



## REFERENCES

- Blazkova, A., Csolleova, I., and Brezova, V. (1998). Effect of light sources on the phenol degradation using Pt/TiO<sub>2</sub> photocatalysts immobilized on glass fibers. *Journal of Photochemistry and Photobiology A: Chemistry*, 113, 251-256.
- Cheng, Y. (1996). Kinetic and Mechanistic Studies of Volatile Organic Compound Oxidation Catalysis Using Thin Film Model Pt Catalysts. A Research Proposal Submitted in partial Fulfillment of The Preliminary Examination Requirements, The University of Michigan.
- De Lasa, H.I., Dogu, G., and Ravella, A. (Eds.). (1992). Chemical Reactor Technology for Environmentally Safe Reactors and Product. Dordrecht/Boston/London: Kluwer Academic Publishers, 577-608.
- De Nevers N. (1995). Air Pollution Control Engineering. International Editions. New York: McGRAW-HILL.
- Einaga, H., Futamura, S., and Ibusuki, T. (2001). Complete oxidation of benzene in gas phase by platinized titania photocatalysts. *Environmental Science & Technology*, 35(9), 1880-1884.
- Eliasson, B., Hirth, M., and Kogelschatz, U. (1987). Ozone synthesis from oxygen in dielectric barrier discharge. *Journal of Applied Physics*, 20, 1421-1437.
- Eliasson, B. and Kogelschatz, U. (1991). Nonequilibrium volume plasma chemical processing. *IEEE Transactions on Plasma Science*, 19(6), 1063-1077.
- Futamura, S. and Yamamoto, T. (1997). Byproduct identification and mechanism determination in plasma chemical decomposition of trichloroethylene. *IEEE Transactions on Industry Applications*, 33(2), 447-453.
- Futamura, S., Zhang, A., and Yamamoto, T. (1999). Mechanisms for formation of inorganic byproducts in plasma chemical processing of hazardous air pollutants. *IEEE Transactions on Industry Applications*, 35(4), 760-766.
- Futamura, S., Einaga, H., and Zhang, A. (2001). Comparison of reactor performance in the nonthermal plasma chemical processing of hazardous air pollutants. *IEEE Transactions on Industry Applications*, 37(4), 978-985.
- Grill, A. (1994). Cold Plasma in Materials Fabrication: From Fundamentals to

- Applications. IEEE Press: New York.
- Harndumrongsak, B. (2002). Oxidation of ethylene in a plasma environment. M.Sc., Thesis, Chulalongkorn University, Bangkok.
- Harndumrongsak, B., Lobban, L.L., Rangsunvigit, P., and Kitiyanan, B. (2002). Oxidation of Ethylene in plasma environment. Proceeding of the 9<sup>th</sup> APCCChE Congress in Christchurch, New Zealand 29 September – 3 October 2002.
- Herrmann, J.-M. (1999). Heterogeneous Photocatalysis: Fundamentals and Applications to the Removal of Various Types of Aqueous Pollutant. Catalysis Today, 53, 155-129.
- Hill, B.J. Master Thesis, University of Oklahoma, 1997.
- Huang, A., Xia, G., Wang, J., Suib, S.L., Hayashi, Y., and Hiroshige, M. (2000). CO<sub>2</sub> reforming of CH<sub>4</sub> by atmospheric pressure ac discharge plasmas. Journal of Catalysis, 189, 349-359.
- Huang, L., Nakajyo, K., Hari, T., Ozawa, S., and Matsuda, H. (2001). Decomposition of carbon tetrachloride by a pulsed corona reactor incorporated with in situ absorption. Industrial and Engineering Chemistry Research, 40, 5481-5486.
- Kruapong, A. (2000). Partial oxidation of methane to synthesis gas in low temperature plasmas. M.Sc., Thesis, Chulalongkorn University, Bangkok.
- Litter, M.I. (1999). Heterogeneous photocatalysis transition metal ions in photocatalytic systems. Applied Catalysis B: Environmental, 13, 89-114.
- Liu, C., A. Marafee, B. Hill, G. Xu, R. Mallinson, and L. Lobban, "Oxidative Coupling of Methane with ac and dc Corona Discharge," *Ind. Eng. Chem. Res.*, 35, 3295 (1996).
- Malik, M.A. and Malik, S.A. (1999). Catalyst enhanced oxidation of VOCs and methane in cold-plasma reactors. Platinum Metal Review, 43(3), 109-113.
- Morinaga, K., Suzuki, M. *Bull. Chem. Soc. of Japan*. 1961, 34(2), 157-161.
- Morinaga, K., Suzuki, M. *Bull. Chem. Soc. of Japan*. 1962, 35(2), 204-207.
- Nakamura, I., Negishi, N., Kutsuna, S., Ihara, T., Sugihara, S., and Takeuchi, K. (2000). Role of oxygen Vacancy in the plasma-treated TiO<sub>2</sub> photocatalyst with visible light activity for NO removal. Journal of Molecular Catalyst A: Chemical, 161, 205-212.

