CHAPTER III
METHODOLOGY

3.1 Materials

Reagent Gases for Reaction

All gases used for reaction testing were obtained from Thai Industrial Gas Co., Ltd, as follows:

1) Helium (HP grade)
2) 40% Ethylene balanced with helium
3) 97% Oxygen balanced with helium
4) 30% Ethylene oxide balanced with helium

3.2 Reaction Activity Testing

The ethylene epoxidation experiments were performed by using a corona discharge reactor, which was operated at atmospheric pressure and ambient temperature, around 25–27 °C (room temperature). The schematic of experimental setup for ethylene epoxidation reaction using a corona discharge system is shown in Figure 3.1(a), and the detailed schematic of corona discharge reactor is shown in Figure 3.1(b). The reactor comprised an 11.25-inch-long quartz tube with an outer diameter of 10 mm and an inner diameter of 8 mm. Plasma was generated in the gap between pin and plate electrodes, which were located at the center of reactor. The power used to generate plasma was alternating current power, 220 V and 50 Hz, which was adjusted by a function generator, whereas the sinusoidal waveform is monitored by an oscilloscope.

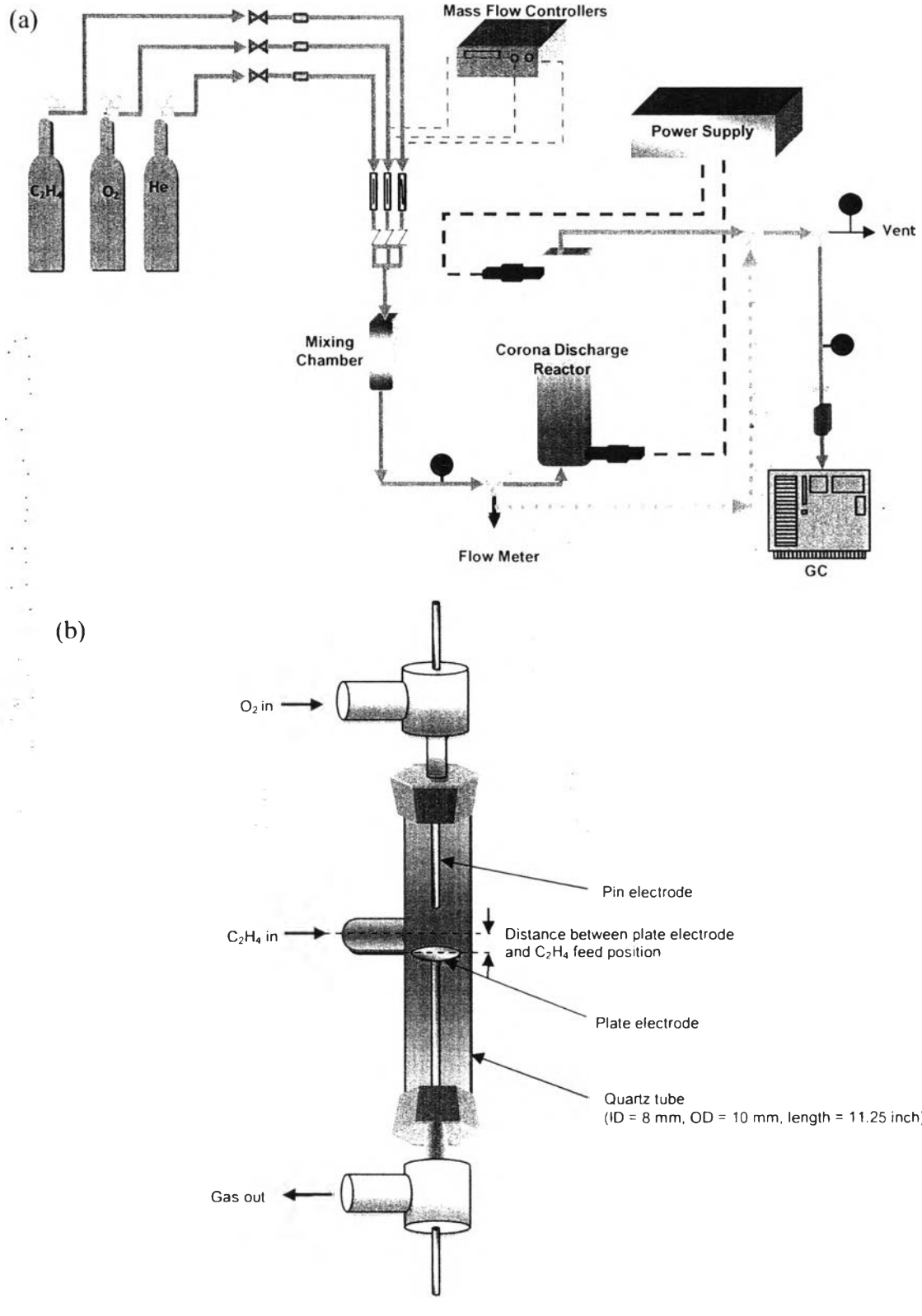

Figure 3.1 (a) Schematic of experimental setup for ethylene epoxidation reaction using a corona discharge reactor and (b) detailed schematic of corona discharge reactor.

