma reactors on reactant conversions and product yields for the combined steam reforming and partial oxidation of natural gas (a) at a constant feed flow rate of 100 cm^3/min and (b) at constant residence time of 4.11 s (steam content, 10 mol%; HCs-to-O_2 feed molar ratio, 2/1 (Oxygen content of 33.33 mol%); input voltage, 14.5 kV; input frequency, 300 Hz; and electrode gap distance, 6 mm).

The reactant conversions and product yields as a function of stage number of plasma reactors at a constant residence time of 4.11 s are depicted in Figure 6.2b. The result reveals that CH_4 and C_2H_6 conversions and the C_2 yield remained almost unchanged with increasing stage number of plasma reactors while the C_3H_8 primarily increased and then remained almost unchanged. Besides, CO_2 conversion tended to increase with increasing stage number of plasma reactors from 1 to 3 stages, and then declined in negative value with further increasing stage number of plasma reactor from 3 to 4 stages. The CO yield increased slightly to reach a maximum at 2 stages after that it decreased with further increasing stage number of plasma reactors. The results suggest that, for a constant residence time, an increase in stage number of plasma reactors directly increases a high opportunity of the collision between all reactants and produced electrons, leading to increases in all reactants (C_3H_8, C_2H_6, CH_4, and CO_2) as supported by the increasing C_3H_8

conversion. The results of both C_2H_6 and CH_4 suggest that the rates of formation and conversion of both reactants were equal.

It is interesting to note that the O_2 conversion showed the opposite trend as compared to the CO_2 conversion, suggesting that an increase in stage number of plasma reactors from 1 to 3 stages provides positive effects to all reactant conversions but the negative effect especially on CO_2 conversion can occur if the stage number of plasma reactor is too high, leading tc increasing CO oxidation to CO_2. Besides, it was found that increasing stage number of plasma reactors from 1 to 4 stages had an insignificant impact on H_2 yield, indication that the increasing rates of both hydrogen formation and conversion were equal.

6.4.2 Effect of Stage Number of Plasma Reactors on Product Selectivity and Product Molar Ratio

The effect of stage number of plasma reactors on product selectivities at a constant feed flow rate of 100 cm^3/min is shown in Figure 6.3a. Interestingly, under the studied conditions, the main products were CO and H_2. With increasing stage number of plasma reactors from 1 to 2 stages, the CO selectivity increased remarkably, whereas it remain almost constant with further increasing stage number of plasma reactors from 2 to 4 stages. The H_2 selectivity also increased significantly with increasing stage number of plasma reactors from 1 to 2 stages and beyond 2 stages, it decreased sharply with further increasing stage number of plasma reactors. It was found that the selectivities for both CO and H_2 reached the maximum at 2 stages of plasma reactors. The selectivities for C_2H_4 and C_4H_{10} decreased with increasing stage number of plasma reactors from 1 to 4 stages, whereas the C_2H_2 selectivity increased significantly. The results are correlative with the decreases in C_2H_4/C_2H_2, H_2/C_2H_2, H_2/CO molar ratios (Figure 6.3a). The results suggest that an increase in stage number of plasma reactors with fixing a total feed flow rate basically increases the residence time, resulting in increasing several subsequential reactions including dehydrogenation, coupling, CO formation and cracking which will be discussed below.

