

## REFERENCES

- Anderson, R. B., Stein, K. C., Feenan, J. J., and Hofer, L. E. J. (1961) Catalytic combustion of methane. *Industrial & Engineering Chemistry*, 53, 809.
- Ari, H. and Machida, M. (1991) Recent progress in high temperature catalytic combustion. *Catalysis Today*, 10, 81-94.
- Baldwin, T. R. and Burch, R. (1990) Catalytic combustion of methane over supported palladium catalyst. *Applied Catalysis*, 66, 235-246.
- Beebe, K.W., Cairns, K.D., Pareek, V.K., and Nickolas, S.G. (2000) Development of catalytic combustion technology for single-digit emissions from industrial gas turbines. *Catalysis Today*, 59, 95-115
- Betta, R. A. D. (1997) Catalytic combustion gas turbine systems: the preferred technology for low emission electric power production and co-generation. *Catalysis Today*, 35, 129-135.
- Briot, P. and Primet, M. (1991) Catalytic combustion of methane over palladium supported on alumina-effect of aging under reactants. *Applied Catalysis*, 69, 301-314.
- Carroni, R., Schmidt, V., and Griffin., T. (2002) Catalytic combustion for power generation, *Catalysis Today*, 75, 287-295
- Carstens, J. N., Su, S. C., and Bell, A. T. (1998) Factors affecting the catalytic activity of Pd/ZrO<sub>2</sub>for the combustion of methane, *Journal of Catalysis*, 176, 136-142.
- Chin, Y. H. and Resasco, D. (1999) *Catalysis-Specialists Periodical Reports*. In Spivey, J. J. (Eds.), Royal Society of Chemistry, New York: Cambridge.
- Climino, S., Pirone, R., and Russo, G. (2001) Thermal stability of perovskite-based monolithic reactors in the catalytic combustion of methane. *Industrial Chemical Research*, 40(1), 80-58.
- Dalla Betta, R.A. and Rostrup-Nielsen, T. (1999) Application of catalytic combustion to a 1.5 MW industrial gas turbine. *Catalysis Today*, 47, 369-375.

- Datye, A. K., Bravoa, J., Nelson, T. R., Atanasova, P., Lyubovskyb, M., and Pfefferle, L. (2000) Catalyst microstructure and methane oxidation reactivity during the  $\text{Pd} \leftrightarrow \text{PdO}$  transformation on alumina supports. *Applied Catalysis A: General*, 198, 179-196.
- Eguchi, K., and Arai, H. (1996) Recent advances in high temperature catalytic combustion. *Catalysis Today*, 29, 379-386.
- Ersson, A. (2003) Material for High-Temperature Catalytic Combustion. Ph.D. Thesis in chemical engineering, Department of Chemical Engineering and Technology, Kingliga Tekniska Högskolan (Royal Institute of Technology).
- Etemad, S., Karim, H., Smith, L. L., and Pfefferle. (1999) Advance technology catalytic combustior for high temperature ground power gas turbine applications. *Catalysis Today*, 47, 305-313.
- Ferraudo, R. J., Hobson, M. C., Kennelly, T., and Waterman, E. M. (1992) Catalytic chemistry of supported palladium for combustion of methane. *Applied Catalysis A: General*, 81, 227-237.
- Ferraudo, R. J., Lampert, J. K., Hobson, M. C., and Waterman, E. M. (1995) Thermal decomposition and reformation of  $\text{PdO}$  catalysts; support effect. *Applied Catalysis B: Environmental*, 6, 263-270.
- Forzatti, P. (2000) Environmental catalysis for stationary applications. *Catalysis Today*, 62, 50-65
- Gélin, P. and Primet, M. (2002) Complete oxidation of methane at low temperature over nobal metal based catalyst: a review. *Applied Catalysis B: Environmental*, 39, 1-37.
- Geus, J. W. and Giez-en, J. C. (1999) Monoliths in catalytic oxidation. *Catalysis Today*, 47, 169-180.
- Griffin, T., Weisenstein, W., and Fowles, M. (1995) Palladium-catalyzed combustion of methane: simulated gas turbine combustion at atmospheric pressure. *Combustion and Flame*, 101, 81-90.
- Hicks, R. F., Qi, H., Young, M. L., and Lee, R. G. (1990) Structure sensitivty of methane oxidation over platinum and palladium. *Journal of Catalysis*, 122, 295-306.