- Nasser, E. (1971). Fundamentals of gaseous ionization and plasma electronics. USA: John Wiley & Sons, Inc.
- Obuchi, E., Sakamoto, T., and Nakano, K. (1999). Photocatalytic Decomposition of Acetaldehyde over TiO<sub>2</sub>/SiO<sub>2</sub> Catalyst. Chemical Engineering Science, 57, 1525-1530.
- Papaethimiou, P., Ioanides, T., and Verykios, X.E. (1997). Combustion of non-halogenated volatile organic compounds over group VIII metal catalysts. Applied Catalysis B: Environmental, 13, 175-184.
- Peral, J., Domenech, X., and Ollis, D.F. (1997). Heterogeneous Photocatalyst for Purification, Decontamination and Deodorization of Air. Journal of Chemical Technology and Biotechnology, 70, 117-140.
- Robertson, P.K.J. (1996). Semiconductor photocatalysis: An environmentally acceptable alternative production technique and effluent treatment process. Journal of Cleaner Production, 4:3-4, 203-212.
- Rosacha, L.A., Anderson, G.K., Bechtold, L.A., Coogan, J.J., Heck, H.G., Kang, M., McCulla, W.H., Tennant, R.A., and Wantuck, P.J. (1993). Treatment of hazardous organic wastes using silent discharge Plasmas. Non-Thermal Plasma Technique for Pollution Control., NATO ASI series, 34, part B, 128-139.
- Sano, N., Nagamoto, T., Tamon, H., Suzuki, T., and Okazaki, M. (1997). Removal of acetaldehyde and skatole in gas by a corona-discharge reactor. Industrial & Engineering Chemistry Research, 36, 3783-3791.
- Sutthiruangwong, S. (1999). Plasma catalytic production of methanol. M.Sc., Thesis, Chulalongkorn University, Bangkok.
- Thanyachotpaiboon, K., Chavadej, S., Caldwell, L., Lobban, L.L., and Mallinson, R.G. (1998). Conversion of methane to higher hydrocarbons in AC nonequilibrium plasmas. AIChE Journal, 44(10), 2252-2257.
- Tsai, C.H., Lee, W.J., Chen, C.Y., and Liao, W.T. (2001). Decomposition of CH<sub>3</sub>SH in a RF plasma reactor: reaction products and mechanisms Industrial & Engineering Chemistry Research 40, 2384 –2395.

Zhang, J., Ayusawa, T., Minagawa, M., Kinugawa, K., Yamashita, H., Matsuoka, M., and Anpo, M. (2001). Investigations of TiO<sub>2</sub> photocatalysts for the decomposition of NO in the flow system. Journal of Catalysis, 198, 1-8.

## APPENDICES

### Appendix A Assumptions, Definitions, and Calculations

In this work, the following assumptions were made:

1. All the gaseous behaviors obey the ideal gas law
2. The change in the system, pressure is very small and negligible.
3. The pressure in the system equals the atmospheric pressure (1 atm)

The total molar flow rate of the gaseous stream can be determined from the following equation:

$$N = q \times (P/RT) \quad (B.1)$$

where

$q$  = total volumetric flow rate

$P$  = total pressure of the system

$R$  = gas constant ( $82.051 \text{ atm}\cdot\text{ml}\cdot\text{mol}^{-1}\cdot\text{min}^{-1}\cdot\text{K}$ )

$T$  = absolute ambient temperature (K)

The molar flow rate of each component can be obtained by multiplying its fraction derived from the gas chromatography analysis by the total molar flow rate.