The reactant gases (ethylene and oxygen, balance with helium) flowing through the reactor were controlled by a set of electronic mass flow controllers. All reactant lines had 7 μm in-line filters before passing through the mass flow controllers in order to trap any foreign particles. The reactor pressure was controlled via a needle valve. The outlet of reactor either was vented to the atmosphere via rubber tube exhaust or entered an on-line gas chromatograph (GC) to analyze the product gases. The moisture in the effluent gas was removed by a water trap before entering the on-line GC. The GC was equipped with both a thermal conductivity detector (TCD) and a flame ionization detector (FID). For the TCD channel, a packed column (Carboxen 1000) was used for separating the product gases, which were hydrogen (H_2), oxygen (O_2), carbon monoxide (CO), carbon dioxide (CO_2), and ethylene (C_2H_4). For FID channel, the capillary column (OV-Plot U) was used for the analysis of ethylene oxide (EO) and other by-product gases, i.e. CH_4, C_2H_2, C_2H_6, and C_3H_8. The compositions of the product gas stream were determined by the GC every 20 min. When the system reached steady state, an analysis of the outlet gas compositions was performed at least three times. The experimental data under steady state conditions were averaged and then used to evaluate the performance of the plasma system. The GC was operated under the following conditions:

TCD injection temperature	120 °C
FID injection temperature	150 °C
Oven temperature	40 °C for 5 min
	165 °C (heating rate 10 °C/min)
Carrier gas	High purity helium
Carrier gas flow rate	30 cm^3/min (TCD channel)
	25 cm^3/min (FID channel)
Detector temperature	190 °C in TCD and 280 °C for FID

To evaluate the system performance, the C_2H_4 and O_2 conversions and the selectivity for products, including EO, CO, CO_2, H_2, CH_4, C_2H_2, C_2H_6, and traces of C_3, were considered. The conversion of either C_2H_4 or O_2 was calculated from the following equation:

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

The product selectivity was calculated from the following equation:

$$\% \text{ Product selectivity} = \frac{[(\text{number of carbon or hydrogen atom in product})(\text{moles of product produced})]}{\sum [(\text{number of carbon or hydrogen atom in product})(\text{moles of product produced})]} \times 100$$

The ethylene oxide yield was calculated from the following equation:

$$\% \text{ Ethylene oxide yield} = (\% \text{ ethylene conversion}) \times (\% \text{ ethylene oxide selectivity}) / 100$$

To determine the energy efficiency of the plasma system, the specific power consumption was calculated in a unit of Ws per molecule of converted ethylene or per molecule of produced ethylene oxide using the following equation:

$$\text{Specific power consumption} = \frac{P \times 60}{N \times M}$$

where P = Power (W)

N = Avogadro's number = 6.02×10^{23} molecules/mol

M = Rate of converted ethylene molecules in feed or rate of produced ethylene oxide molecules (mol/min).

3.3 Power Supply Unit

The block diagram of the power supply unit is shown in Figure 3.2. For the first step, the AC input of 220 V and 50 Hz was converted to DC of about 70-80 V by a DC power supply converter. For the second step, 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 alternating current was a sinusoidal waveform. For the final step, the amplified AC passed through the input 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 (input). An Extech® series 380801 power analyzer was used to measure current, frequency, and voltage at the low side of the power supply unit.

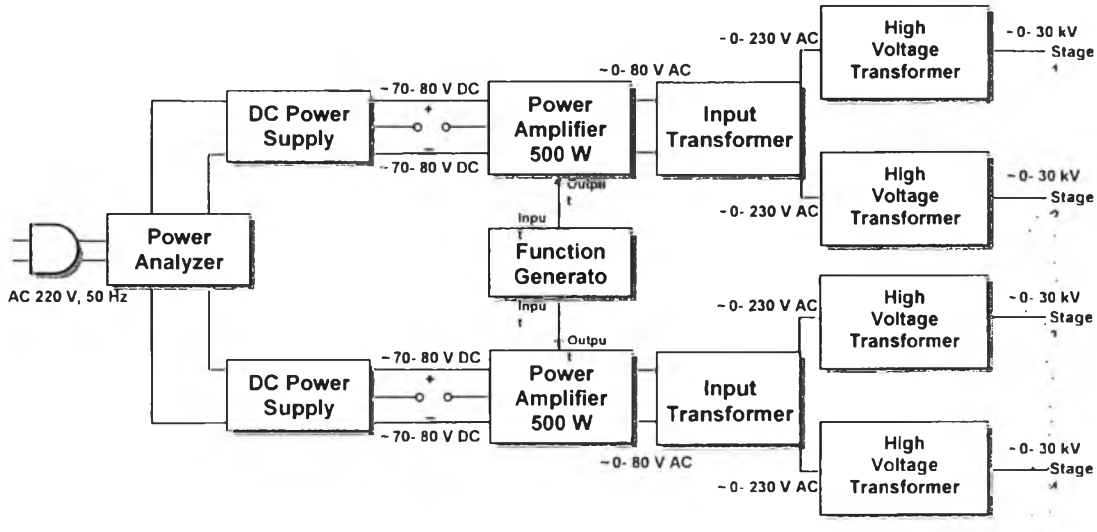

Figure 3.2 Block diagram of the power supply unit.

3.4 Studied Conditions

After the compositions of the feed remained constant, the power supply unit was turned on. After 60 min, the compositions of the effluent gas were analyzed for every time interval of 20 min until the gas compositions were invariant. After reaching a steady state, reactant conversion, production selectivity, product yield, and power consumption were calculated for evaluating the system performance. The conditions for all studied parameters (i.e. distance between plate electrode and C_2H_4 feed position, O_2/C_2H_4 feed molar ratio, applied voltage, input frequency, total feed flow rate, and gap distance between pin and plate electrodes) are shown as follows:

Distance between plate electrode and C_2H_4 feed position	0.1–0.5 cm
O_2/C_2H_4 feed molar ratio:	0.25:1–1:1
Applied voltage:	15–19 kV
Input frequency:	400–700 Hz
Total feed flow rate:	100–150 cm^3/min
Gap distance between pin and plate electrodes:	0.8–1.2 cm