- Heck, R. M., Gulati, S., and Farrauto, R. J. (2001) The application of monoliths for gasphase catalytic reaction. Chemical Engineering Journal, 82, 149-156.
- Holzwarth, A., Denton, P., Zanthonoff, H., and Mirodatos, C. (2001) Combinatorial approaches to heterogeneous catalysts strategies and perspective for academic research. Catalysis Today, 67, 309-318.
- Hoyos, L. J., Praliaud, H., and Primet, M. (1993) Catalytic combustion of methane over palladium supported on alumina and silica in presence of hydrogen sulfide. Applied Catalysis A: General, 98, 125-138.
- IEA Greenhouse Gas R&D Programme June (1998) Abatement of methane emission.
- Kapteijn, F. and Moulijn, J. A. (1996) Handbook of Heterogeneous Catalysis Vol. 3, In Ertl, G., Knözinger, H. and Weitkamp (Eds.), VCH: Weinheim.
- Kennelly, J., Mead, B., and Freeauto, R. J. (1993) Catalytic combustion process using supported palladium oxide catalysts. US Patent 5,216,875.
- Kennelly, J., Mead, B., Chou, T. C., Jose, S., and Freeauto, R. J. (1999) Combustion catalyst containing oxides and processes using the same. US Patent 5,863,851.
- Kikuchi, R. Maeda, S., Sasaki, K., Wennerström, S., Ozawa, Y. and Eguchi, K. (2003) Catalytic activity of oxide-supported Pd catalysts on a honeycomb for low-temperature methane oxidation. Applied Catalysis A: General, 239, 169-179.
- Ledwich, J. and Su, S. (2001) Catalytic combustion of coal mine ventilation air: literature review and experimental preparation. CSIRO Exploration and Mining, May 2001, Australia.
- Lee, J. H. and Trimm, D. L. (1995) Catalytic combustion of methane. Fuel Processing Technology, 42, 339-359.
- Lefebvre, A.H. (1983) Gas Turbine Combustion, Hemisphere Publishing Corp., Bristol.
- Lopez, T., Asomoza, M., Bosch, P., Garcia-Fiueroa, E., and Gomez, R. (1992) Journal of Catalysis, 138, 463.
- Lyubovsky, M., LaPierre, R., Smith, L., Castaldi, M., Nentwick, B., and Pefferle, W.C. (2002) Catalytic combustion of methane under fuel lean and fuel rich

- conditions over supported Pt, Pd and Rh catalysts. Book of abstracts - 5<sup>th</sup> International symposium on catalytic combustion, Seoul.
- McFarland, E.W. and Weinberg, W.H. (1999) Combinatorial approaches to materials discovery. Tibtech, 17, 107-115.
- Méthivier, Ch., Beguin, B., Brun, M., Massardier, J., and Bertolini, J. C. (1998) Pd/SiC catalysts: characterization and catalytic activity for the methane total oxidation. Journal of Catalysis, 173, 374-382.
- Méthivier, Ch., Massardier, J., and Bertolini, J. C. (1999) Pd/Si<sub>3</sub>N<sub>4</sub> catalysts: preparation, characterization and catalytic activity for the methane total oxidation. Applied Catalysis A: General, 182, 337-344.
- Muller, M., Maciejewski, R. A., Koeppel, R., Tschan, A., and Baiker (1996) Role of lattice oxygen in the combustion of methane over PdO/ZrO<sub>2</sub>: combined pulse TG/DTA and MS study with <sup>18</sup>O-labelled catalyst. Journal of Physical Chemistry, 100, 20006-20014.
- Müller, C. A., Maciejewski, M., Koeppel, R. A., and Baiker, A. (1997) Combustion of methane over palladium/zirconia derived from a glassy Pd-Zr alloy: effect of Pd particle size on catalytic behavior. Journal of Catalysis, 166, 36-43.
- Narui, K., Yata, H., Furuta, K., Nishida, A., Kohtoku, Y. and Matsuzaki, T. (1999) Effects of adding of Pt to PdO/Al<sub>2</sub>O<sub>3</sub> catalyst on catalytic activity for methane combustion and TEM observations of supported particles. Applied Catalysis A: General, 179, 165-173.
- Neomangus, H. W. J. P., Saracco, G., Wessel, H. F. W., and Versteeg, G. F. (2000) The catalytic combustion of natural gas in a membrane reactor with separate feed of reactants. Chemical Engineering Journal, 77, 165-177.
- Oh, S. H., Mitchell, P. J., and Siewert, R. M. (1991) Catalytic control of air pollution: mobile and stationary sources. In Silver, J. E. (Eds.), 202<sup>nd</sup> National Meeting of the American Chemical Society, 25-30 August 1991, ACS Series, Vol. 495.
- Ozawa, Y., Fujii, T., Sato, M., Kanazawa, T., and Inoue, H. (1999) Development of a catalytically assisted combustor for a gas turbine. Catalysis Today, 47, 399-405.

