

## CHAPTER III

### EXPERIMENTAL SECTION

In this research work, the plasma promoting partial oxidation of methane was carried out at atmospheric condition. The wire and plate electrodes made of stainless steel were used to generate an AC corona discharge in the quartz tube reactor where the reaction took place. Both the feed and product streams were analyzed by two gas chromatographs comprising of thermal conductivity and flame ionization detector. The purpose of this work was to determine the effects of applied voltage, frequency, ratio of component, flow rate and gap width on conversions and product selectivities.

#### 3.1 Materials

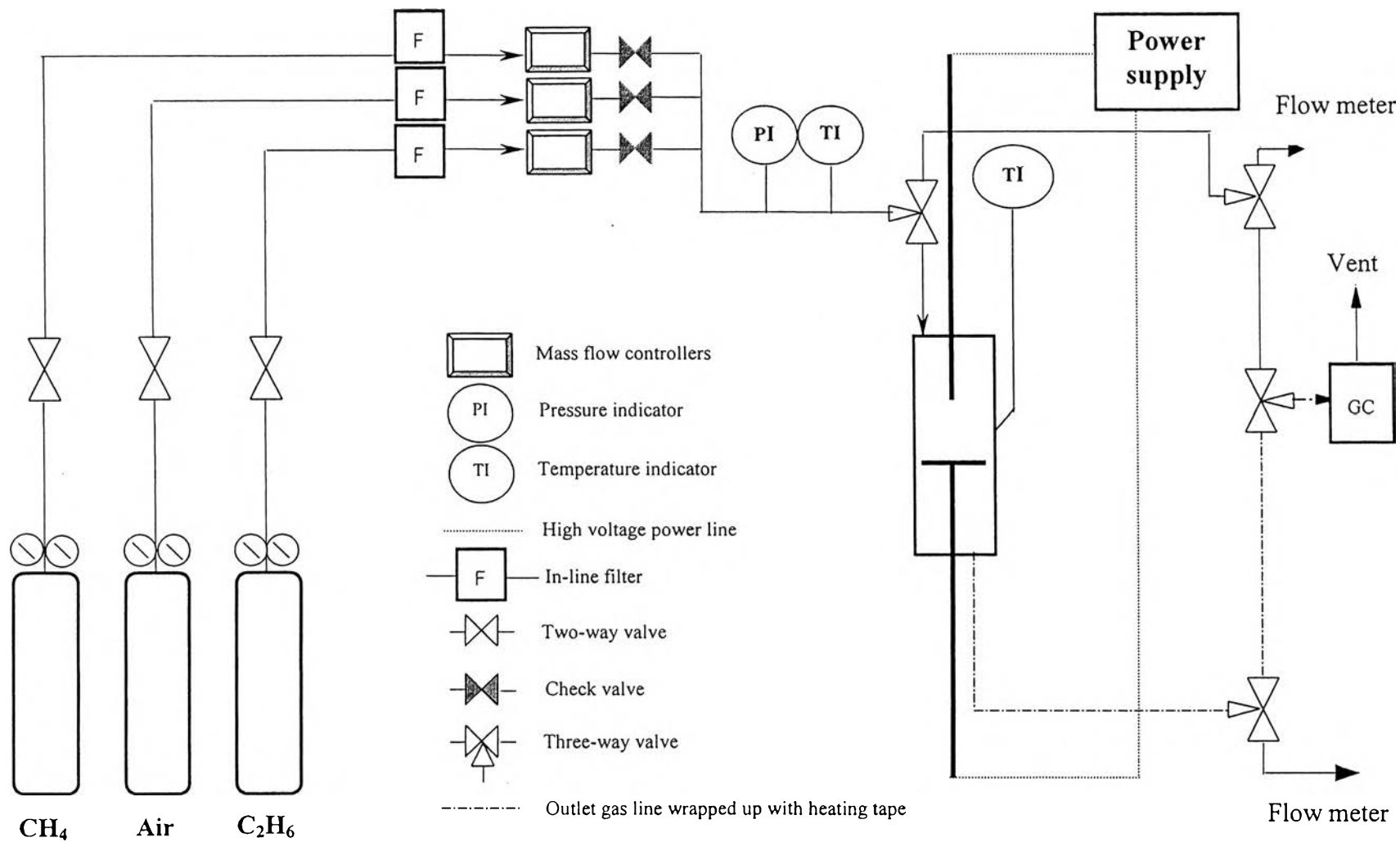
Methane ( $\text{CH}_4$ ) UHP grade of 99.97% purity, air zero grade moisture less than 25 ppm, oxygen range  $21 \pm 1\%$  and ethane ( $\text{C}_2\text{H}_6$ ) research grade, 99.99% were used in this experimental study.

