CHAPTER III
EXPERIMENTAL

3.1 Materials

The simulated natural gas use in this study consists of methane, ethane, propane, and carbon dioxide, with a CH_4:C_2H_6:C_3H_8:CO_2 molar ratio of 70:5:5:20, is specially manufactured by Thai industry Gas (Public) Co., Ltd. Ultra-high purity oxygen used for performing the plasma reforming of the simulated natural gas with partial oxidation, is also obtained from Thai industry Gas (Public) Co., Ltd.

3.2 Experimental Setup

The schematic of the multistage gliding arc discharge system with 4 stages in series used in this research is shown in Figure 3.1. The system consists of 4 sections: feed gas mixing section, reactor section, power supply section, and analytical section.

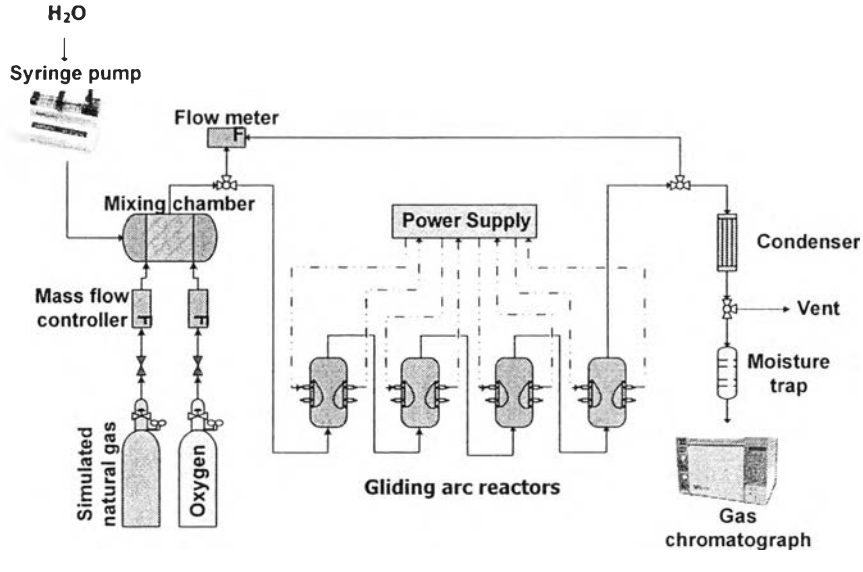

Figure 3.1 Schematic of gliding arc discharge system.

3.2.1 Feed Gas Mixing Section

3.2.1.1 Gas Mixing Section

The flow rate of the simulated natural gas, and the O_2, are controlled by a set of mass flow controllers and transducers supplied by AALBORG. A 7-μm stainless steel filter is placed upstream of each mass flow controllers in order to trap any solid particles in the reactant gas. The check valve is also placed downstream of each mass flow controller to prevent any backflow.

3.2.1.2 Steam Generation Section

The steam fed into the system was achieved by vaporizing water in a mixing chamber at a controlled temperature of 120 °C. The water flow rate to generate steam was controlled by a syringe pump supplied by Cole-Parmer. The simulated natural gas, oxygen, and steam were well mixed in the mixing chamber before being introduced upward into the plasma reactor at atmospheric pressure. To prevent the water condensation in the feed line and plasma reactor, the temperature of stainless tube from syringe pump to the gliding arc reactor, as well as the plasma reactor itself, was maintained at 120 °C by using a heating tape.

3.2.2 Reactor Section

The schematic of the gliding arc reactor is illustrated in Figure 3.2. The reactor is made of a glass tube with 9 cm OD and 8.5 cm ID. The reactor consisted of two diverging knife-shaped electrodes that were made from stainless steel sheets. The width of each electrode is 12 mm. The gap distance between the electrode pairs is fixed at 6 mm. Two teflon sheets are placed at top and bottom of the electrodes to direct the feed gas to pass through the reaction zone.

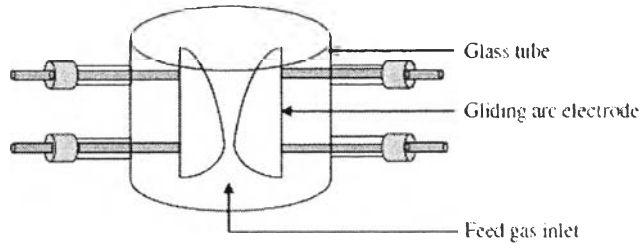

Figure 3.2 Schematic of the gliding arc reactor.

3.2.3 Power Supply Section

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. For the third step, the outlet voltage was stepped up by using a high voltage transformer. The output voltage and frequency were controlled 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.

