



CHAPTER III EXPERIMENTAL

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) 10% Ethylene oxide balanced with carbon dioxide.

3.2 Reaction Activity Measurement

The experimental study of ethylene epoxidation was comparatively conducted using dielectric barrier discharge (DBD) reactors with two electrode geometries, i.e. parallel DBD and cylindrical DBD, which were operated at ambient temperature and atmospheric pressure. The schematics of experimental setup for ethylene epoxidation reaction using both DBD systems are shown in Figure 3.1. For the parallel DBD system, the reactor configuration is shown in Figure 3.2(a). The reactor sizes were 1.5 cm height x 5.5 cm width x 17.5 cm length for inner dimension, and 3.9 cm height x 9.5 cm width x 21.5 cm length for outer dimension. Between the two electrodes, there was a dielectric glass plate. The gap distance between the two electrodes was fixed at 5 mm. For the cylindrical DBD system, the reactor configuration is shown in Figure 3.2(b). The reactor comprised two concentric electrode tubes. The inner electrode had 5 mm OD and 13.67 cm height, and the outer electrode had 19.05 mm OD, 15.8 mm ID and 16.2 cm height. Between the two concentric electrodes, there was a dielectric glass tube. The gap distance between two electrodes was also fixed at 5 mm. The microdischarge plasma was generated in the electrode gap. The detailed dimensions of both DBD reactors are given in Table 3.1. The power used to generate plasma was alternating current power, 220 V and 50 Hz, which was transmitted to a high voltage current via a

power supply unit. The output voltage was adjusted by a function generator, whereas the sinusoidal wave signal was controlled and monitored by an oscilloscope.