#### 3.2 Experimental Setup

The schematic diagram of the entire reaction system used throughout this study was illustrated in Figure 3.1. The system could be categorized into three parts as reactant make-up section, reaction section and analysis section.

##### 3.2.1 Reaction Make-Up Section

All reactant gases used in this work were used without any purification. The flow rates of all gases fed into the reactor were controlled by



**Figure 3.1** Flow diagram of plasma reactor system

a set of mass flow control box and transducers supplied by SIERRA<sup>®</sup> Instrument, Inc. The error of the apparatus was reported to be less than 1%. During conducting the experiment, all mass flow controller were regularly calibrated with the digital bubble flow meter.

The pressure read out at the discharge end of all pressure regulators connected to the gas cylinders had to be maintained in range of 20-30 psia to guarantee the reliable function of the transducers. The 0.7 $\mu$ m in-line filters were placed upstream of all mass flow transducers in order to trap any foreign particles. The check valves were placed downstream of all mass flow controllers to protect from any back pressure effects.

Pressure and temperature of the feed gas were read out directly from a pressure gauge and thermocouple type- K installed downstream of make-up section.

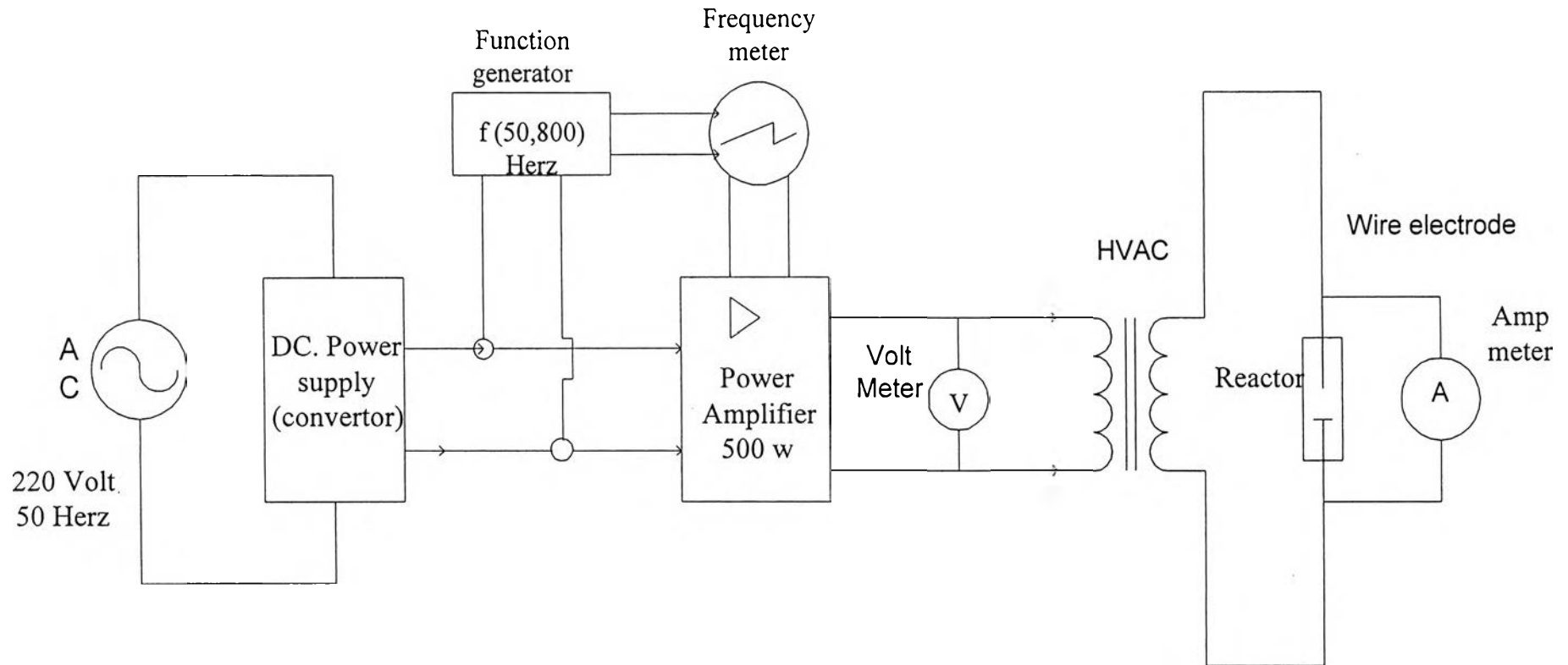
### 3.2.2 Reaction Section

#### 3.2.2.1 *Power Supply Unit*

Figure 3.2 illustrates the schematic diagram of the power supply unit. First, domestic AC power 220 V, 50 Hz was connected to the DC power supply converter to convert to relatively low DC current, then passed through a function generator for varying frequency in range 0- 800 Hz. The output passed through amplifier, 500 watt in order to amplify voltage in range 0 to 80 V. Eventually, the output was then transmitted to high voltage alternating current (HVAC) transformer. The HVAC could step up the voltage to a maximum value of 10,000 V.