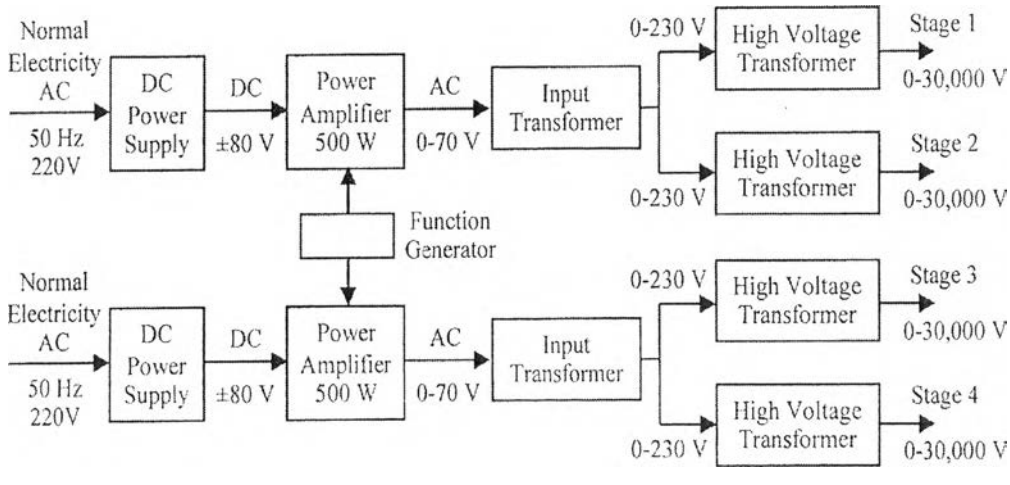

Figure 3.3 Schematic of the power supply unit.

3.2.4 Analytical Section

The feed mixture and the effluent gas are analyzed by an on-line gas chromatograph (HP, 5890) equipped with a thermal conductivity detector (TCD) and a flame ionization detector (FID). The quantitative analysis of the percent volumes of all gaseous components is carried out by correlating their peak area responses obtained from the GC chromatograms. The HP 5890 gas chromatograph is installed with a 10-port valve in order to separate the analyzed gas into two parts with independent sample loops. The first part is connected to a Carboxen 1000 column with the TCD. The second part is sent to a PLOT Al_2O_3"S" column connected with the FID. The GC conditions are summarized as follows:

Injector type:	Automatic sampling valve (programmable)
Injection temperature:	120°C
Oven temperature:	Initial temperature 40°C, ramp up at 70°C/min to 100°C followed by ramping up again at 30°C/min to 190°C and then hold for 17 min
Detectors:	Thermal conductivity detector (TCD) and flame ionization detector (FID)
Detector temperature:	190°C
GC columns:	Carboxen 1000 (15' x 1/8') and PLOT Al_2O_3 "S" (30 m x 0.53 mm)
Carrier gas:	High purity helium (99.995%)
Carrier gas flow rates:	35 cm^3/min for Carboxen 1000 column and 105 cm^3/min for PLOT Al_2O_3 "S" column

3.3 Reaction Performance Evaluation

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:

$$\% \text{ Reactant conversion} = \frac{(\text{Moles of reactant in} - \text{Moles of reactant out}) \times (100)}{\text{Moles of reactant in}} \quad (1)$$

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 2. The percentage of coke formed can be calculated from the difference between the total reactant conversions and total C-containing products, as given in Equation 3. In the instance of the H_2 product, its selectivity is calculated based on the amount of H-containing reactants converted, as stated in Equation 4:

$$\% \text{ Selectivity for any hydrocarbon product} = \frac{[P]\, C_p 100}{\sum [R_i](C_{Ri})} \quad (2)$$

$$\% \text{ Coke} = \frac{\sum [R_i]\, C_{Ri} - \sum [P_i]\, C_{Pi}}{\sum [P_i]\, C_{pi}} \quad (3)$$

$$\% \text{ Selectivity for hydrogen} = \frac{[H]\, H_P\, 100}{\sum [R_i]\, H_{Ri}} \quad (4)$$

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

[R] = moles 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:

$$\% \text{ } C_2 \text{ hydrocarbon yield} = \frac{[\sum(\% \text{ } CH_4, \% \text{ } C_2H_6, \% \text{ } C_3H_8, \% \text{ } CO_2 \text{ conversions})][\sum(\% \text{ } C_2H_2, \% \text{ } C_2H_4 \text{ selectivities})]}{(100)} \quad (5)$$

$$\%\ H_2\ \text{yield} = \frac{[\Sigma(\%\ CH_4, \%\ C_2H_6, \%\ C_3H_8\ \text{conversions})][\%\ H_2\ \text{selectivity}]}{(100)} \quad (6)$$

$$\%\ CO\ \text{yield} = \frac{[\Sigma(\%\ CH_4, \%\ C_2H_6, \%\ C_3H_8\ , \%\ CO_2\ \text{conversions})][\%\ CO\ \text{selectivity}]}{(100)} \quad (7)$$

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:

$$\text{Power consumption} = \frac{P \times 60}{N \times M} \quad (8)$$

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.

3.4 Studied Conditions

For any studied conditions, the system will be operated to reach steady state before taking samples for analysis. The steady state will be justified when the composition of effluent gas is invariant with time. After the composition of the reactant gases fed into the system is constant, the power supply unit will be turned on. The product gas will be analyzed to obtain the reactant conversions, product selectivities, and product yields.

The effect of the stage number of plasma reactors with fixed feed flow rate will be first investigated. The investigation of fixed residence time will then be

further examined on reactant conversions, product selectivities and yields, and power consumptions in the presence of both steam and oxygen addition in natural gas feed.

The experimental conditions will be as follows:

- HCs/O_2 feed molar ratio: 2/1
- Input voltage: 14.5 kV
- Input frequency: 300 Hz
- Electrode gap distance: 6 mm
- Steam content: 10 mol%