The conversion is defined as:

$$\% \text{ Conversion} = \frac{\text{Mole reactant in} - \text{Mole reactant out}}{\text{Mole reactant in}} \times 100 \quad (B.2)$$

The first selectivity is defined as:

$$\% \text{ Selectivity} = \frac{P \times \text{Mole of } C_p \text{ produced}}{R \times \text{Mole of } C_R \text{ converted}} \times 100 \quad (B.3)$$

where

$P$  = number of carbon atom in product

$R$  = number of carbon atom in reactant

$C_p$  = product that has carbon  $P$  atom

$C_R$  = reactant that has carbon  $R$  atom

The second selectivity is defined as:

$$\% \text{ Selectivity} = \frac{\text{Mole of product}}{\text{Total mole of all products}} \times 100 \quad (\text{B.4})$$

To determine the energy efficiency of corona discharge system, the specific energy consumption was calculated in a unit of electron-volt per molecule of converted carbon (eV/m<sub>c</sub>) from the following equation:

$$\text{Specific energy consumption} = \frac{P \times 60}{(1.602 \times 10^{-19}) \times \tilde{N} \times M_C} \text{ eV/ mol C}_2\text{H}_4$$

Where P = Power (W)

$\tilde{N}$  = Avogadro's number =  $6.02 \times 10^{23}$  molecules.g-mole<sup>-1</sup>

M<sub>C</sub> = Rate of carbon in feed gas converted (g-mole.min<sup>-1</sup>)

1 eV =  $1.602 \times 10^{-19}$  Ws

To determine the UV light intensity of corona discharge system, the intensity was calculated in a unit of μW. In this work, the following assumption was that the UV light spread out in all direction.

$$\text{Intensity } (\mu\text{W}) = \frac{\text{Intensity measured from UV meter } (\mu\text{W/cm}^2)}{\text{Area of sphere (cm}^2)}$$

Where Area of sphere =  $4\pi r^2$

r = 1.5 cm

## Appendix B Experimental data

**Table B.1** Effect of total feed flow rate at 11,000, 200 Hz, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1

Feed flow rate (ml/min)	Stage (s)	% Conversion		% Selectivity				
		C <sub>2</sub> H <sub>4</sub>	O <sub>2</sub>	CO	CO <sub>2</sub>	CH <sub>4</sub>	C <sub>2</sub> H <sub>2</sub>	C <sub>2</sub> H <sub>6</sub>
40	1	100	57.32	38.33	54.59	u	u	u
	2	100	68.03	20.22	78.65	u	u	u
	3	100	69.53	10.59	90.92	u	u	u
	4	100	71.11	5.86	99.58	u	u	u
80	1	85.37	50.89	52.74	45.83	0.22	0.39	0.39
	2	99.77	62.49	39.18	62.94	0.03	0.1	0.38
	3	99.88	65.82	27.32	76.57	0.05	0.19	0.18
	4	99.90	69.54	16.81	83.04	0.17	u	0.19
160	1	72.26	34.58	53.31	47.68	0.65	0.26	0.78
	2	96.15	49.19	45.59	55.67	u	0.13	0.11
	3	99.39	54.62	36.24	66.97	0.19	0.11	0.34
	4	99.56	55.45	28.15	75.49	u	0.48	0.25
200	1	65.45	30.89	51.45	43.74	0.65	0.45	0.24
	2	90.23	44.91	49.43	51.02	0.05	0.13	0.07
	3	100	53.07	41.10	59.50	u	0.19	0.51
	4	99.81	56.72	34.90	66.99	u	0.05	0.02
240	1	62.10	27.67	48.86	40.18	0.19	u	0.25
	2	90.27	40.89	46.87	42.82	0.38	0.19	0.39
	3	100	48.82	42.98	52.54	0.03	0.28	0.27
	4	99.96	53.00	38.29	61.38	0.09	u	0.18

u = below the GC detecting limit

**Table B.2** Effect of frequency at 11,000, feed flow rate 160 ml/min, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1