- Ozawa, Y., Tochihara, Y., Nagai, M., and Omi, S. (2003) Effect of addition of Nd<sub>2</sub>O<sub>3</sub> and La<sub>2</sub>O<sub>3</sub> to PdO/Al<sub>2</sub>O<sub>3</sub> in catalytic combustion of methane. Catalysis Communications, 4, 87-90.
- Ozawa, Y., Tochihara, Y., Watanabe, A., Nagai, M., and Omi, S. (2004) Deactivation of Pt-Pt/Al<sub>2</sub>O<sub>3</sub> in catalytic combustion of methane, Applied Catalysis A: General, 259, 1-7.
- Papadias, D. (2001) Mathematical Modelling of Structured Reactors with Emphasis on Catalytic Combustion Reactions. Ph.D. Thesis in chemical engineering, Department of Chemical Engineering and Technology, Kingliga Tekniska Högskolan (Royal Institute of Technology).
- Pitchon, V. and Fritz, A. (1997) The current state of research on automotive lean NO<sub>x</sub> catalysis, Applied Catalysis B: Environmental, 13, 1-25.
- Pérez-Ramírez, J., Berger, R., Mul, G., Kapteijn, F., and Moulijn, J. A. (2000) The six-flow reactor technology: a review on fast catalyst screening and kinetic studies. Catalysis Today, 60, 93-109.
- Robeiro, F. H., Chow, M., and Dalla Betta, R. A. (1994) Kinetics of the complete oxidation of methane over supported palladium catalysts. Journal of Catalysis, 146, 537-544.
- Ryu, C.K., Ryoo, M.W., Ryu, I. S., and Kang, S.K. (1999) Catalytic combustion of methane over supported bimetallic Pd catalysts: Effects of Ru or Rh addition. Catalysis Today, 47, 141-147
- Sadamori, H. (1999) Application concepts and evaluation of small-scale catalytic combustors for natural gas. Catalysis Today, 47, 325-338.
- Sadamori, H., Tanioka, T., and Matsuhisa, T. (1995) Development of a high temperature combustion catalyst system and prototype catalytic combustor turbine test result. Catalysis Today, 26, 337-344.
- Salomonsson, P., Johansson, S., and Kasermo, B. (1995) Catalysis Letters, 33, 1.
- Sapoundjiew, H. (1999) High potential energy from dilute methane emission. Natural Resources Canada.
- Sekizawa, K., Erguchi, K., Widjaja, H., and Machida, M. (1996) Property of Pd-supported catalysts for catalytic combustion. Catalysis Today, 28, 245-250.

- Sekizawa, K., Widjaja, H., Maeda, S., Ozawa, Y., and Erguchi, K. (2000) Low temperature oxidation of methane supported on metal oxides. Catalysis Today, 59, 69-74.
- Senken, S. (2001) Combinatorial heterogeneous catalysis-A new path in an old field. Angewandte Chemie International Edition, 40, 312-329.
- Snapping Shoals EMC (2001) Electronic service-Green power (Landfill gas project).
- Su, S.C., Carstens, J. N., and Bell, A. T. (1998) A study of the dynamics of Pd oxidation and PdO reduction by H<sub>2</sub> and CH<sub>4</sub>, Journal of Catalysis, 176, 125-135.
- Thevenin, P. (2002) Catalytic Combustion of Methane. Ph.D. Thesis in chemical engineering, Department of Chemical Engineering and Technology, Kingliga Tekniska Högskolan (Royal Institute of Technology).
- Trimm, D. L. (1983) Catalytic combustion (review). Applied Catalysis, 7, 249-282.
- Vatcha, S. R. (1997) Low emission gas turbine using catalytic combustion. Energy Conservation and Management, 38(10-B), 1327-1334.
- Widjaja, H., Sekizawa, K., Erguchi, K., and Arai, H. (1999) Oxidation of methane over Pd/mixed oxide for catalytic combustion. Catalysis Today, 47, 95-101.
- Wit, J. D., Johansen, K., Hansen, P. L., Rossen, H., and Rasmussen, N. B. (2000) Catalytic emission control with respect to CH<sub>4</sub> and CO for highly efficient gas fueled decentralized heat and power production. 5<sup>th</sup> Internatinal Conference on Furnaces and Boilers (INFUB), April 2000, Portugal.
- Yang, L. F., Shi, C. K., He, X., and Cai, J. X. (2002) Catalytic combustion of methane over PdO supported on Mg-modified alumina. Applied Catalysis B: Environmental, 38, 117-125.

