CHAPTER VIII
CONCLUSIONS AND RECOMMENDATIONS

8.1 Conclusions

In this research, the epoxidation of ethylene to EO was studied in four different non-thermal plasma systems. For a first part, a parallel-plate DBD reactor, cooperating with the Al_2O_3- and SiO_2-supported Ag catalysts was used to investigate the ethylene epoxidation. The presence of Ag catalysts was found to greatly enhance EO selectivity. The DBD system with the Ag catalysts on the SiO_2 support was found to provide a better epoxidation performance, as compared to these on the Al_2O_3 particles. The optimum Ag loading on the SiO_2 particles was found to be 20 wt.%, resulting in a maximum EO selectivity of 30.6% with the minimum H_2 and CO selectivities.

For a second part, a cylindrical DBD system with separate C_2H_4/O_2 feed was investigated for ethylene epoxidation. The effects of the C_2H_4 feed position and operational parameters on ethylene epoxidation reaction were investigated to find out best operating conditions. A C_2H_4 feed position of 0.25, an O_2/C_2H_4 feed molar ratio of 1/4, an applied voltage of 13 kV, an input frequency of 550 Hz, and a total feed flow rate of 75 cm^3/min provided the highest ethylene epoxidation performance, in terms of maximum EO selectivity of 33.9% and yield of 7.5% with both minimum CO selectivity and power consumption per EO molecule produced. The system with the separate C_2H_4/O_2 feed at a C_2H_4 feed position of 0.25 provided a superior epoxidation performance in terms of a higher EO selectivity and yield, lower CO selectivity, and lower power consumption per EO molecule produced, as compared with the system with the mixed C_2H_4/O_2 feed.

For a third part, a corona discharge reactor with different C_2H_4 feed position was employed for ethylene epoxidation. The highest EO selectivity was obtained at the most suitable C_2H_4 feed position of 2 mm. In addition, the O_2/C_2H_4 feed molar ratio also significantly influenced ethylene epoxidation activity and the optimum O_2/C_2H_4 feed molar ratio of 0.5:1 was needed to operate with the highest efficiency. An increase in applied voltage increased O_2 conversion, EO selectivity and yield, and reduced energy consumption, in contrast to the effect of input frequency. Increasing applied voltage and decreasing frequency increases the number of energetic electrons in the discharge zone. The total feed flow rate has also a significant effect on the residence time of gas molecules in the plasma zone, affecting the overall performance of the plasma system, as well as the gap distance between pin and plate electrodes. The optimum conditions for acheiving the highest EO selectivity of 8.4% and yield of 1.8% were a C_2H_4 feed position of 2 mm, an O_2/C_2H_4 feed molar ratio of 0.5:1, an applied voltage of 18 kV, an input frequency of 500 Hz, a total feed flow rate of 100 cm^3/min, and a gap distance between pin and plate electrodes of 1 cm.

For a last part, a DBD jet reactor with a separate C_2H_4/O_2 feed was used to enhance epoxidation efficiency. From the results, the DBD jet system provided obviously high potential for ethylene epoxidation with very low power consumption. The highest EO selectivity of 55.2% and yield of 27.6%, as well as the lowest power consumption were obtained at a total feed flow rate of 1,625 cm^3/min, an O_2/C_2H_4 feed molar ratio of 0.25:1, an applied voltage of 9 kV, an input frequency of 300 Hz, and an inner electrode position of 0.3 mm.

The ethylene epoxidation performance of the catalytic process with the parallel plate DBD system, the sole cylindrical DBD, corona discharge and DBD jet system was comparatively studied as compared to the previous works (Chavadej *et al.*, 2008; Sreethawong *et al.*, 2008; Sreethawong *et al.*, 2009; Sreethawong *et al.*, 2010) as shown in Table 8.1 and 8.2. The DBD jet system greatly enhanced the ethylene epoxidation activity in terms of the highest C_2H_4 and O_2 conversions as well as the highest EO selectivity (based on total mole of product) and yield, but it required very low power consumption. However, there was high quantity of coke formation depositing on the inner electrode surface. The active catalysts and redesigned configuration of electrode are interesting to further study in order to reduce coke formation and to improve the efficiency of epoxidation process. Comparing all of the plasma systems, the generated current under the DBD jet system was the lowest, leading to lower the opportunity of C_2H_4 molecules/highly energetic electrons collision. Besides, the C_2H_4 molecules were directly injected at the end of plasma region, resulting in reduction of C_2H_4 cracking as well as secondary side reactions. Therefore, selectivities for other products such as H_2, CO, and C_2H_2 were the lowest under the DBD jet system. Comparing this work with the previous work, the sole DBD jet system with the separate feed still provided much higher EO selectivity and yield as well as extremely lower power consumption.