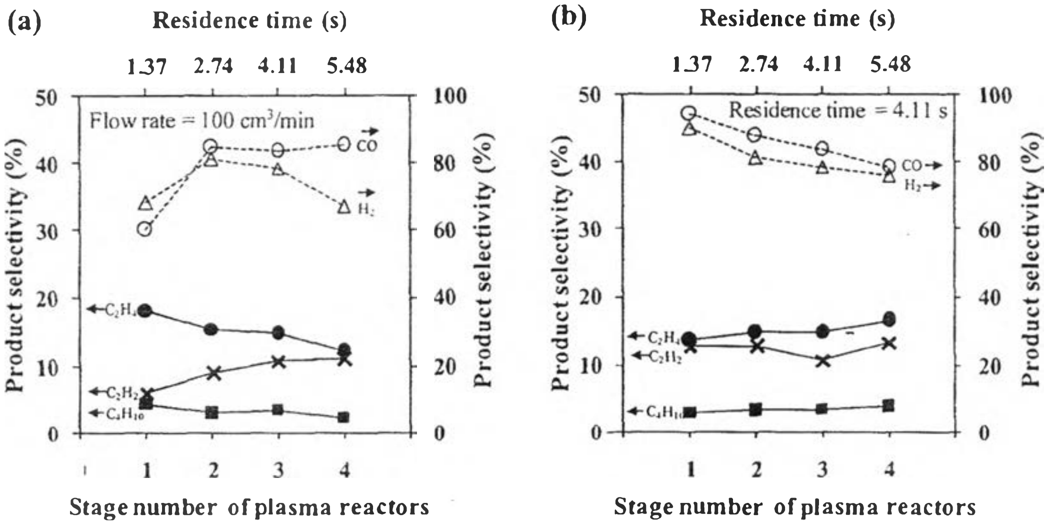

Fig. 6.3 Effect of stage number of plasma reactors on product selectivities for the combined steam reforming and partial oxidation of natural gas (a) at a constant feed flow rate of 100 cm^3/min and (b) at constant residence time of 4.11 s (steam content, 10 mol%; HCs-to-O_2 feed molar ratio, 2/1 (Oxygen content of 33.33 mol%); input voltage, 14.5 kV; input frequency, 300 Hz; and electrode gap distance, 6 mm).

Figure 6.3b shows the effect of stage number of plasma reactors on product selectivities at a constant residence time of 4.11 s. Interestingly, both CO and H_2 were also the main products of the plasma reforming of CO_2-containing natural gas with steam and partial oxidation under the studied conditions. The selectivities for both H_2 and CO gradually decrease with increasing stage number of plasma reactors from 1 to 4 stages. It is known that increasing in stage number of plasma reactors results in a higher probability of the collision of all reactants and generated electrons to produce active species, leading to increasing subsequential reactions. However, too high residence time and/or too many times passing through the plasma zone can increase undesirable reactions such as CO oxidation, hydrogenation, and H_2 oxidation, leading to the reduction of both selectivities of CO and H_2 in this study. On the other hand, selectivities for C_2H_4 and C_4H_{10} slightly increased with increasing stage number of plasma reactors, corresponding to the decreases in the ratios of H_2/C_2H_4 and H_2/C_2H_2, whereas C_2H_4/C_2H_2 slightly increased (Figure 6.4b).

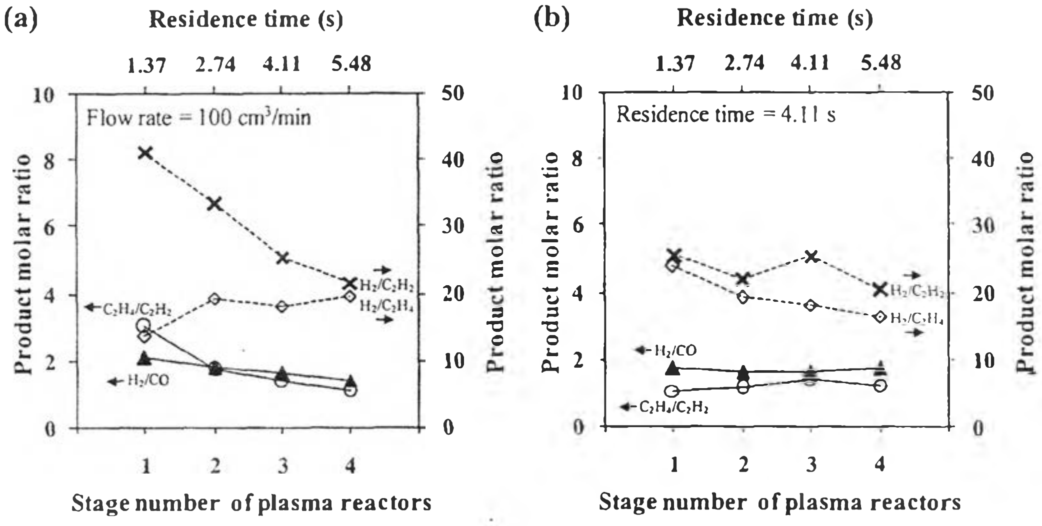

Fig. 6.4 Effect of stage number of plasma reactors on product molar ratio for the combined steam reforming and partial oxidation of natural gas (a) at a constant feed flow rate of 100 cm^3/min and (b) at constant residence time of 4.11 s (steam content, 10 mol%; HCs-to-O_2 feed molar ratio, 2/1 (Oxygen content of 33.33 mol%); input voltage, 14.5 kV; input frequency, 300 Hz; and electrode gap distance, 6 mm).

6.4.3 Effect of Stage Number of Plasma Reactors on Power Consumption and Coke Formation

As shown in Figure 6.5a, in the case of fixed feed flow rate, a sharp decrease in the power consumption per mole of all hydrocarbon reactants converted or hydrogen produced with increasing stage number of plasma reactors from 1 to 3 stages and beyond 3 stages, it adversely increased substantially with further increasing stage number of plasma reactors from 3 to 4 stages. In contrast, the coke formation increase markedly with increasing stage number of plasma reactors. In the case of fixed total flow rate, an increase in stage number of plasma reactors directly increases the residence time, leading to increases in all subsequential reaction including the cracking reactions to yield coke in the system. The minimum power consumption were 3.49×10^{-17} Ws per mole of hydrocarbon reactants converted and 2.04×10^{-17} Ws per mole of hydrogen produced at 3 stages of plasma reactors.