Frequency (Hz)	Stage (s)	% Conversion		% Selectivity				
		C <sub>2</sub> H <sub>4</sub>	O <sub>2</sub>	CO	CO <sub>2</sub>	CH <sub>4</sub>	C <sub>2</sub> H <sub>2</sub>	C <sub>2</sub> H <sub>6</sub>
50	1	96.55	55.92	18.12	84.60	0.16	0.13	0.06
	2	99.90	60.85	10.53	92.98	u	0.06	0.20
	3	100	62.25	5.85	98.29	0.09	u	0.59
	4	100	63.07	4.99	99.53	u	u	u
100	1	92.75	47.36	34.21	63.87	u	u	u
	2	100	55.34	26.64	75.94	u	u	u
	3	100	58.58	18.15	86.74	u	0.08	u
	4	100	59.47	14.04	92.12	0.03	0.04	0.10
200	1	72.26	34.58	53.31	47.68	0.65	0.26	0.78
	2	96.15	49.19	45.59	55.67	u	0.13	0.11
	3	99.39	54.62	36.24	66.97	0.19	0.11	0.34
	4	99.56	55.45	28.15	75.49	u	0.48	0.25
300	1	47.23	20.96	71.71	21.54	0.27	u	u
	2	84.63	37.15	63.83	28.22	0.94	0.06	0.05
	3	95.96	47.29	57.08	40.77	0.16	0.12	u
	4	100	51.53	48.38	49.30	u	u	u
400	1	40.11	13.92	69.18	12.78	1.24	0.16	u
	2	69.62	25.91	67.96	16.30	1.62	u	0.13
	3	88.35	36.92	65.21	24.46	1.43	0.02	1.41
	4	95.98	43.59	60.03	32.40	0.98	u	u
500	1	30.51	11.14	73.31	10.86	1.54	0.47	1.01
	2	61.19	21.66	67.86	13.30	2.42	0.23	u
	3	81.38	31.82	68.35	16.89	2.26	0.11	u
	4	91.14	37.71	66.64	22.97	1.63	u	u
700	1	25.15	8.40	59.54	7.59	2.45	0.16	0.15
	2	46.68	15.00	61.77	9.68	2.61	u	u
	3	65.55	20.75	63.36	12.46	2.69	0.09	u
	4	77.74	26.82	64.35	13.73	1.12	0.14	0.12



**Table B.3** Effect of frequency on current and power consumption at 11,000, feed flow rate 160 ml/min, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1

Frequency (Hz)	Stage (s)	Current (A)	Power Consumption (eV/ C <sub>2</sub> H <sub>4</sub> Molecule)
50	1	1.528	730
	2	2.300	1168
	3	3.710	1838
	4	4.610	2318
100	1	1.076	506
	2	1.518	700
	3	2.460	1163
	4	2.910	1399
200	1	0.860	466
	2	1.063	474
	3	1.859	775
	4	1.980	883
300	1	0.776	599
	2	0.915	443
	3	1.645	674
	4	1.798	739
400	1	0.744	642
	2	0.844	464
	3	1.524	665
	4	1.678	679
500	1	0.734	781
	2	0.799	474
	3	1.442	639
	4	1.529	634
700	1	0.686	850
	2	0.771	577
	3	1.405	751
	4	1.501	710

**Table B.4** Effect of voltage at feed flow rate 160 ml/min, 200 Hz, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1