Table 8.1 Comparisons the ethylene epoxidation performance of the sole plasma systems and combined catalytic-plasma systems

Plasma reactors	Conversions (%)		EO selectivity (%)		EO yield (%)	Power consumption (x10^{16} Ws/molecule)	
	C_2H_4	O_2	*	**		C_2H_4 converted	EO produced
Part1-Parallel DBD with 20 wt.% Ag/ SiO_2	7.0	64.36	30.6	16.81	2.1%	4.6	15.26
Part2-Cylindrical DBD	3.68	74.57	203.16	33.85	7.47	3.49	5.21
Part3-Corona discharge	2.25	44.33	78.12	8.42	1.76	4.74	6.07
Part4- DBD jet	49.95	91.23	55.19	85.24	27.57	0.000030	0.000065
Corona discharge with 12.5 wt.% Ag/(LSA)α-Al_2O_3 (Chavadej *et al.*, 2008)	27.18	57.62	11.91	16.54	3.24%	1.7	12.6
Parallel DBD (Sreethawong *et al.*, 2008)	90.95	93.67	6.18	9.70	5.62%	0.37	6.07
Corona discharge with 0.2 wt.% Au-12.5 wt.% Ag/ α-Al_2O_3 (Sreethawong *et al.*, 2009)	46.04	65.23	9.39	14.55	4.32%	0.62	6.5
Cylindrical DBD (Sreethawong *et al.*, 2010)	18.86	91.38	13.5	5.71	1.8%	0.75	5.9

* Selectivity based on C_2H_4 conversion

** Selectivity based on total mole of product

Table 8.2 Comparisons the selectivities for other products and the current of the sole plasma systems and combined catalytic-plasma systems

Plasma reactors	H_2 selectivity(%)		CO selectivity (%)		C_2H_2 selectivity (%)		Current (mA)
	*	**	*	**	*	**	
Part1-Parallel DBD with 20 wt.% Ag/ SiO_2	61.56	33.86	29.62	16.30	49.49	27.22	0.805
Part2-Cylindrical DBD	40.07	13.35	16.27	5.42	255.23	42.53	0.756
Part3-Corona discharge	123.48	26.63	212.60	45.85	101.94	10.99	0.539
Part4- DBD jet	4.04	12.48	0.66	2.04	0	0	0.441
Corona discharge with 12.5 wt.% Ag/(LSA)α-Al_2O_3 (Chavadej *et al.*, 2008)	0	0	24.72	34.29	2.62	30.34	0.685
Parallel DBD (Sreethawong *et al.*, 2008)	15.83	24.86	24.06	37.78	0.46	0.73	0.854
Corona discharge with 0.2 wt.% Au-12.5 wt.% Ag/ α-Al_2O_3 (Sreethawong *et al.*, 2009)	0	0	52.03	47.21	0.02	0.18	0.769
Cylindrical DBD (Sreethawong *et al.*, 2010)	62.99	28.20	78.37	35.08	60.55	27.11	0.768

* Selectivity based on C_2H_4 conversion

** Selectivity based on total mole of product

8.2 Recommendations

The recommendations for future work are as follows:

1. In addition to O_2, nitrous oxide (N_2O) is also used as an oxygen source, especially for propylene epoxidation. N_2O creates less oxygen species than O_2 that should prefer to ethylene epoxidation because this reaction with high efficiency is obtained at an O_2-lean condition. Therefore, the effect of nitrous oxide (N_2O) as oxygen source on ethylene epoxidation should be investigated.
2. Methane is commercially used as a balancing gas in conventional catalytic processes. The ethylene epoxidation should be investigated in a low-temperature plasma system to clearly understand about the role of methane on the chemical pathways and how it affects the plasma system.
3. The gap distance between two electrodes of a DBD reactor is one of the most parameters that significantly affect the plasma behavior. To study the effect of the gap distance, corresponding to the plasma volume and residence time is of interest.