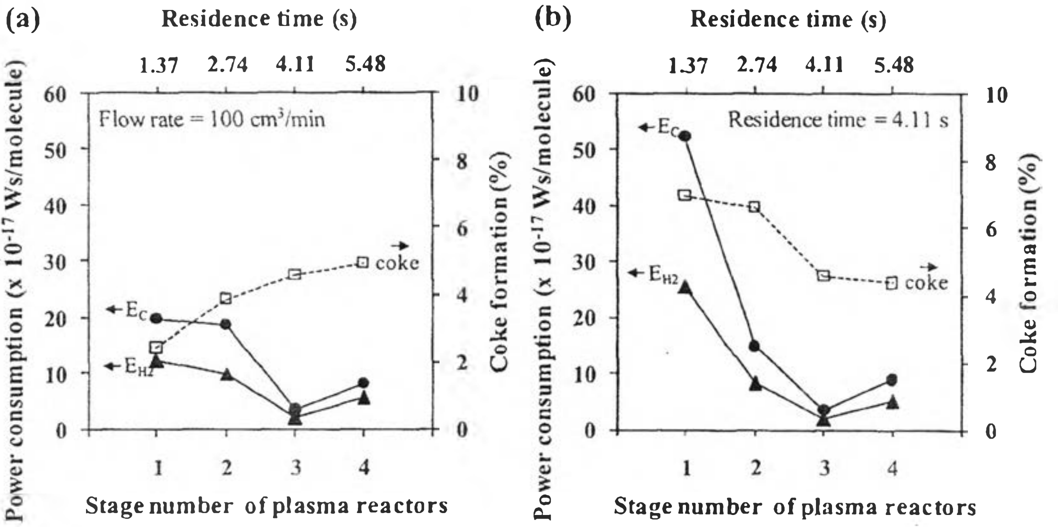

Fig. 6.5 Effect of stage number of plasma reactor on power consumptions and coke formation for the combined steam reforming and partial oxidation of natural gas (a) at a constant feed flow rate of 100 cm^3/min and (b) at constant residence time of 4.11 s (steam content, 10 mol%; HCs-to-O_2 feed molar ratio, 2/1 (Oxygen content of 33.33 mol%); input voltage, 14.5 kV; input frequency, 300 Hz; and electrode gap distance, 6 mm).

As shown in Figure 6.5b, the effect of stage number of plasma reactors on power consumptions and coke formation at a constant residence time of 4.11. It can be clearly seen that the power consumption per mole of all hydrocarbon reactants converted or hydrogen produced tends to dramatically decease with increasing stage number of plasma reactors from 1 to 3 stages, and beyond 3 stages, it slightly increases. In comparisons of both case of constant total feed flow rate and constant residence time, the effect of stage number of plasma reactors at constant residence time showed more prominently than that at a constant total feed flow rate especially on power consumptions. Interestingly, the minimum power consumption was found at the same condition stage number of 3 stages. From the results, the optimum conditions of a total feed flow rate of 100 cm^3/min, a steam content of 10 mol%, HCs-to-O_2 feed molar ratio of 2/1, 3 stages number of plasma reactors, electrode gap distance of 6 mm, input voltage of 14.5 kV, and input frequency of 300 Hz provided the lowest power consumption with reasonably high selectivities for both H_2 and CO.

6.5 Conclusion

The combined steam reforming and partial oxidation of CO_2-containing natural gas was investigated under two series of systems with a constant feed flow rate and a constant residence time by using a non-thermal multistage gliding arc discharge. The major products were mainly hydrogen and carbon monoxide. From the results, the stage number of plasma reactors under a constant residence time of 4.11 s showed the dominant effects on reactant conversions, product yields, product selectivities, and power consumption higher than that under a constant total feed flow rate of 100 cm^3/min. Under the operating conditions at a HCs-to-O_2 feed molar ratio of 2/1, an applied voltage of 14.5 kV, an input frequency of 300 Hz, and an electrode gap distance of 6 mm, the lowest power consumptions of 3.49×10^{-17} Ws per mole of hydrocarbon reactants converted or 2.04×10^{-17} Ws per mole of hydrogen produced was obtained from 3 stages of plasma reactors at a residence time and feed flow rate of 4.11 s and 100 cm^3/min, respectively.

6.6 Acknowledgements

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