#### 3.2.2.2 *Reactor Unit*

The quartz tube reactor used in this study was shown in Figure 3.3 and the reactor tube had an inside diameter of 8 mm. The wire electrode was made of 1/16 inch diameter stainless steel rod. The stainless steel plate electrode had the shape of a circular stainless steel with a thickness



**Figure 3.2** Schematic diagram of power supply unit

of 1 mm. The disk was drilled with 8 holes and each hole had a diameter of 1 mm. A type- K thermocouple was connected at the middle of the two electrodes and at the outside tube wall for monitoring the wall temperature.

### 3.2.3 Analysis Section

The product gas stream was heated along the line to an on-line GC unit in order to prevent any liquid condensation in the line before reaching the GC. The volumetric flow rates of product stream and feed stream were measured by using a digital flow meter.

A Perkin-Elmer GC with a thermal conductivity detector (TCD) and a flame ionization detector (FID) was used as an on-line GC. The quantitative analysis of the percent volume of all gaseous components was carried out by correlating their peak-area response derived from the GC chromatograms. The Perkin-Elmer GC 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 HayeSep-T column connected with FID allowing the separation of methanol. The second part was sent to a Carbonxen-1000 column connected with the TCD allowing the separation of H<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub>, CH<sub>4</sub>, CO<sub>2</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub>. The GC conditions are summarized as follows:

Injector type:	Automatic sampling 10-port valve
Injector temperature:	110 °C
Oven temperature:	35 °C hold for 5 minutes, then rise to 170 °C at rate 15 °C/min and eventually hold for 30 minutes.
Detectors:	Thermal conductivity detector (TCD) and flame ionization detector (FID)
Detector temperature:	180 °C

Columns: HayeSep-T (5' x 1/8'') and  
Columns: Carbonxen-1000 (15' x 1/8'')  
Carrier gas: High purity helium  
Carrier gas flow rates: 30 ml/min for HayeSep-T  
30 ml/min for Carbonxen-1000

