CHAPTER III

EXPERIMENTAL

3.1 Materials

3.1.1 Reactant Gases for Reaction Experiments

- Simulated natural gas ($CH_4:C_2H_6:C_3H_8:CO_2 = 70:5:5:20$)

- Air zero grade

- Oxygen (O_2)

3.1.2 Gases for GC Analysis

- Air zero grade

- H_2 99.99% purity

- Helium (He)

3.2 Equipment Setup

The schematic diagram of the multistage gliding arc discharge system is shown in Figure 3.1. The system can be classified into 3 sections as follows:
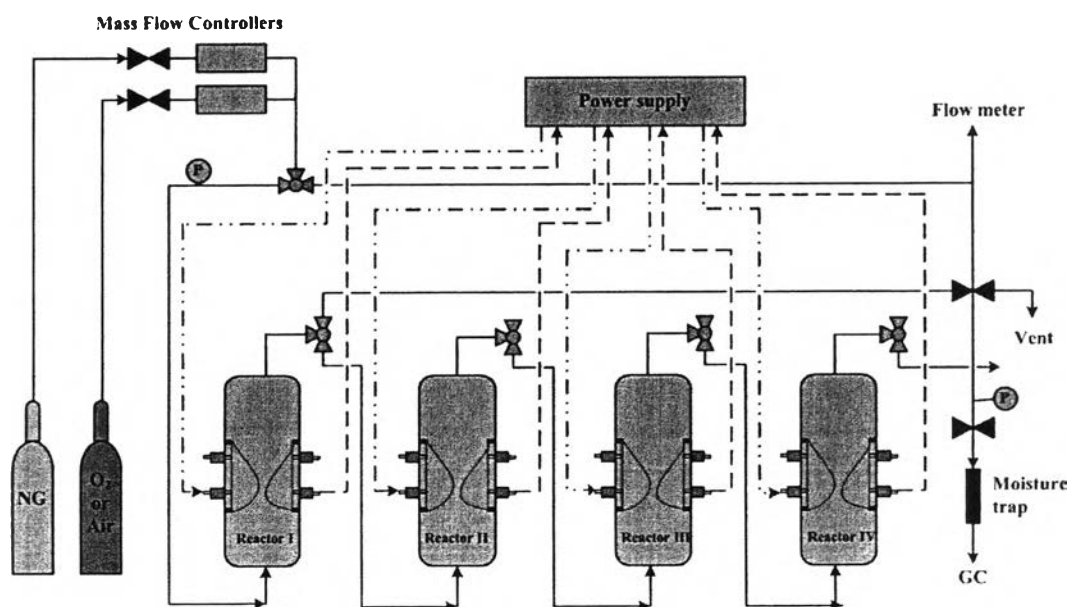

Figure 3.1 The schematic of the multistage gliding arc discharge system.

3.2.1 Feed Gas Mixing Section

Reactant gases, i.e. simulated natural gas, O_2/air, and helium balance, were controlled by a set of mass flow controllers and transducers supplied by SIERRA® Instrument, Inc. The 7-μm stainless steel filters were placed upstream of all mass flow controllers in order to trap any solid particles in the reactant gases. The check valves were also placed downstream of the mass flow controllers to prevent any back flow. All of the reactant gases and steam were well mixed and introduced upward into the first reactor at ambient temperature and atmospheric pressure.

3.2.2 Reaction Section

3.2.2.1 Reactor Units

The schematic diagram of the gliding arc reactor is illustrated in Figure 3.2. The reactor was made of 9 cm OD and 8.5 cm ID glass tubes. The reactor consisted of two diverging knife-shaped electrodes that were made from stainless steel sheets. The width of each electrode was 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 allow the feed gas for passing through the reaction zone.
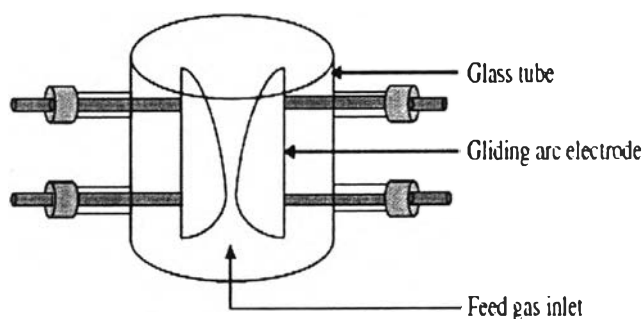

Figure 3.2 Schematic of the gliding arc reactor.

3.2.2.2 Power Supply Unit

The schematic diagram of the power supply unit is depicted in Figure 3.3. The domestic AC input of 220 V and 50 Hz was connected to the DC power supply converter to convert to DC of about 80 V. The DC was supplied through a 500 W power amplifier, which was connected to the Instek function