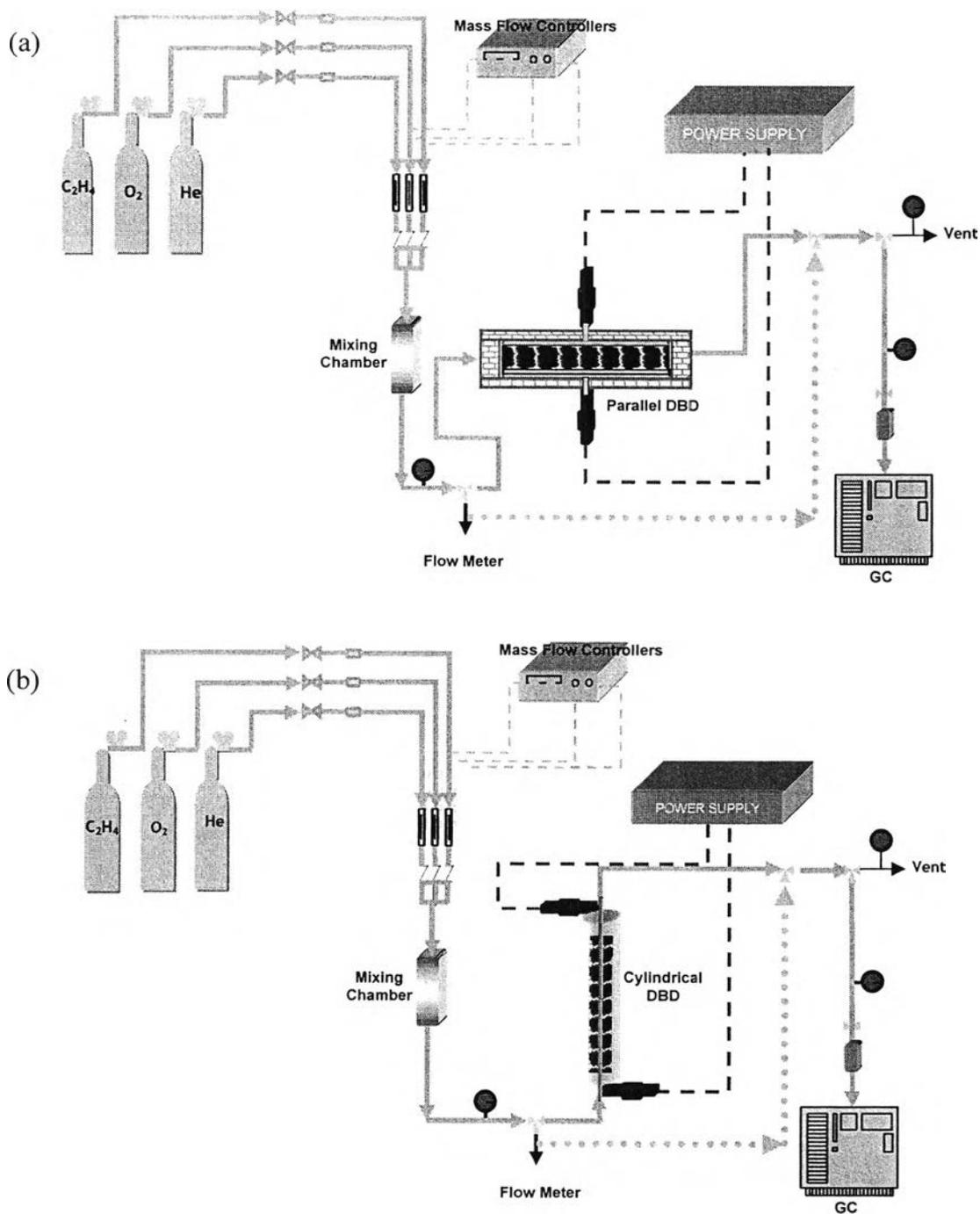


Figure 3.1 Schematics of experimental setup for ethylene epoxidation reaction using (a) parallel DBD and (b) cylindrical DBD.