Voltage (V)	Stage (s)	% Conversion		% Selectivity				
		C <sub>2</sub> H <sub>4</sub>	O <sub>2</sub>	CO	CO <sub>2</sub>	CH <sub>4</sub>	C <sub>2</sub> H <sub>2</sub>	C <sub>2</sub> H <sub>6</sub>
9000	1	67.88	33.12	58.02	38.93	u	u	u
	2	92.9	47.07	54.32	43.95	0.36	u	u
	3	99.31	53.90	46.78	55.07	0.08	u	u
	4	99.80	56.56	39.59	64.42	0.14	u	u
11,000	1	72.26	34.58	53.31	47.68	0.65	0.26	0.78
	2	96.15	49.19	45.59	55.67	u	0.13	0.11
	3	99.39	54.62	36.24	66.97	0.19	0.11	0.34
	4	99.56	55.45	28.15	75.49	u	0.48	0.25
13,000	1	71.25	40.73	51.35	52.29	0.29	0.09	u
	2	98.80	56.52	40.24	58.40	0.10	0.52	0.65
	3	99.89	61.17	33.72	71.00	u	0.01	0.08
	4	100	63.10	25.52	79.04	u	0.23	0.08
15,000	1	78.64	41.43	47.05	55.32	u	0.25	0.15
	2	96.03	53.89	40.38	64.89	u	u	0.06
	3	99.81	59.71	29.99	76.06	u	0.09	0.03
	4	100	61.34	23.05	82.70	u	u	0.08

**Table B.5** Effect of voltage on current at feed flow rate 160 ml/min, 200 Hz, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1

Voltage (V)	Stage (s)	Current
9000	1	0.803
	2	0.972
	3	1.741
	4	1.907
11,000	1	0.860
	2	1.063
	3	1.859
	4	1.980
13,000	1	0.907
	2	1.173
	3	1.972
	4	2.180
15,000	1	0.942
	2	1.232
	3	2.040
	4	2.370

**Table B.6** Effect of stage number of reactor with different residence time at 11,000, 200 Hz, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1

Residence time (s)	Stage (s)	% Conversion		% Selectivity				
		C <sub>2</sub> H <sub>4</sub>	O <sub>2</sub>	CO	CO <sub>2</sub>	CH <sub>4</sub>	C <sub>2</sub> H <sub>2</sub>	C <sub>2</sub> H <sub>6</sub>
0.38	1	85.37	50.89	52.74	45.83	u	u	u
	2	96.15	49.19	45.59	55.67	u	u	u
	3	100	48.82	42.98	52.54	u	u	u
0.75	1	100	57.32	38.33	54.59	u	u	u
	2	99.77	62.49	39.18	62.94	u	u	u
	3	99.00	62.00	30.00	72.00	u	u	u
	4	99.56	55.45	28.15	75.49	u	u	u
1.00	1	100	58.56	36.71	63.97	u	u	u
	2	99.86	61.47	32.86	73.77	u	u	u
	3	99.88	59.75	25.66	78.13	u	u	u
	4	99.95	62.55	24.82	80.31	u	u	u

**Table B.7** The UV light intensity measure by UV meter at feed flow rate 160 ml/min, 200 Hz, 9,000 V, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1.

min	Intensity ( $\mu\text{W}/\text{cm}^2$ )	min	Intensity ( $\mu\text{W}/\text{cm}^2$ )	min	Intensity ( $\mu\text{W}/\text{cm}^2$ )	min	Intensity ( $\mu\text{W}/\text{cm}^2$ )
1	0.01470	26	0.01212	51	0.01314	76	0.01300
2	0.01301	27	0.01281	52	0.01252	77	0.01112
3	0.01281	28	0.01180	53	0.01302	78	0.01234
4	0.01062	29	0.01090	54	0.01130	79	0.01080
5	0.01180	30	0.01290	55	0.01252	80	0.01121
6	0.01283	31	0.01194	56	0.01052	81	0.01272
7	0.01180	32	0.01180	57	0.01193	82	0.00829
8	0.01310	33	0.01280	58	0.01210	83	0.01162
9	0.01200	34	0.01260	59	0.01180	84	0.01170
10	0.01050	35	0.01250	60	0.01333	85	0.01220
11	0.01161	36	0.01312	61	0.01240	86	0.01230
12	0.01160	37	0.01360	62	0.01184	87	0.01190
13	0.01150	38	0.01221	63	0.01241	88	0.01240
14	0.01273	39	0.01260	64	0.00969	89	0.01270
15	0.01210	40	0.01190	65	0.01140	90	0.01340
16	0.01060	41	0.01192	66	0.01390	91	0.01174
17	0.01200	42	0.01120	67	0.01173	92	0.01270
18	0.01261	43	0.01224	68	0.01230	93	0.01193
19	0.01252	44	0.01330	69	0.01190	94	0.01160
20	0.01180	45	0.01050	70	0.01324	95	0.01230
21	0.01200	46	0.01220	71	0.01143	96	0.01200
22	0.01213	47	0.01150	72	0.01124	97	0.01340
23	0.01150	48	0.01073	73	0.01230	98	0.01110
24	0.01151	49	0.01261	74	0.01232	99	0.01232
25	0.01081	50	0.01213	75	0.01320	100	0.01171