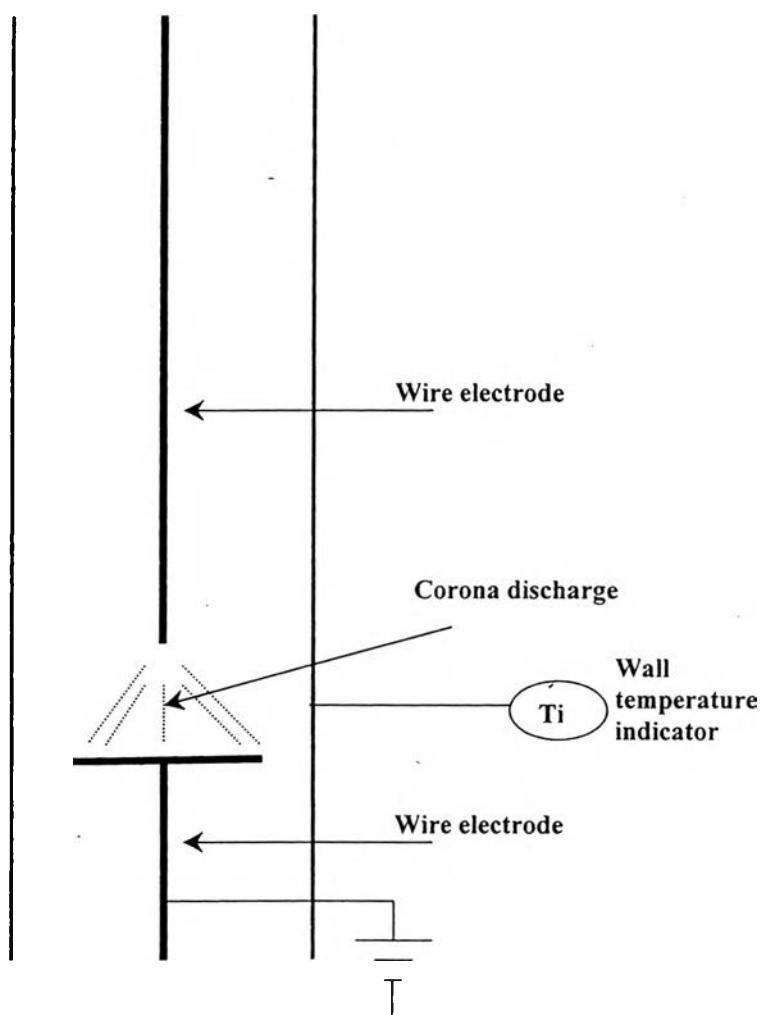


Figure 3.3 Plasma reactor

### 3.3 Experimental Procedure

The experiment was divided into two parts. The first part was to experiment with only methane and air for studying the effects of voltage, frequency, ratio of component, flow rate and gap width. The second was to experiment with methane, air and ethane for studying the effects of ethane addition, voltage and frequency. All of experiments were run at atmospheric pressure and room temperature.

#### 3.3.1 Methane/Air System

For the methane/ air system, the effects of voltage, frequency, methane partial pressure, gas flow rate, and the electrode gap width on methane conversion and product selectivities were determined.

##### 3.3.1.1 *Study of Voltage Effect*

The feed gas was a mixture of methane and air having a constant ratio of methane to air of 3:4.8, which corresponded to  $\text{CH}_4:\text{O}_2:\text{N}_2=3:1:3.8$ . The total flow rate of the feed gas was kept constant at 100 ml/min. The gap width of the electrodes was set at 0.9 cm. The supply power was then turned on at the desired voltage and frequency. The frequencies of 300 and 400 Hz were selected in this experiment and the applied voltage varied from 5,000 to 9,000 V. The feed gas was then continuously fed to the reactor for measuring the exact gas composition. Next, the gas composition of the outlet stream was examined by GC every 1 hour. The system reached a steady state condition when the compositions of the outlet gas was relatively constant. The current was also measured every 20 minutes. The flow rate of the outlet gas was measured by using the digital flow meter. Afterwards, the experiment was repeated by changing the new voltage while the others kept constant.