## APPENDICES

### Appendix A

**Table A1** Raw data obtained from the screening of catalysts in Library I using eight tubular flow reactors

No	Catalyst	Relative ratio of elemental loading (%)			Remark	Screening results at 650°C		
		Pd	Pt	La		Time (min)	%Conversion	%Selectivity
1	CM	-	-	-	Bare Monolith	15	0.00	100
						83	0.00	100
						151	0.00	100
						219	0.00	100
						287	0.00	100
						355	0.00	100
2	C22	-	-	-	Washcoated Monolith	24	0.00	100
						92	0.00	100
						160	0.00	100
						228	0.00	100
						296	0.00	100
						364	0.00	100
3	C19A	80	20	-	Method A (Pd-based)	32	99.5	100
						100	99.0	100
						168	98.7	100
						236	99.7	100
						304	98.0	100
						372	96.81	100
4	C19B	80	20	-	Method B (Pd-based)	41	99.9	100
						109	100	100
						177	100	100
						245	100	100
						313	100	100
						381	100	100
5	C17A	60	20	20	Method A (Pd-based)	49	97.7	100
						117	97.4	100
						185	97.0	100
						253	96.3	100
						321	95.9	100
						389	95.9	100
6	C17B	60	20	20	Method B (Pd-based)	58	98.9	100
						126	99.0	100
						194	98.5	100
						262	98.2	100
						330	97.8	100
						398	96.1	100

Note: Method A: washcoated monolith was calcined at 500°C and re-calcined at 900°C for 3 hrs after metal loading.

Method B: washcoated monolith was calcined at 900°C and re-calcined at 900°C for 3 hrs after metal loading.

**Table A1** continued

No	Catalyst	Relative ratio of elemental loading (%)			Remark	Screening results at 650°C		
		Pd	Pt	La		Time (min)	%Conversion	%Selectivity
7	C02A	-	80	20	Method A (Pt-based)	66	44.2	100
						134	44.9	100
						202	44.5	100
						270	43.6	100
						338	43.3	100
						406	30.2	100
8	C02B	-	80	20	Method B (Pt-based)	75	34.9	100
						143	33.7	100
						211	28.2	100
						279	27.3	100
						347	17.9	100
						415	18.6	100

Note: Method A: washcoated monolith was calcined at 500°C and re-calcined at 900°C for 3 hrs after metal loading.

Method B: washcoated monolith was calcined at 900°C and re-calcined at 900°C for 3 hrs after metal loading.

**Table A2** Raw data obtained from the screening of catalysts in Library II using eight tubular flow reactors

No	Catalyst	Pd loading (%wt)	Screening results at 350°C			Screening results at 400°C			Screening results at 4500°C		
			Time (min)	%Conversion	%Selectivity	Time (min)	%Conversion	%Selectivity	Time (min)	%Conversion	%Selectivity
1	ML1	1%	15	0.00	100	15	84.03	100	15	0.00	100
			83	0.00	100	83	1.50	100	83	0.00	100
			151	0.53	100	151	1.75	100			
			219	0.41	100	219	0.87	100			
					287		1.02	100			
2	ML2	2%	24	0.00	100	24	0.00	100	24	0.00	100
			92	0.00	100	92	0.86	100	92	0.00	100
			160	0.43	100	160	1.31	100			
			228	0.30	100	228	1.07	100			
					296		0.90	100			
3	ML3	4%	32	19.6	100	32	76.2	100	32	98.6	100
			100	16.9	100	100	43.1	100	100	93.0	100
			168	15.6	100	168	36.3	100			
			236	12.0	100	236	26.4	100			
					304		27.1	100			
4	ML4	5%	41	22.5	100	41	78.8	100	41	99.3	100
			109	15.9	100	109	51.4	100	109	95.3	100
			177	12.6	100	177	25.7	100			
			245	10.5	100	245	20.0	100			
					313		24.7	100			
5	ML5	7%	49	13.3	100	49	77.6	100	49	96.5	100
			117	10.5	100	117	57.9	100	117	93.4	100
			185	10.4	100	185	28.9	100			
			253	8.43	100	253	34.5	100			
					321		28.2	100			

**Table A2** continued

No	Catalyst	Pd loading (%wt)	Screening results at 350°C			Screening results at 400°C			Screening results at 4500°C		
			Time (min)	%Conversion	%Selectivity	Time (min)	%Conversion	%Selectivity	Time (min)	%Conversion	%Selectivity
6	ML6	8%	58	16.9	100	58	80.5	100	58	97.5	100
			126	12.3	100	126	50.9	100	126	92.5	100
			194	13.9	100	194	46.6	100			
			262	11.0	100	262	27.4	100			
						330	23.9	100			
7	ML7	10%	66	37.0	100	66	93.2	100	66	99.0	100
			134	18.3	100	134	80.2	100	134	98.0	100
			202	17.1	100	202	61.4	100			
			270	13.5	100	270	42.6	100			
						338	31.8	100			
8	ML8	12%	75	33.4	100	75	96.1	100	75	99.6	100
			143	18.1	100	143	90.8	100	143	98.5	100
			211	10.6	100	211	73.9	100			
			279	14.5	100	279	68.6	100			
						347	52.9	100			

