REFERENCES

Bhatnagar, R. (1993) A Study of Partial Oxidation of Methane under the Influence of an AC Electric discharge. M.S. Thesis, University of Oklahoma.

Casey, P.S., McAllister, T., and Foger, K. (1994) Selective oxidation of methane to methanol at high pressures. Industrial Engineering Chemistry Research, 33, 1120-1125.

Eliasson, B., Hirth, M., and Kogeischatz, U. (1987) Ozone synthesis from oxygen in dielectric barrier discharge. Journal of Applied Physics, 20, 1421-1437.

Eliasson, B. and Kogeischatz, U. (1991) Nonequilibrium volume plasma chemical processing. IEEE Transactions on Plasma Science, 19(6), 1063-1077.

Fraser, M., Fee, D., and Sheinson, R. (1985) Decomposition of methane in an AC discharge. Plasma Chemistry and Plasma Processing, 5, 163-173.

Fridman, A., Naster, S., Kennedy, L.A., Savaliev, A., and Mutaf-Yardimci O. (1999) Gliding arc discharge. Journal of Progress in Energy and Combustion Science, 25, 211-213.

Gesser, H.D., Hunter, N.R. and Prakash, C.B. (1985) The direct conversion of methane to methanol by controlled oxidation. Industrial Engineering Chemistry Research, 85(4), 235-244.

Hill, B.J., (1997) The oxidative coupling of methane using an AC electric gas discharge. M.S., Thesis, The university of Oklahoma.

Huang, J., Badani, M.V., Suib, S.L., Harrison, J.B., and Kablauoi, M. (1994) Partial oxidation of methane to methanol through microwave plasmas. Journal of Physical Chemistry, 98, 206-210.

Larkin W. David, Lobban L. Lance and Millinson G. Richard (2001) Production of organic oxygenates in the partial oxidation of methane in a silent electric discharge reactor. Industrial Engineering Chemistry Research, 40, 1594-1601.

Leethochawalit, K. (1998) Effect of carbon dioxide on methane conversion under electric discharge. M.S. Thesis, Chulalongkorn University, Bangkok.

Lui, C., Marafee, A., Hill, B., Xu, G., Mallinson, R., and Lobban, L. (1996) Oxidative coupling of methane with AC and DC corona discharges. Industrial & Engineering Chemistry Research, 35, 3295-3301.

Lui, C., Marafee, A., Hill, B., Xu, G., Mallinson, R., and Lobban, L. (1997) The oxidative coupling of methane in DC corona discharges reactor over Sr/La_2O_3 catalyst. Industrial & Engineering Chemistry Research, 36, 632-637.

Mallinson, R.G., and Sliepecevich, C.M. (1986) Field effect catalysis program final report, University Technologist, Inc.

Mizuno, A., Chakrabari, A., and Okazaki, K. (1992) Application of corona technology in the production of greenhouse gases and other gaseous pollutants. Non-thermal Plasma Technique for Pollution Control., NATO ASI series, 34, part B, 167-185.

Nasser, E. (1971) Fundamentals of gaseous ionization and plasma electronics, USA: John Wiley & Sons, Inc.

Okumoto, M., Rajanikanth, B.S., Katsura, S., and Mizuno, A. (1998) Nonthermal plasma approach in direct methanol synthesis from methane. IEEE Transactions on Industry Applications, 34, 940-944.

Oumghar, A., Legrand, J.C., Diamy, A.M., and Turillon, N. (1995) Methane conversion by an air microwave plasma. Plasma Chemistry and Plasma Procassing, 15, 87-107.

Perry R.H., Green D.W., and Maloney J.O., (1997) Perry's Chemical Engineering' Handbook. 7th, USA: McGraw-Hill, Inc

Poonohatanapricha, P. (1998) Methane conversion in an AC electric discharge. M.S. Thesis, Chulalongkorn University, Bangkok.

Rusu, I., and Cormier, J.M. (2003) On a possible mechanism of the methane steam reforming in a gliding arc reactor. Chemical Engineering Journal, 15(1), 23-31.

Supat, K., Kruapong, A., and Chavadej, S., Lobban L.L, and Millinson G.R. (2003) Synthesis gas production from partial oxidation of methane with air in AC electric gas discharge. Energy & Fuels, 17 , 474-481.

Supat, K., Chavadej, S., Lobban L.L, and Millinson G.R. (2002) Combined steam reforming and partial oxidation of methane to synthesis gas under electrical discharge. Industrial & Engineering Chemistry Research, 42, 1654-1661.

