

## REFERENCES

- Bhatnagar, R. (1993). A study of partial oxidation of methane under the influence of an AC electric discharge. M.Sc. Thesis, University of Oklahoma.
- Casey, P. S., McAllister, T., and Foger, K. (1994). Selective oxidation of methane to methanol at high pressure. Industrial & Engineering Chemistry Research, 33, 1120-1125.
- Eliasson, B., Hirth, M., and Kogelschatz, U. (1987). Ozone synthesis from oxygen in dielectric barrier discharge. Journal of Physics D: Applied Physics, 20, 1421-1437.
- Foulds, G. A., and Gray, B. F. (1995). Homogeneous gas-phase partial oxidation of methane to methanol and formaldehyde. Fuel Processing Technology, 42, 129-150.
- Gesser, H. D., and Hunter, N. R. (1998). A review of C-1 conversion chemistry. Catalysis Today, 42, 183-189.
- Jingfa, D., Qi, S., Yulong, Z., Songying, C., and Dong, W. (1996). A novel process for preparation of a Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> ultrafine catalyst for methanol synthesis from CO<sub>2</sub> + H<sub>2</sub>: comparison of various preparation methods. Applied Catalysis, 139, 75-85.
- Liu, Q., Rogut, J., Chen, B., Falconer, J. L., and Noble, R. D. (1996). Improved methanol yield from methane oxidation in a non-isothermal reactor. Fuel, 75(15), 1748-1754.
- Lu, G., Shen, S., and Wang, R. (1996). Direct oxidation of methane to methanol at atmospheric pressure in CMR and RSCMR. Catalysis Today, 30, 41-48.

- Mallinson, R. G., Silpcevich, C. M., and Rusek, S. (1987). Methane partial oxidation in alternating electric field. Preprints of the 197th National Meeting of the ACS, Section of Fuel Chemistry, 32(3).
- Marafee, A., Liu, C., Xu, G., Mallinson, R., and Lobban, L. (1997). An experimental study on the oxidative coupling of methane in a direct current corona discharge reactor over Sr/La<sub>2</sub>O<sub>3</sub> catalyst. Industrial & Engineering Chemistry Research, 36(3), 632-637.
- Nasser, E. (1971). Fundamentals of Gaseous Ionization and Plasma Electronics. USA: John Wiley & Sons.
- Okazaki, K., Hirai, S., Nozaki, T., Ogawa, K., and Hijikata K. (1997). Plasma chemical reactions at atmospheric pressure for high efficiency use of hydrocarbon fuels. Energy, 22, 369-374.
- Okumoto, M., Tsunoda, K., Katsura, S., and Mizuno, A. (1997). Direct methanol synthesis using non-thermal pulsed plasma generated by a solid state pulse generator. Journal of Electrostatics, 42, 167-175.
- 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. NATO ASI Series., 34, Part B.
- Satterfield, C. N. (1991). Heterogeneous Catalysis in Industrial Practice. 2nd ed. New York: McGraw-Hill.
- Sokolovskii, V. D., Coville, N. J., Parmaliana, A., Eskendirov, I., and Makoa, M. (1998). Methane partial oxidation. Challenge and perspective. Catalysis Today, 42, 191-195.
- Struis, R. P. W. J., Stucki, S., and Wiedorn M. (1996). A membrane reactor for methanol synthesis. Journal of Membrane Science, 113, 96-100.

- Taylor, S. H., Hargreaves, J. S. J., Hutchings, G. J., Joyner, R. W., and Lembacher, C. W. (1998). The partial oxidation of methane to methanol: An approach to catalyst design. Catalysis Today, 42, 217-224.
- Thanyachotpaiboon, K., Chavadej, S., Caldwell, T. A., Lobban, L. L., and Mallinson, R. G. (1998). Conversion of methane to higher hydrocarbons in AC nonequilibrium plasmas. American Institute of Chemical Engineering Journal, 44, 2252-2257.
- Twigg, M. V. (1989). Catalyst Handbook. England: Butler & Tanner.
- Walker, G. S., Lapszewicz, J. A., and Foulds G. A. (1994). Partial oxidation of methane to methanol comparison of heterogeneous catalyst and homogeneous gas phase reactions. Catalysis Today, 21, 519-526.
- Walsh, D. E., Martenak, D. J., Han, S., and Palermo, R. E. (1992). Direct oxidative methane conversion at elevated pressure and moderate temperatures. Industrial & Engineering Chemistry Research, 31, 1259-1262.