**Table A3** Raw data obtained from the screening of catalysts in Library III using eight tubular flow reactors

No	Catalysts	Ratio of elements loading (%)			Screening Data											
		Pd	Pt	La	T = 400°C		T = 450°C		T = 500°C		T = 550°C		T = 600°C		T = 800°C	
					Time (min)	% Conv.	Time	% Conv.	Time (min)	% Conv.						
1	C01	0.00	1.00	0.00	15	0.00	15	0.00	15	3.38	15	7.54	15	48.9	15	100
					83	0.00	83	0.00	83	3.54	83	8.12	83	55.0	83	100
					151	0.00	151	0.00	151	3.67	151	7.74	151	54.9	151	100
					219	0.00	219	0.00	219	3.51	219	7.46	219	56.5	219	100
					287	0.00	287	0.00	287	3.28	287	7.33	287	53.3	287	100
					355	0.00	355	0.00	355	3.29	355	7.32	355	51.7	355	100
					423	0.00	423	0.00	423	3.29	423	6.43	423	49.8	423	100
2	C02	0.00	0.80	0.20	15	0.00	15	0.00	15	3.35	15	5.48	15	12.4	15	89.9
					83	0.00	83	0.00	83	0.00	83	4.88	83	11.7	83	89.9
					151	0.00	151	0.00	151	0.00	151	4.41	151	11.0	151	100
					219	0.00	219	0.00	219	0.00	219	5.26	219	9.92	219	100
					287	0.00	287	0.00	287	0.00	287	4.82	287	10.3	287	100
					355	0.00	355	0.00	355	0.00	355	4.76	355	10.2	355	100
					423	0.00	423	0.00	423	0.00	423	3.60	423	10.4	423	100
3	C03	0.00	0.60	0.40	24	0.00	24	0.00	24	0.00	24	6.25	24	6.19	24	100
					92	0.00	92	0.00	92	0.00	92	6.32	92	6.94	92	100
					160	0.00	160	0.00	160	0.00	160	6.12	160	6.66	160	100
					228	0.00	228	0.00	228	0.00	228	6.15	228	5.42	228	100
					296	0.00	296	0.00	296	0.00	296	5.78	296	4.55	296	100
					364	0.00	364	0.00	364	0.00	364	5.75	364	4.33	364	100
					432	0.00	432	0.00	432	0.00	432	5.58	432	3.81	432	100
4	C04	0.00	0.40	0.60	24	0.00	24	0.00	24	0.00	24	6.83	24	12.2	24	99.2
					92	0.00	92	0.00	92	0.00	92	5.80	92	12.4	92	99.4
					160	0.00	160	0.00	160	0.00	160	5.74	160	12.1	160	100
					228	0.00	228	0.00	228	0.00	228	6.17	228	11.3	228	100
					296	0.00	296	0.00	296	0.00	296	5.59	296	10.7	296	100
					364	0.00	364	0.00	364	0.00	364	5.03	364	10.8	364	100
					432	0.00	432	0.00	432	0.00	432	4.13	432	10.5	432	100
5	C05	0.00	0.20	0.80	32	0.00	32	0.00	32	0.00	32	4.05	32	7.45	32	97.4
					100	0.00	100	0.00	100	0.00	100	0.00	100	6.80	100	98.2
					168	0.00	168	0.00	168	0.00	168	0.00	168	6.69	168	95.9
					236	0.00	236	0.00	236	0.00	236	0.00	236	6.28	236	94.8
					304	0.00	304	0.00	304	0.00	304	0.00	304	6.61	304	93.0
					372	0.00	372	0.00	372	0.00	372	0.00	372	6.30	372	90.9
					440	0.00	440	0.00	440	0.00	440	0.00	440	6.26	440	89.4