Sutthiruangwong, S. (1999) Plasma catalytic production of methanol. M.S. 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.

Wang, S., and Lu, G.Q. (1996). Carbon dioxide reforming of methane to produce synthesis gas over metal-supported catalysts: State of art. Energy & Fuels, 10, 896-904.

Zerger, R.P., Sui, S.L., and Zhang, Z. (1993) Preparation of oxygenates with methane/oxygen microwave plasmas. American Chemical Society, 37, 344 -348.

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:

$$N = q \times (P/RT) \qquad \text{(B.1)}$$

where

q = total volumetric flow rate

P = total pressure of the system

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

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:

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

The first selectivity is defined as:

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

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 second selectivity is defined as:

$$\% \text{ Selectivity of } H_2 = \frac{\text{Molar flowrate of } H_2 \text{ produced}}{\text{Rate of H reacted}} \times 100 \quad (B.4)$$

To determine the energy efficiency of gliding arc discharge system, the specific energy consumption was calculated in a unit of electron-volt per molecule of converted carbon (eV/m_c) from the following equation:

$$\text{Specific energy consumption} = \frac{P \times 60}{(1.602 \times 10^{-19}) \times \tilde{N} \times M_C}$$

eV/ molecule of CH_4 converted (B.5)

Where

P = Power (W)

$\tilde{N}$ = Avogadro's number = 6.02×10^{23} molecules.g-mole^{-1}

M_C = Rate of methane in feed gas converted (g-mole.min^{-1})

1 eV = 1.602×10^{-19} W

Appendix B Experimental data.

Table B.1 Effect of CH_4/O_2 molar ratio at 200 cm^3/min, 300 Hz, 15,500 V, and gap distance of 6 mm

Molar ratio	Stage (s)	%Conversion		%Selectivity					
		CH_4	O_2	H_2	CO	CO_2	C_2H_2	C_2H_4	C_2H_6
2/1	1	18.28	29.79	34.67	77.30	3.47	14.61	9.22	6.73
	2	35.09	46.70	28.94	58.10	4.14	15.82	9.79	4.44
	3	52.04	55.30	21.64	51.93	5.71	18.49	7.72	3.05
	4	68.02	69.99	17.93	44.61	6.00	19.51	5.65	1.90
3/1	1	10.64	22.58	35.46	61.63	3.29	14.51	21.51	13.24
	2	22.91	38.22	31.88	52.65	3.80	16.26	16.39	8.58
	3	37.28	53.69	27.13	44.46	5.03	18.92	12.41	5.65
	4	52.14	69.92	24.74	39.37	5.46	20.58	9.41	3.56
4/1	1	4.16	15.27	45.05	58.69	1.96	15.60	22.47	32.19
	2	13.06	29.10	33.61	47.68	2.27	17.22	20.36	18.20
	3	25.14	44.10	28.38	38.57	2.66	19.05	19.31	10.93
	4	38.27	59.83	25.25	32.49	3.24	20.92	14.60	6.94
5/1	1	2.30	25.66	50.24	50.57	1.55	16.92	38.75	46.27
	2	6.69	32.36	36.16	40.23	1.75	17.83	31.05	32.13
	3	16.64	40.55	23.33	30.99	2.17	20.00	22.32	17.47
	4	28.81	51.50	20.86	26.06	2.85	21.70	16.80	10.54

Table B.2 Effect of CH_4/O_2 mole ratio on power consumption at 200 cm^3/min, 300 Hz, 15,500 V, and a gap distance of 6 mm

Molar ratio	Stage (s)	Power Consumption (eV/ CH_4 Molecule converted)
2/1	1	16.18
	2	15.17
	3	14.20
	4	13.93
3/1	1	20.04
	2	19.37
	3	14.56
	4	14.33
4/1	1	32.88
	2	23.12
	3	19.11
	4	15.02
5/1	1	41.26
	2	39.24
	3	22.76
	4	18.12

Table B.3 Effect of feed flowrate at CH_4/O_2 molar ratio of 3/1, 300 Hz, 15,500 V, and a gap distance of 6 mm