## **APPENDICES**

**APPENDIX A**  
**EXPERIMENTAL DATA**

**Table A.1** Effect of oxygen partial pressure at total flow rate = 100 ml/min and applied voltage = 5,000 V

CH <sub>4</sub> :O <sub>2</sub>	%CH <sub>4</sub> conv.	%O <sub>2</sub> conv.	% Selectivity							
			CO	H <sub>2</sub>	HCOH	CH <sub>3</sub> OH	C <sub>2</sub> <sup>•</sup>	C <sub>2</sub> <sup>=</sup>	C <sub>2</sub> <sup>≡</sup>	CO <sub>2</sub>
2:1	19.8	68.5	4.0	0	4.6	0	0	7.4	0	85.9
4:1	15.6	94.5	16.0	10.8	7.8	0	0	19.0	0	68.0
8:1	8.9	88.2	19.3	33.6	13.5	0.02	0	31.9	0	42.0

**Table A.2** Effect of applied voltage at total flow rate = 100 ml/min and CH<sub>4</sub>:O<sub>2</sub> = 4:1

Applied Voltage (V)	%CH <sub>4</sub> conv.	%O <sub>2</sub> conv.	% Selectivity							
			CO	H <sub>2</sub>	HCOH	CH <sub>3</sub> OH	C <sub>2</sub> <sup>•</sup>	C <sub>2</sub> <sup>=</sup>	C <sub>2</sub> <sup>≡</sup>	CO <sub>2</sub>
5000	15.6	94.5	16.0	10.8	7.8	0	0	19.0	0	68.0
6000	22.6	97.2	13.0	22.4	6.9	0.02	13.4	20.1	0	47.9
8000	22.7	93.9	12.4	22.5	6.6	0.01	13.7	22.1	0	43.5
10000	23.4	84.8	11.1	29.8	5.5	0.02	10.0	19.7	0	40.8

**Table A.3** Effect of total flow rate at  $\text{CH}_4:\text{O}_2 = 4:1$  and applied voltage = 5,000 V

Total Flow Rate (ml/min)	%CH <sub>4</sub> conv.	%O <sub>2</sub> conv.	% Selectivity							
			CO	H <sub>2</sub>	HCOH	CH <sub>3</sub> OH	C <sub>2</sub>	C <sub>2</sub> <sup>=</sup>	C <sub>2</sub> <sup>≡</sup>	CO <sub>2</sub>
50	16.6	91.4	17.0	33.9	10.9	0.01	7.2	26.3	0	52.5
100	15.6	94.5	16.0	10.8	7.8	0	0	19.0	0	68.0
150	12.1	69.6	13.0	6.8	7.4	0	0	18.0	0	60.2
200	11.5	76.2	5.2	0	5.6	0	0	10.3	0	73.8

**Table A.4** Effect of oxygen partial pressure on non-catalytic system at total flow rate = 100 ml/min and applied voltage = 5,000 V

CH <sub>4</sub> :O <sub>2</sub>	%CH <sub>4</sub> conv.	%O <sub>2</sub> conv.	% Selectivity							
			CO	H <sub>2</sub>	HCOH	CH <sub>3</sub> OH	C <sub>2</sub>	C <sub>2</sub> <sup>=</sup>	C <sub>2</sub> <sup>≡</sup>	CO <sub>2</sub>
2:1	48.6	46.9	34.5	52.6	2.2	0	12.0	37.2	9.1	9.9
4:1	35.4	56.6	26.6	59.0	4.6	0.01	18.7	30.8	10.4	9.9
8:1	27.3	53.0	11.2	55.1	5.9	0.01	22.5	19.3	12.2	12.4

## APPENDIX B

### ASSUMPTIONS, DEFINITIONS AND CALCULATIONS

To facilitate the calculations, some valid assumptions were made as follows:

1. All the gaseous behaviors obey the ideal gas law.
2. Pressure drop across the system is very small and can be negligible.
3. The pressure in the system equals atmospheric pressure (1 atm).
4. The flow rate change across the reactor due to the variation in the gaseous compositions during the reaction time is very small and can be negligible.

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 atm·ml·mol<sup>-1</sup>·min<sup>-1</sup>·K<sup>-1</sup>)

$T$  = absolute ambient temperature (K)

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

The conversion is defined as:

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

The selectivity of each product is defined on the basis of the amount of carbon converted from the reactant into the specified products. The product selectivity is defined as follows:

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

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 selectivity of Hydrogen is defined as follows:

$$\% H_2 \text{ Selectivity} = \frac{\text{Mole of } H_2 \text{ produced}}{2 \times \text{Mole of } CH_4 \text{ converted}} \times 100 \quad (\text{B.4})$$



## CURRICULUM VITAE

**Name:** Sutha Sutthiruangwong

**Date of Birth:** 29 August 1975

**Nationality:** Thai

**University Education:**

1993-1997 Bachelor's Degree of Engineering in Chemical  
Engineering, Burapha University