**Table A3** continued

No	Catalysts	Ratio of elements loading (%)			Screening Data											
		Pd	Pt	La	T = 400°C		T = 450°C		T = 500°C		T = 550°C		T = 600°C		T = 800°C	
					Time (min)	% Conv.	Time	% Conv.	Time (min)	% Conv.						
6	C06	0.00	0.00	1.00	32	0.00	32	0.00	32	0.00	32	0.00	32	9.92	32	63.1
					100	0.00	100	0.00	100	0.00	100	0.00	100	7.43	100	62.3
					168	0.00	168	0.00	168	0.00	168	0.00	168	7.32	168	60.1
					236	0.00	236	0.00	236	0.00	236	0.00	236	5.73	236	64.2
					304	0.00	304	0.00	304	0.00	304	0.00	304	5.60	304	59.5
					372	0.00	372	0.60	372	0.00	372	0.00	372	4.75	372	65.1
					440	0.00	440	0.00	440	0.00	440	0.00	440	3.31	440	63.0
7	C07	0.20	0.80	0.00	15	6.08	15	38.2	15	67.3	15	87.7	15	29.1	15	100
					83	9.58	83	44.8	83	82.5	83	94.8	83	93.0	83	100
					151	9.94	151	49.2	151	89.2	151	96.2	151	92.1	151	100
					219	10.5	219	54.0	219	91.1	219	98.0	219	91.2	219	100
					287	10.8	287	56.4	287	91.6	287	98.4	287	90.6	287	100
					355	11.5	355	54.7	355	91.4	355	98.0	355	90.2	355	100
					423	12.1	423	56.6	423	91.6	423	97.7	423	89.9	423	100
8	C08	0.20	0.60	0.20	24	8.04	24	37.2	24	64.8	24	87.1	24	82.2	24	100
					92	9.55	92	43.5	92	79.9	92	89.7	92	88.7	92	100
					160	9.94	160	46.7	160	87.1	160	89.4	160	90.2	160	100
					228	10.2	228	50.2	228	89.3	228	88.6	228	91.0	228	100
					296	10.0	296	52.2	296	90.1	296	88.9	296	90.3	296	100
					364	10.8	364	51.6	364	89.8	364	87.7	364	89.1	364	100
					432	11.2	432	53.3	432	89.9	432	87.4	432	89.6	432	100
9	C09	0.20	0.40	0.40	41	6.46	41	23.2	41	51.1	41	72.5	41	69.9	41	98.3
					109	6.03	109	27.6	109	58.6	109	75.4	109	78.9	109	98.2
					177	6.96	177	30.3	177	65.2	177	74.9	177	84.4	177	98.3
					245	6.71	245	30.6	245	70.0	245	74.1	245	83.8	245	98.0
					313	5.94	313	31.7	313	72.6	313	73.1	313	89.7	313	97.1
					381	6.77	381	33.1	381	74.2	381	72.6	381	89.3	381	96.7
					449	6.87	449	35.5	449	75.2	449	72.6	449	88.9	449	96.2
10	C10	0.20	0.20	0.60	41	12.5	41	48.3	41	72.4	41	89.3	41	67.6	41	95.1
					109	9.09	109	51.0	109	79.4	109	88.7	109	71.5	109	94.4
					177	8.48	177	54.1	177	84.6	177	89.0	177	73.6	177	92.0
					245	8.31	245	56.6	245	79.4	245	92.5	245	74.7	245	85.7
					313	7.14	313	59.3	313	77.8	313	92.5	313	75.7	313	89.9
					381	7.84	381	61.9	381	79.9	381	90.8	381	76.4	381	88.6
					449	11.6	449	64.5	449	86.3	449	89.9	449	76.8	449	87.5