generator to generate waveform and to amplify voltage and frequency. The signal of alternative current is a sinusoidal waveform. The amplified current passed through the transformer to convert to 230 V AC. Thereafter, the variable output was transmitted to a high voltage current by nominal factor 130 times of low side voltage (input). A Lutron power analyzer was used to measure power, power factor, current, frequency, and voltage at the low side voltage of the power supply unit.
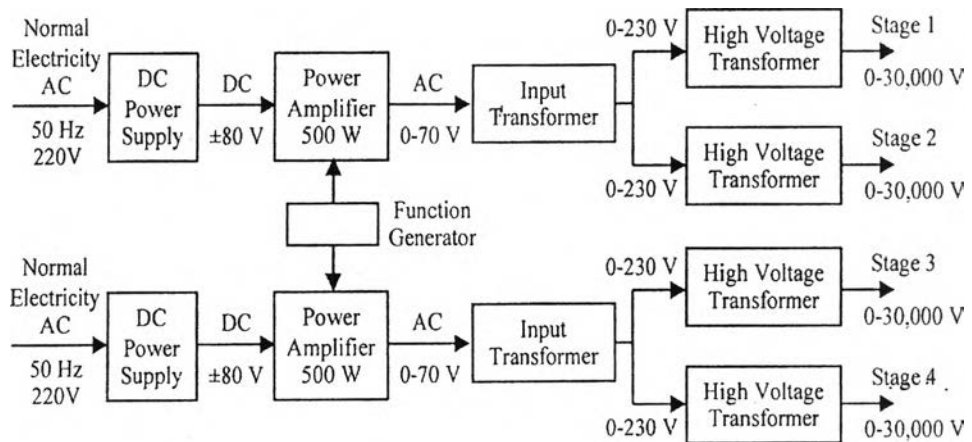

Figure 3.3 Schematic diagram of the power supply unit.

3.2.3 Analytical Section

The feed mixture and the effluent gas 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 HP, 5890 gas chromatography was installed with a 10-port valve in order to separate the analyzed gas into two parts with independent sample loops. The first part was introduced to a Carboxen 1000 column connected with the TCD. The second part was 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 Experimental Procedure

The main objective of this research was to investigate the conversion of natural gas to hydrogen and higher valuable products via plasma reforming and partial oxidation reactions. In this study, a simulated natural gas consisting of 70% CH_4, 5% C_2H_6, 5% C_3H_8, and 20% CO_2 was used as a feed gas.

The experiments were divided into 2 parts: reforming of natural gas in an AC multistage gliding arc system without partial oxidation and reforming of natural gas in an AC multistage gliding arc system with partial oxidation. The details of the experiments are described below.

3.3.1 Reforming of Natural Gas in an AC Multistage Gliding Arc System without Partial Oxidation

The first part of this work was to investigate the reforming of simulated natural gas without O_2 addition for partial oxidation in low temperature plasma using the AC multistage gliding arc system. The experiment was conducted by varying feed flow rate, residence time, and stage number of plasma reactors while the other operating parameters (applied voltage; 17.5 kV, input frequency; 300 Hz, and electrode gap distance; 6 mm) were kept at optimum values obtained from previous work (Rueangjitt et al, 2007).

3.3.2 Reforming of Natural Gas in an AC Multistage Gliding Arc System with Partial Oxidation

The objective of the second part was to study the effect of added air and O_2 on the reforming of the simulated natural gas. The optimum operating conditions for the plasma reforming of the simulated natural gas used in the first part were also used. The hydrocarbons-to-oxygen molar ratio was adjusted to 2/1.

3.4 Studied Conditions

After the composition of the reactant gases fed into the system was constant, the power supply unit was turned on. The exhaust gas was analyzed to obtain the reactant conversions, product selectivities, and product yields.

All operating parameters used in this study are summarized in Table 3.1.

Table 3.1 Experimental conditions used in this study

Varied parameter	Feed flow rate (cm^3/min)	Residence time (s)	Stage number of plasma reactor	Frequency (Hz)	Voltage (kV)	Electrode gap distance (mm)	HCs/O_2 molar ratio
Feed flow rate	31.25	4.38	1	300	17.5	6	2/1
	62.50	4.38	2				
	93.75	4.38	3				
	125.00	4.38	4				
Residence time	125.00	1.10	1	300	17.5	6	2/1
	125.00	2.19	2				
	125.00	3.29	3				
	125.00	4.38	4				

3.5 Reaction Performance Evaluation

In general, the conversion is defined as:

$$\% \text{ conversion} = \frac{(\text{ moles of reactant in} - \text{moles of reactant out})(100)}{\text{moles of reactant in}}$$

The percent selectivity of products containing carbon atoms is defined on the basis of the amount of carbon converted from the reactants into any specified products. In case of hydrogen product, the hydrogen selectivity is calculated based on hydrogen converted from the reactants.

$$\% \text{ selectivity of any hydrocarbon product} = [P](C_P)(100) / \sum[R](C_R)$$

where

[P]	=	moles of product in effluent
[R]	=	moles of reactant in feed to be converted
C_P	=	numbers of carbon atom in a product molecule
C_R	=	numbers of carbon atom in a reactant molecule

$$\% \text{ selectivity of hydrogen} = [P](H_P)(100) / \sum[R](H_R)$$

where

H_P	=	numbers of hydrogen atom in a product molecule
H_R	=	numbers of hydrogen atom in a reactant molecule

The product yield is formulated as follows:

$$\% \text{ yield of } C_2 \text{ hydrocarbons} = \sum(\% \text{ conversion of } CH_4, C_2H_6, C_3H_8, CO_2)$$
$$\times \sum(\% \text{ selectivity of } C_2H_2, C_2H_4, C_2H_6) / (100)$$

$$\% \text{ yield of } H_2 = \frac{\sum(\% \text{ conversion of } CH_4, C_2H_6, C_3H_8) \times (\% \text{ selectivity of } H_2)}{100}$$

The specific energy consumption is calculated in a unit of Ws per a C-containing reactant molecule converted or per a hydrogen molecule produced (Ws/M) using the following equation:

$$\text{Specific energy consumption} = (P)\,(60)\,/\,(\tilde{N})\,(M)$$

where

P = Power (W)

$\tilde{N}$ = Avogadro's number = 6.02×10^{23} molecules·g-mole^{-1}

M = Rate of converted carbon in feed or rate of produced hydrogen molecule (g-mole·min^{-1})