**Table B.7** The UV light intensity measure by UV meter at feed flow rate 160 ml/min, 200 Hz, 9,000 V, a gap distance of 10 mm, and O<sub>2</sub>: C<sub>2</sub>H<sub>4</sub> ratio of 5:1.

min	Intensity ( $\mu\text{W}/\text{cm}^2$ )	min	Intensity ( $\mu\text{W}/\text{cm}^2$ )	min	Intensity ( $\mu\text{W}/\text{cm}^2$ )
101	0.01251	124	0.01122	147	0.01151
102	0.01230	125	0.01080	148	0.01240
103	0.01320	126	0.01292	149	0.01340
104	0.01160	127	0.01250	150	0.01160
105	0.01182	128	0.01152	151	0.01330
106	0.01203	129	0.01190	152	0.01162
107	0.01130	130	0.01251	153	0.01130
108	0.01070	131	0.01260	154	0.01260
109	0.01120	132	0.01190	155	0.01154
110	0.01120	133	0.01100	156	0.01180
111	0.01100	134	0.01313	157	0.01262
112	0.01191	135	0.01180	158	0.01220
113	0.01220	136	0.01140	159	0.01170
114	0.01080	137	0.01053	160	0.01241
115	0.01144	138	0.01040	161	0.01350
116	0.01120	139	0.01213	162	0.01370
117	0.01290	140	0.01230	163	0.01333
118	0.01293	141	0.01271	164	0.01323
119	0.01143	142	0.01192	165	0.01490
120	0.01221	143	0.01160	166	0.01480
121	0.01280	144	0.01093	167	0.01400
122	0.01110	145	0.01264	168	0.01421
123	0.01170	146	0.01350	169	0.01220

**Table B.8** Comparative results of different selectivity calculation.

Product	% Selectivity					
	Degussa P25		Sol-Gel TiO <sub>2</sub>		1%Pt/ Sol-Gel TiO <sub>2</sub>	
	First	Second	First	Second	First	Second
4 Stages						
CO	41	40	41	41	35	33
CO <sub>2</sub>	60	60	60	59	70	67
3 Stages						
CO	48	49	48	49	46	45
CO <sub>2</sub>	51	51	51	51	56	55
2 Stages						
CO	56	57	55	56	55	52
CO <sub>2</sub>	42	43	43	44	46	44
1 Stage						
CO	58	62	57	55	56	55
CO <sub>2</sub>	36	38	38	37	46	45

## CURRICULUM VITAE

**Name:** Ms. Kanokwan Saktrakool

**Date of Birth:** February 13, 1980

**Nationality:** Thai

**University Education:**

1996-2000 Bachelor Degree of Science in Chemical Engineering, Faculty of Science, Chulalongkorn University, Bangkok, Thailand

**Proceedings and presentations:**

1. **Saktrakool, K.**, Chavadej, S., Rangsunvigit, P., and Lobban, L. L. (2003) “Oxidative Removal of Ethylene in a Multistage Plasma Reactor in the Presence of  $\text{TiO}_2$ ” *Proceedings of First International Symposium on Process Intensification and Miniaturization*, Newcastle upon Tyne, England, 18-21 August, 5 pages.
2. **Saktrakool, K.**, Chavadej, S., Rangsunvigit, P., and Lobban, L. L. (2003) “Ethylene Oxidation in a Multistage Plasma Reactor in The Presence of  $\text{TiO}_2$ ” *The 8<sup>th</sup> International Conference on Atmospheric Sciences and Applications to Air Quality*, Tsukuba, Japan 11 – 13 March.