Flowrate (cm^3/min)	Stage (s)	%Conversion		%Selectivity					
		CH_4	O_2	H_2	CO	CO_2	C_2H_2	C_2H_4	C_2H_6
50	1	24.16	37.09	30.56	39.24	2.88	16.62	18.11	12.85
	2	28.65	50.16	30.52	39.01	2.98	20.21	16.92	12.44
	3	49.45	73.75	28.79	36.86	3.07	21.30	16.27	12.25
	4	69.89	84.07	28.34	33.70	3.19	22.41	15.12	11.95
100	1	15.99	32.18	28.96	41.24	2.85	16.30	16.89	12.29
	2	28.33	49.12	28.86	39.51	3.00	17.05	16.20	11.94
	3	47.83	71.94	27.78	36.91	3.06	21.13	15.83	11.64
	4	65.69	83.42	27.20	36.49	3.12	21.84	15.37	11.23
150	1	16.29	26.79	28.73	46.27	2.83	15.82	16.30	11.25
	2	28.31	46.29	28.38	41.60	2.97	17.00	15.79	11.23
	3	43.07	59.61	27.56	39.92	3.01	17.64	15.50	11.22
	4	63.51	78.77	26.40	36.62	3.07	17.89	15.18	11.13
200	1	13.13	24.74	27.49	48.56	2.81	14.28	15.27	11.38
	2	26.88	41.41	25.60	42.56	2.86	15.31	15.00	11.30
	3	39.68	55.47	24.78	40.17	2.90	15.86	14.90	11.20
	4	52.88	69.91	23.94	37.06	2.95	16.59	14.15	11.12
250	1	7.49	8.04	26.90	49.38	2.76	13.48	15.08	11.30
	2	17.01	27.12	24.33	42.66	2.78	14.52	14.95	11.18
	3	31.42	43.47	22.59	41.03	2.82	14.66	14.85	11.16
	4	45.48	49.21	19.78	39.61	2.89	16.04	14.52	11.18
300	1	6.22	8.12	22.48	49.67	2.67	11.30	14.04	11.23
	2	16.56	18.37	22.24	49.04	2.72	11.65	13.51	11.15
	3	27.24	30.76	19.84	42.63	2.75	13.17	12.00	11.08
	4	35.98	43.54	17.36	40.62	2.84	13.36	8.80	10.45

Table B.4 Effect of feed flowrate on power consumption at CH_4/O_2 molar ratio of 3/1, 300 Hz, 15,500 V, and a gap distance of 6 mm

Flowrate (cm^3/min)	Stage (s)	Power Consumption (eV/ CH_4 Molecule converted)
50	1	39.46
	2	45.47
	3	47.26
	4	49.44
100	1	23.75
	2	25.32
	3	28.23
	4	28.90
150	1	16.68
	2	17.28
	3	18.30
	4	19.26
200	1	15.86
	2	17.04
	3	17.31
	4	18.10
250	1	13.58
	2	15.73
	3	21.79
	4	28.87
300	1	13.59
	2	14.26
	3	18.16
	4	34.27

Table B.5 Effect of stage number of reactor with different residence time at CH_4 molar ratio of 3/1, 300 Hz, 14,500 V and a gap distance of 6 mm

Residence time (sec)	Stage (s)	%Conversion		%Selectivity					
		CH_4	O_2	H_2	CO	CO_2	C_2H_2	C_2H_4	C_2H_6
0.5517	1	6.22	8.12	22.48	49.67	2.67	11.30	14.04	11.23
	2	16.56	18.37	22.24	49.04	2.72	11.65	13.51	11.15
	3	27.24	30.76	19.84	42.63	2.75	13.17	12.00	11.08
	4	35.98	43.54	17.36	40.62	2.84	13.36	8.80	10.45
0.8276	1	13.13	24.74	27.49	48.56	2.81	14.28	15.27	11.38
	2	26.88	41.41	25.60	42.56	2.86	15.31	15.00	11.30
	3	39.68	55.47	24.78	40.17	2.90	15.86	14.90	11.20
	4	52.88	69.91	23.94	37.06	2.95	16.59	14.15	11.12
1.6652	1	15.99	32.18	28.96	41.24	2.85	16.30	16.89	12.29
	2	28.33	49.12	28.86	39.51	3.00	17.05	16.20	11.94
	3	47.83	71.94	27.78	36.91	3.06	21.13	15.83	11.64
	4	65.69	83.42	27.20	36.49	3.12	21.84	15.37	11.23
3.3105	1	24.16	37.09	30.56	39.24	2.88	16.62	18.11	12.85
	2	28.65	50.16	30.52	39.01	2.98	20.21	16.92	12.44
	3	49.45	73.75	28.79	36.86	3.07	21.30	16.27	12.25
	4	69.89	84.07	28.34	33.70	3.19	22.41	15.12	11.95