### 3.3.1.2 Study of Frequency Effect

It was found that the coke deposit on the electrode surface appeared when the system was operated under frequency below 300 Hz. Therefore in this experiment study frequency had to be varied from 300 to 700 Hz. The mixture was kept constant at a methane to air ratio of 3:4.8 ( $\text{CH}_4:\text{O}_2:\text{N}_2= 3:1:3.8$ ) while the total flow rate of 100 ml/min and a gap width of 0.9 cm were set in the experiment. In addition, the voltage was fixed at 6,500 and 9,000 V. Next, the gas compositions of the inlet and outlet streams were examined by GC every 1 hour. The system was assumed a steady state condition when the composition of the outlet gas was relatively constant. The current was also measured every 20 minutes. It was found that the current varied in a some extent due to its nature of nonequilibrium plasma. Hence, an average value of the current was taken to represent each run. The flow rate of the outlet gas was measured by using the digital flow meter. Afterwards, the experiment was repeated by changing a new frequency while the others kept constant.

### 3.3.1.3 Study of Methane Partial Pressure Effect

In this experiment, there were three different  $\text{CH}_4$  to air ratios of experiments; 2:1, 3:1 and 4:1 corresponding to  $\text{CH}_4:\text{O}_2:\text{N}_2= 2:1:3.8$ ,  $\text{CH}_4:\text{O}_2:\text{N}_2= 3:1:3.8$ , and  $\text{CH}_4:\text{O}_2:\text{N}_2= 4:1:3.8$ , respectively. The studied system was run at a total flow rate of 100 ml/min, the gap width of 0.9cm, an applied voltage of 9,000 V and a frequency of 300 Hz. After the desired composition of feed gas was obtained, the supply power unit was then turned on at the desired voltage and frequency. Next, the gas composition of the outlet stream was examined by GC. Then, the similar procedure was taken as described before.

### 3.3.1.4 Study of Flow Rate Effect

The flow rate was varied from 50 to 125 ml/min while a ratio of methane to air of 3:4.8 ( $\text{CH}_4:\text{O}_2:\text{N}_2= 3:1:3.8$ ), a gap width of 0.9 cm.



An applied voltage of 9,000 V and the frequency of 300 Hz were set in this experimental study. The similar procedure was taken as described before.

#### *3.3.1.5 Study of Gap Width Effect*

The gap width was varied from 0.7 to 1.9 cm while a ratio of methane to air of 3:4.8 ( $\text{CH}_4:\text{O}_2:\text{N}_2 = 3:1:3.8$ ). The total flow rate of 100 ml/min, the applied voltage of 9,000 V and the frequency of 300 Hz were set for this experiment. Again the same procedure was then taken as describe before.

### 3.3.2 Methane/Air/Ethane System

#### *3.3.2.1 Study of Voltage Effect*

In this experimental study, ethane was introduced in order to determine its effects on the partial oxidation of methane. A ratio of  $\text{CH}_4:\text{C}_2\text{H}_6:\text{air}$  in the feed mixture was kept at 2:1:4.8 corresponding to  $\text{CH}_4:\text{C}_2\text{H}_6:\text{O}_2:\text{N}_2$  of 2:1:1:3.8. The applied voltage was only varied from 5,000 to 7,250 V while the other parameters were kept constant throughout the experiment. The total flow rate of the feed mixture was 100 ml/min and the gap width was 0.5 cm. In addition, the frequency of 300 Hz was chosen for this experiment. Again, the similar procedure was carried out as described before.

#### *3.3.2.2 Study of Frequency Effect*

In this experiment, the frequency was only varied from 300 to 700 Hz while the other parameters were kept constant. The ratio of  $\text{CH}_4$  to  $\text{C}_2\text{H}_6$  to air was 2:1:4.8. The total flow rate was 100 ml/min. The gap width of 0.9 cm and applied voltage of 9,000V were set in this experiment. The similar procedure was then taken as described before.