

CHAPTER III EXPERIMENTAL

3.1 Materials

The simulated natural gas used in this study consisted of methane, ethane, propane, and carbon dioxide, with a $\text{CH}_4:\text{C}_2\text{H}_6:\text{C}_3\text{H}_8:\text{CO}_2$ molar ratio of 70:5:5:20, and was 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 gliding arc system used in this study is shown in Figure 3.1. The system consisted of 4 sections: feed gas mixing section, reactor section, power supply section, and analytical section.

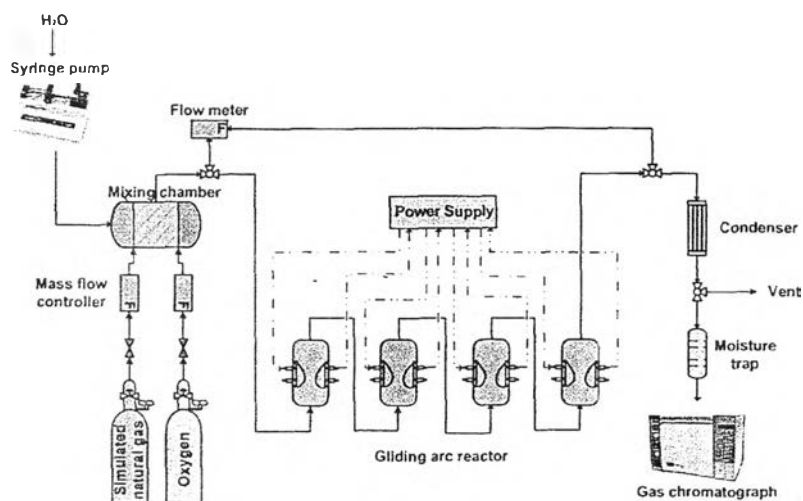


Figure 3.1 Schematic of gliding arc discharge system.

3.2.1 Feed Gas Mixing Section

3.2.1.1 *Gas Mixing Section*

The flow rates of the simulated natural gas, and the O₂, were controlled by mass flow controllers and transducers supplied by AALBORG. A 7- μ m stainless steel filter was placed upstream of each mass flow controller in order to trap any solid particles in the reactant gas. The check valve was 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 (xx). 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, 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 was 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 was fixed at 6 mm. Two teflon sheets were placed at top and bottom of the electrodes to direct the feed gas to pass through the plasma zone.

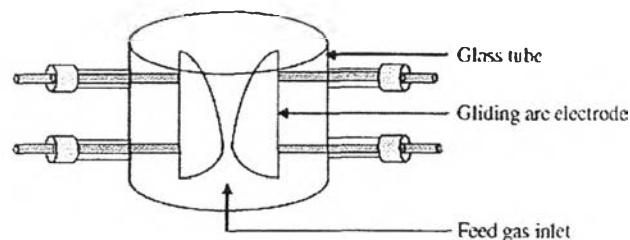


Figure 3.2 Schematic of the gliding arc reactor used in this study.

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 convert 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 show the electric diagram of the power supply unit used in this study.

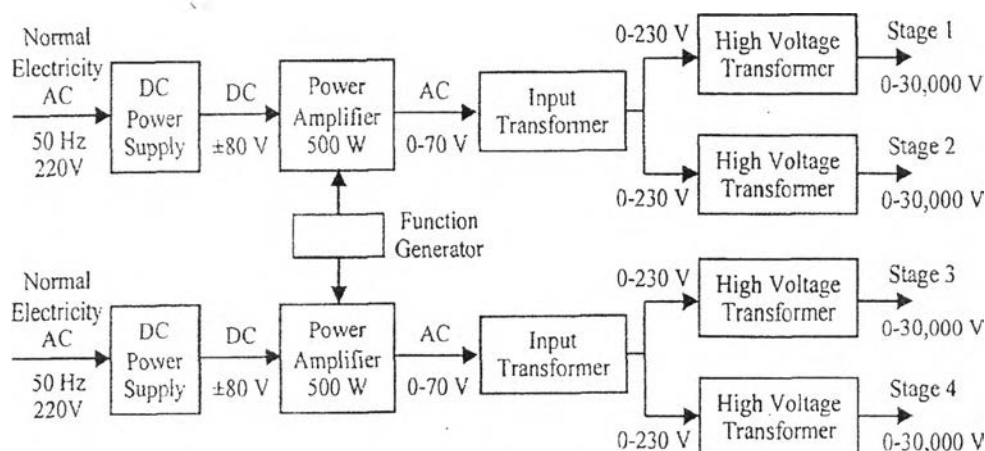


Figure 3.3 Schematic of the power supply unit.

3.2.4 Analytical Section

The feed mixture and the effluent gas samples were 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 was carried out by correlating their peak area responses obtained from the GC chromatograms. The gas chromatograph was equipped with a 10-port valve in order to separate the gas sample into two parts with independent sample loops. The first part was flow through a Carboxen 1000 column with the TCD. The second part was fed into a PLOT Al₂O₃ "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 ₂ O 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 ₂ O ₃ "S" column

3.3 Experimental Procedure

The main objective of this research was to investigate the reforming performance of natural gas to hydrogen and valuable higher hydrocarbon products via steam reforming and partial oxidation reactions under low-temperature gliding arc discharge. In this study, a simulated natural gas, containing of 70% CH₄, 5% C₂H₆, 5% C₃H₈, and 20% CO₂, and steam was used as a feed gas. The experiments

were divided into 3 main parts: (1) synthesis gas production from the reforming of CO₂-containing natural gas with steam using an AC gliding arc discharge system, (2) synthesis gas production from CO₂-containing natural gas by combined steam reforming and partial oxidation in an AC gliding arc discharge, (3) synthesis gas production from combined plasma reforming of CO₂-containing natural gas with steam and partial oxidation in a multistage gliding arc discharge system

3.4 Reaction Performance Evaluation

The plasma system performance was evaluated from reactant conversions, product selectivities, H₂, CO, and C₂ 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 case of the H₂ 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:

$$\% \text{ C}_2 \text{ hydrocarbon yield} = \frac{[\sum(\% \text{ CH}_4, \% \text{ C}_2\text{H}_6, \% \text{ C}_3\text{H}_8, \% \text{ CO}_2 \text{ conversions})][\sum(\% \text{ C}_2\text{H}_2, \% \text{ C}_2\text{H}_4 \text{ selectivities})]}{(100)} \quad (5)$$

$$\% \text{ H}_2 \text{ yield} = \frac{[\sum(\% \text{ CH}_4, \% \text{ C}_2\text{H}_6, \% \text{ C}_3\text{H}_8 \text{ conversions})][\% \text{ H}_2 \text{ selectivity}]}{(100)} \quad (6)$$

$$\% \text{ CO yield} = \frac{[\sum(\% \text{ CH}_4, \% \text{ C}_2\text{H}_6, \% \text{ C}_3\text{H}_8, \% \text{ CO}_2 \text{ conversions})][\% \text{ CO selectivity}]}{(100)} \quad (7)$$

The power consumption is calculated in a unit of Ws per C-containing reactant molecule converted and Ws per H₂ 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 H₂ molecules
(g mole min⁻¹)

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