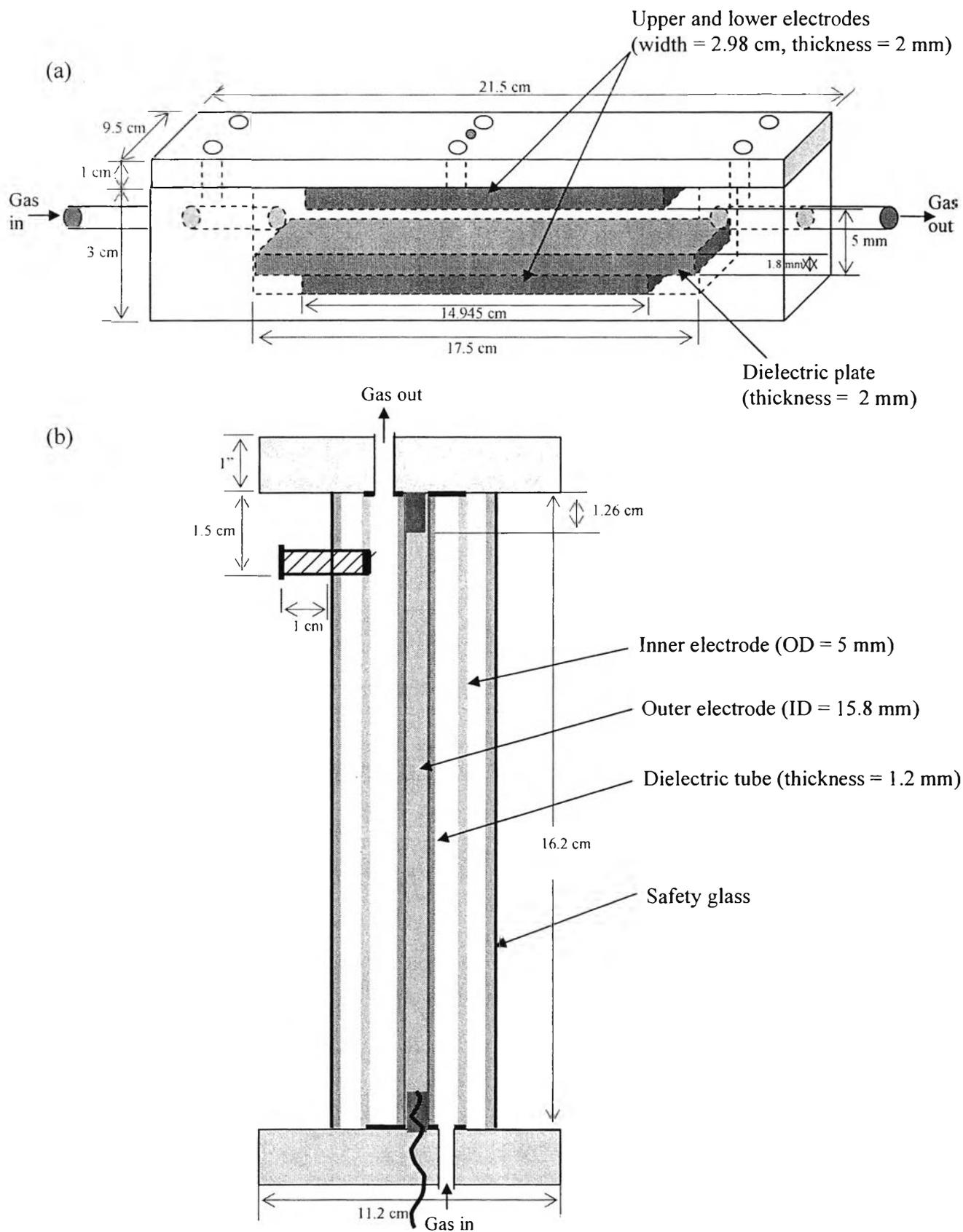


Figure 3.2 Schematics of reactor setup for (a) parallel DBD and (b) cylindrical DBD.

Table 3.1 The detailed dimensions of both DBD reactors

Dimension	Parallel DBD	Cylindrical DBD
Electrode width (cm)	2.98	inner electrode = 0.5 (OD) outer electrode = 1.58 (ID) = 1.905 (OD)
Electrode length (cm)	14.945	inner electrode = 13.67 outer electrode = 16.2
Electrode thickness (cm)	0.14	-
Electrode gap distance (cm)	0.5	0.84
Glass thickness (cm)	0.18	0.12
Electrode edge length (cm)	71.70	13.07
Electrode surface area (cm ²)	89.07	89.36
Reaction volume (cm ³)	14.25	20.93
Electrode edge length-to-reaction volume ratio (cm ⁻²)	5.03	0.63
Electrode surface area-to-reaction volume ratio (cm ⁻¹)	6.25	4.27

The flow rates of ethylene, oxygen, and helium flowing through the reactor were controlled by electronic mass flow controllers. Impurities in all reactant gases were trapped by 7 μm in-line filter before the reactant gases passed through electronic mass flow controllers. A mixture of 40% ethylene in helium, pure oxygen, and pure helium was used as feed gases with $\text{O}_2/\text{C}_2\text{H}_4$ molar ratio ranging from 0.2:1 to 1:1. The reactor pressure was controlled by a needle valve, and the outlet of reactor was vented to the atmosphere via an exhausted rubber tube before entering an on-line gas chromatograph. The moisture in the product gas stream was trapped by a water trap filter before entering a heated stainless steel line to an on-line gas chromatograph. The composition of product gases was analyzed by an automatic gas chromatograph (Perkin-Elmer, AutoSystem GC). The gas chromatograph was equipped with both a thermal conductivity detector (TCD) and a flame ionization detector (FID). For the TCD channel, the packed column (Carboxen 1000) was used

for separating the product gases, which were hydrogen (H₂), oxygen (O₂), carbon monoxide (CO), and carbon dioxide (CO₂). For the FID channel, the capillary column (OV-Plot U) was used for analysis of ethylene (C₂H₄), ethylene oxide (C₂H₄O), and other product gases, i.e. CH₄, C₂H₂, C₂H₆, and C₃H₈. The GC was operated under the following conditions:

TCD injector temperature	120°C
FID injector temperature	150°C
TCD detector temperature	190°C
FID detector temperature	280°C
TCD range	4
FID range	1
TCD carrier gas flow rate	30 ml/min
FID carrier gas flow rate	25 ml/min
Carrier gas	High purity helium
Oven temperature	40°C for 5 min 165°C (heating rate 10°C/min)

To evaluate the process performance, the conversions of ethylene and oxygen, the selectivities for products, including H₂, CO, CO₂, C₂H₄O, CH₄, C₂H₆, C₂H₂, and traces of C₃, and the ethylene oxide yield were considered. The conversion of either ethylene or oxygen was defined as:

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

The product selectivity was calculated from the following equation:

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

The ethylene oxide yield was calculated from the following equation:

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

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 3.3. For the first step, the AC input of 220 V and 50 Hz was converted to DC of about 70-80 V by 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 the 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 a nominal factor of 130 times of low side (input). An Extech[®] series 380801 power analyzer was used to measure power, power factor, current, frequency, and voltage at the low side of the power supply unit.

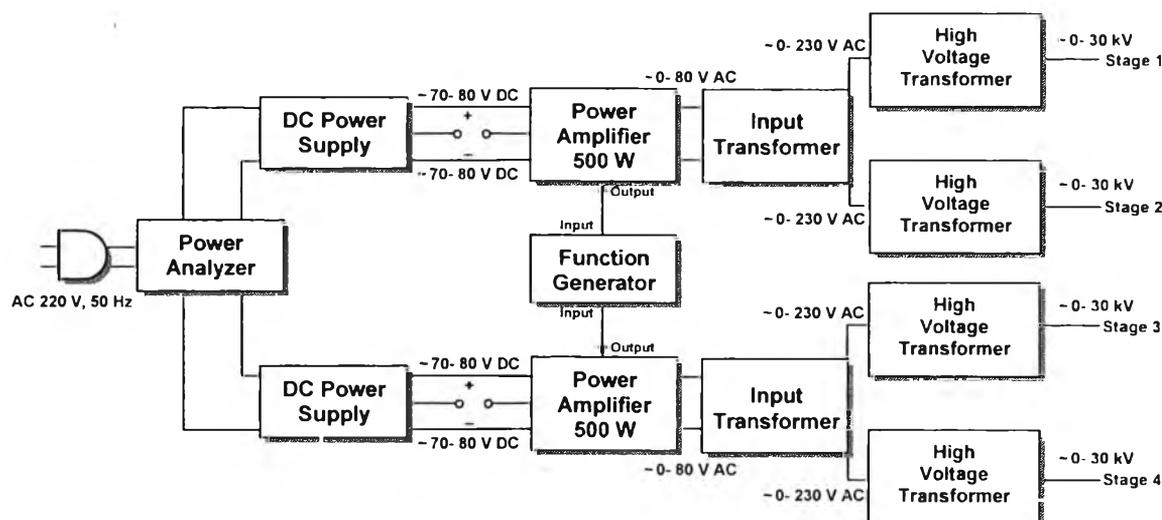


Figure 3.3 Block diagram of the power supply unit.

3.4 Studied Conditions

After the composition of the feed remained constant, the power supply unit was turned on. After 60 min, the composition of the effluent gas was analyzed for every time interval of 20 min until the exhaust gas composition was invariant. After reaching a steady state, reactant conversion, product selectivity, product yield, and power consumption were calculated for evaluating the system performance. The experiments were performed by the two DBD reactors with different electrode geometries. The effects of various operating parameters, including O_2/C_2H_4 molar ratio, applied voltage, input frequency, and feed flow rate, were comparatively investigated.

The experimental conditions for this research were as follows:

O_2/C_2H_4 molar ratio:	0.2:1-1:1
Applied voltage:	13-19 kV (Cylindrical DBD) 15-21 kV (Parallel DBD)
Input frequency:	400-800 Hz
Feed flow rate:	50-150 cm^3/min
Gap distance:	5 mm