**Table A3** continued

No	Catalysts	Ratio of elements loading (%)			Screening Data											
		Pd	Pt	La	T = 400°C		T = 450°C		T = 500°C		T = 550°C		T = 600°C		T = 800°C	
					Time (min)	% Conv.	Time	% Conv.	Time (min)	% Conv.						
11	C11	0.20	0.00	0.80	49	11.7	66	9.69	66	28.2	66	47.0	49	96.9	49	88.4
					117	13.1	134	9.15	134	26.2	134	43.9	117	96.9	117	90.6
					185	12.9	202	9.23	202	25.5	202	42.6	185	96.7	185	90.0
					253	13.1	270	8.96	270	24.8	270	41.4	253	96.5	253	88.1
					321	12.9	338	14.4	338	30.0	338	40.4	321	96.4	321	88.6
					389	11.4	406	16.0	406	31.4	406	39.9	389	96.2	389	88.0
					457	10.3	474	22.7	474	30.0	474	39.8	457	96.0	457	87.3
12	C12	0.40	0.60	0.00	49	9.44	58	30.2	49	67.4	49	84.9	49	81.1	49	100
					117	9.87	126	38.5	117	75.9	117	87.3	117	87.7	117	100
					185	9.71	194	42.0	185	81.0	185	87.3	185	89.9	185	100
					253	10.2	262	44.5	253	83.1	253	87.1	253	88.0	253	100
					321	9.97	330	50.5	321	83.9	321	87.0	321	88.7	321	100
					389	11.1	398	55.2	389	84.2	389	86.9	389	86.7	389	100
					457	11.5	466	58.8	457	84.7	457	87.0	457	86.0	457	100
13	C13	0.40	0.40	0.20	32	6.47	32	35.6	32	59.2	32	75.6	32	95.2	32	100
					100	6.71	100	39.0	100	68.3	100	77.9	100	94.1	100	100
					168	6.98	168	42.1	168	74.7	168	78.4	168	93.5	168	100
					236	7.61	236	45.0	236	77.8	236	77.7	236	93.1	236	100
					304	7.49	304	45.7	304	79.6	304	77.1	304	92.6	304	100
					372	7.91	372	43.1	372	79.2	372	76.5	372	92.2	372	100
					440	8.10	440	44.2	440	80.6	440	76.3	440	92.0	440	100
14	C14	0.40	0.20	0.40	58	14.8	58	35.0	58	67.9	58	75.2	58	93.2	58	89.3
					126	14.9	126	37.8	126	73.7	126	75.7	126	93.8	126	90.9
					194	14.0	194	39.6	194	75.8	194	74.1	194	93.8	194	93.1
					262	13.5	262	42.5	262	76.5	262	72.9	262	92.1	262	87.2
					330	14.4	330	43.6	330	76.4	330	71.6	330	93.1	330	82.8
					398	13.6	398	45.9	398	76.2	398	71.6	398	90.9	398	79.7
					466	13.9	466	45.0	466	75.9	466	71.4	466	90.4	466	77.2
15	C15	0.40	0.00	0.60	58	15.7	58	50.5	58	83.3	58	96.7	58	97.9	58	96.8
					126	10.5	126	45.6	126	76.8	126	95.0	126	96.2	126	98.5
					194	10.7	194	45.0	194	72.3	194	95.0	194	95.1	194	98.2
					262	13.2	262	43.5	262	74.2	262	92.6	262	93.7	262	98.5
					330	13.1	330	43.0	330	74.4	330	90.9	330	92.8	330	95.8
					398	13.8	398	42.2	398	73.3	398	90.4	398	91.5	398	93.2
					466	11.6	466	41.7	466	75.2	466	91.9	466	90.6	466	91.5

**Table A3** continued

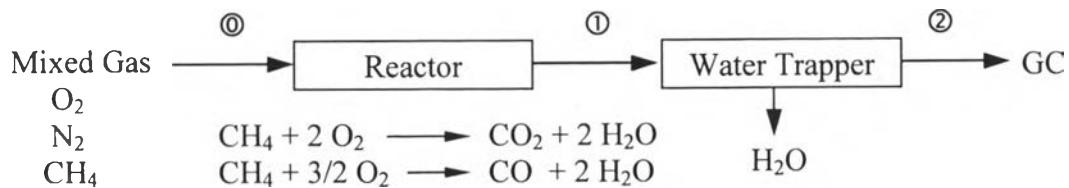
No	Catalysts	Ratio of elements loading (%)			Screening Data											
		Pd	Pt	La	T = 400°C		T = 450°C		T = 500°C		T = 550°C		T = 600°C		T = 800°C	
					Time (min)	% Conv.	Time	% Conv.	Time (min)	% Conv.						
16	C16	0.60	0.40	0.00	66	5.46	66	40.5	66	82.3	66	95.7	66	93.9	66	96.8
					134	11.1	134	41.8	134	84.6	134	96.1	134	94.3	134	89.2
					202	11.0	202	47.8	202	89.2	202	96.4	202	93.9	202	88.1
					270	12.3	270	46.3	270	90.8	270	94.6	270	93.8	270	87.7
					338	12.3	338	48.5	338	90.4	338	94.4	338	93.8	338	87.1
					406	13.6	406	49.2	406	85.6	406	96.0	406	94.0	406	87.9
					474	10.5	474	52.1	474	82.2	474	96.8	474	93.4	474	85.5
17	C17	0.60	0.20	0.20	66	23.2	66	43.3	66	73.5	66	77.4	66	96.9	66	96.0
					134	23.0	134	45.7	134	76.4	134	78.2	134	97.0	134	96.1
					202	21.8	202	47.6	202	77.7	202	76.9	202	96.9	202	94.6
					270	22.0	270	49.5	270	78.2	270	74.8	270	96.0	270	91.7
					338	23.4	338	48.9	338	77.7	338	74.4	338	96.7	338	89.3
					406	23.6	406	49.4	406	78.5	406	74.1	406	90.7	406	88.3
					474	23.9	474	49.1	474	78.4	474	74.1	474	90.0	474	86.9
18	C18	0.60	0.00	0.40	41	17.4	41	40.8	41	61.0	41	88.0	41	99.6	41	100
					109	15.5	109	38.5	109	55.9	109	85.0	109	98.2	109	100
					177	21.6	177	36.8	177	53.8	177	84.1	177	98.2	177	100
					245	15.4	245	35.7	245	50.4	245	82.8	245	97.6	245	95.4
					313	14.4	313	30.7	313	50.1	313	81.8	313	97.3	313	96.4
					381	14.5	381	25.9	381	46.0	381	81.1	381	96.8	381	98.7
					449	15.3	449	25.7	449	47.3	449	80.5	449	96.3	449	100
19	C19	0.80	0.20	0.00	75	26.6	75	56.0	75	84.1	75	89.9	75	98.2	75	90.0
					143	26.0	143	54.6	143	83.5	143	89.7	143	97.7	143	91.8
					211	24.3	211	56.9	211	82.3	211	89.1	211	97.3	211	90.4
					279	24.8	279	57.9	279	81.7	279	88.4	279	97.2	279	93.4
					347	25.6	347	58.0	347	81.7	347	87.0	347	96.7	347	90.3
					415	26.6	415	58.4	415	80.1	415	86.6	415	96.3	415	88.0
					483	26.3	483	58.0	483	79.9	483	86.0	483	96.3	483	87.5
20	C20	0.80	0.00	0.20	49	27.8	49	52.2	49	76.4	49	97.0	49	97.8	49	80.5
					117	25.3	117	48.9	117	69.8	117	95.3	117	96.2	117	80.9
					185	23.4	185	46.1	185	65.5	185	94.3	185	94.6	185	80.6
					253	23.2	253	45.7	253	62.3	253	92.5	253	93.7	253	77.0
					321	22.4	321	43.6	321	57.3	321	91.2	321	92.6	321	75.8
					389	22.5	389	48.6	389	50.3	389	90.3	389	91.5	389	77.2
					457	20.9	457	49.0	457	53.7	457	89.4	457	90.8	457	76.6