Table B.6 Effect of frequency at CH_4/O_2 molar ratio of 3/1, feed flowrate of 150 cm^3/min, 15,500 V, and a gap distance of 6 mm

Frequency (Hz)	Stage (s)	%Conversion		%Selectivity					
		CH_4	O_2	H_2	CO	CO_2	C_2H_2	C_2H_4	C_2H_6
300	1	16.31	26.81	28.70	46.22	2.89	15.72	16.20	11.48
	2	28.33	46.30	28.36	41.56	3.19	16.99	15.77	11.22
	3	43.09	59.62	27.54	39.88	3.23	17.62	15.48	11.21
	4	63.65	78.85	26.24	37.59	3.33	17.87	15.17	11.12
400	1	7.55	13.64	28.56	46.04	2.63	21.43	13.90	13.90
	2	19.32	28.59	28.34	41.41	2.62	20.47	13.77	13.77
	3	33.61	43.71	26.67	39.71	2.91	20.55	13.40	13.40
	4	45.51	60.91	26.22	37.62	3.20	20.87	13.16	13.16
500	1	6.65	12.34	28.51	45.76	2.47	21.31	18.02	14.09
	2	15.93	24.89	28.28	41.26	2.54	21.48	17.24	13.50
	3	27.84	36.97	26.56	38.95	2.96	24.59	17.14	13.45
	4	17.02	42.25	25.92	37.49	3.20	24.70	17.02	13.02
600	1	6.31	11.06	28.36	45.61	2.40	23.35	18.29	15.04
	2	14.55	20.83	28.17	41.26	2.50	24.12	18.50	14.46
	3	26.42	32.92	26.52	38.69	3.03	25.84	17.98	14.14
	4	31.23	39.06	25.90	37.37	3.22	26.20	17.88	13.95
700	1	5.86	10.37	28.05	45.55	1.90	20.91	15.37	14.49
	2	13.72	19.47	27.78	40.96	2.03	22.12	15.22	14.46
	3	22.02	28.08	26.32	37.96	2.45	24.05	15.64	14.38
	4	28.94	36.22	25.68	36.97	2.89	28.75	16.69	14.19

Table B.7 Effect of frequency on current and power consumption at CH_4/O_2 molar ratio of 3/1, feed flowrate of 150 cm^3/min, 15,500 V, and a gap distance of 6 mm

Frequency (Hz)	Stage (s)	Current (A)	Power Consumption (eV/ CH_4 Molecule converted)
300	1	0.83	16.67
	2	0.51	17.27
	3	0.39	18.29
	4	0.23	19.25
400	1	0.77	20.74
	2	0.49	21.62
	3	0.36	22.53
	4	0.2	23.52
500	1	0.68	27.96
	2	0.45	30.00
	3	0.32	31.48
	4	0.19	34.16
600	1	0.6	29.15
	2	0.42	32.52
	3	0.3	34.83
	4	0.18	35.35
700	1	0.54	40.04
	2	0.4	41.78
	3	0.28	42.59
	4	0.18	43.22

Table B.8 Effect of voltage at CH_4/O_2 molar ratio of 3/1, feed flowrate of 150 cm^3/min, 300 Hz and gap distance of 6 mm

Voltage (V)	Stage (s)	%Conversion		%Selectivity					
		CH_4	O_2	H_2	CO	CO_2	C_2H_2	C_2H_4	C_2H_6
10000	1	5.06	12.59	42.42	75.19	2.46	13.55	20.71	21.19
	2	8.31	16.01	38.58	59.82	2.70	14.04	20.51	20.99
	3	17.68	25.69	34.90	49.98	2.81	14.24	19.94	20.29
	4	20.69	27.22	33.71	47.27	3.03	14.46	19.42	19.86
12500	1	7.67	13.31	36.62	57.87	2.74	13.60	20.12	19.15
	2	12.02	21.25	34.05	51.97	2.87	14.12	19.58	18.90
	3	21.10	29.46	32.36	45.85	2.94	14.40	19.13	18.68
	4	33.19	42.49	30.63	38.04	3.13	15.77	18.86	18.07
14500	1	12.28	14.73	31.83	46.48	2.82	14.86	18.97	14.41
	2	16.83	27.27	31.24	45.98	3.05	15.36	18.63	13.85
	3	34.43	48.04	29.10	42.44	3.15	16.00	18.23	13.44
	4	45.12	62.17	28.25	37.33	3.28	16.43	18.11	12.17
15500	1	16.29	26.79	28.73	46.27	2.94	16.39	16.78	11.25
	2	28.32	46.30	28.37	41.57	3.13	16.92	16.25	11.21
	3	43.10	59.63	27.53	39.87	3.22	17.11	15.77	11.11
	4	63.60	78.82	26.30	36.48	3.38	17.91	15.01	10.90
17500	1	28.66	35.60	28.33	45.75	3.01	17.20	13.82	8.89
	2	41.13	53.08	27.75	37.37	3.20	17.98	13.61	8.42
	3	54.51	67.91	26.43	33.00	3.25	18.97	13.44	8.21
	4	65.22	80.53	25.57	29.30	3.42	19.55	12.97	7.43
19000	1	29.77	40.21	27.49	35.84	3.13	17.97	11.83	5.70
	2	48.91	62.22	26.92	32.18	3.23	18.29	11.56	5.45
	3	55.59	69.67	26.45	32.56	3.30	19.38	11.47	5.24
	4	71.00	84.24	24.68	25.71	3.53	20.82	10.21	5.20

Table B.9 Effect of voltage on current and power consumption at CH_4/O_2 molar ratio of 3/1, feed flowrate of 150 cm^3/min, 300 Hz and a gap distance of 6 mm

Voltage (V)	Stage (s)	Current (A)	Power Consumption (eV/ CH_4 Molecule converted)
10000	1	0.38	13.30
	2	0.34	14.57
	3	0.29	15.23
	4	0.15	15.62
12500	1	0.53	14.05
	2	0.48	15.69
	3	0.33	16.59
	4	0.17	17.85
14500	1	0.67	15.32
	2	0.51	16.76
	3	0.35	17.95
	4	0.18	18.46
15500	1	0.72	16.68
	2	0.53	17.27
	3	0.36	18.92
	4	0.19	19.23
17500	1	0.81	19.95
	2	0.65	21.48
	3	0.39	23.84
	4	0.2	25.90
19000	1	0.85	22.52
	2	0.67	25.30
	3	0.46	26.44
	4	0.22	27.96

Table B.10 Effect of gap distance at CH_4/O_2 molar ratio of 3/1, feed flowrate of 150 cm^3/min, 300 Hz and 14,500 V

Gap distance (mm)	Stage (s)	%Conversion		%Selectivity					
		CH_4	O_2	H_2	CO	CO_2	C_2H_2	C_2H_4	C_2H_6
2	1	9.31	13.08	33.93	43.75	1.56	8.07	16.29	12.48
	2	13.74	21.73	33.79	43.30	2.15	12.31	14.52	10.36
	3	18.95	27.17	33.04	42.46	2.37	14.78	13.71	9.97
	4	24.47	30.37	31.81	41.89	2.56	15.06	12.91	9.23
4	1	13.27	13.24	28.75	41.34	1.76	12.25	12.10	8.25
	2	15.96	23.39	28.31	41.23	2.31	12.83	12.02	7.92
	3	22.24	32.49	27.60	40.89	2.60	14.80	11.18	7.83
	4	32.33	39.40	27.38	39.01	2.75	15.89	10.82	7.75
6	1	15.89	22.74	28.18	40.49	2.39	13.03	12.02	8.12
	2	18.08	29.85	27.86	39.82	2.91	13.38	11.68	7.87
	3	30.53	44.97	27.51	39.58	3.26	14.92	11.13	7.81
	4	38.90	57.88	27.27	38.89	3.38	15.94	10.75	7.38
8	1	20.21	33.46	26.62	38.52	2.45	15.38	11.88	8.09
	2	20.59	35.10	26.49	37.64	3.04	15.87	11.62	7.82
	3	34.57	53.34	24.77	36.81	3.30	16.64	11.08	7.53
	4	45.71	69.66	22.03	32.36	3.41	16.91	10.54	7.30

Table B.11 Effect of gap distance on power consumption at CH_4/O_2 molar ratio of 3/1, feed flowrate of 150 cm^3/min, 300 Hz and 14,500 V

Gap distance (mm)	Stage (s)	Power Consumption (eV/ CH_4 Molecule converted)
2	1	14.30
	2	15.51
	3	16.87
	4	17.42
4	1	14.84
	2	17.62
	3	17.73
	4	18.30
6	1	16.91
	2	17.84
	3	18.48
	4	19.34
8	1	17.63
	2	20.76
	3	21.26
	4	22.75

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