**Table A3** continued

No	Catalysts	Ratio of elements loading (%)			Screening Data													
		Pd	Pt	La	T = 400°C		T = 450°C		T = 500°C		T = 550°C		T = 600°C		T = 800°C			
					Time (min)	% Conv.	Time	% Conv.	Time (min)	% Conv.								
21	C21	1.00	0.00	0.00	58	42.1	75	38.6	75	70.0	75	95.7	58	100	58	100		
					126	37.9	143	34.1	143	63.5	143	94.3	126	99.4	126	100		
					194	36.2	211	32.3	211	59.6	211	92.4	194	98.7	194	100		
					262	35.1	279	32.2	279	56.1	279	91.7	262	98.3	262	99.1		
					330	33.7	347	49.9	347	59.0	347	90.0	330	97.8	330	99.0		
					398	33.0	415	60.5	415	58.8	415	88.9	398	97.6	398	100		
					466	32.3	483	60.7	483	56.8	483	88.7	466	97.0	466	100		
22	C22	Washcoated Monolith			66	0.00	75	0.00	75	0.00	75	0.00	66	0.00	66	97.9		
					134	0.00	143	0.00	143	0.00	143	0.00	134	0.00	134	98.1		
					202	0.00	211	0.00	211	0.00	211	0.00	202	0.00	202	98.7		
					270	0.00	279	0.00	279	0.00	279	0.00	270	0.00	270	99.2		
					338	0.00	347	0.00	347	0.00	347	0.00	338	0.00	338	98.9		
					406	0.00	415	0.00	415	0.00	415	0.00	406	0.00	406	98.5		
					474	0.00	483	0.00	483	0.00	483	0.00	474	0.00	474	98.9		
23	C23	Only quartz wool			75	0.00	75	0.00	75	0.00	75	0.00	75	0.00	75	22.6		
					143	0.00	143	0.00	143	0.00	143	0.00	143	0.00	143	28.9		
					211	0.00	211	0.00	211	0.00	211	0.00	211	0.00	211	21.1		
					279	0.00	279	0.00	279	0.00	279	0.00	279	0.00	279	19.0		
					347	0.00	347	0.00	347	0.00	347	0.00	347	0.00	347	18.1		
					415	0.00	415	0.00	415	0.00	415	0.00	415	0.00	415	20.4		
					483	0.00	483	0.00	483	0.00	483	0.00	483	0.00	483	12.3		

## Appendix B

**System schematic:**



**Streams details:**

◆ Stream #0: Volumetric flow rate:  $F_0 \text{ cm}^3/\text{min}$

Compositions:	- Methane fraction: $y_{\text{Me},0}$
	- Oxygen fraction: $y_{\text{O}_2,0}$
	- Nitrogen fraction: $y_{\text{N}_2,0}$

◆ Stream #1: Volumetric flow rate:  $F_1 \text{ cm}^3/\text{min}$

Compositions:	- Methane fraction: $y_{\text{Me},1}$
	- Oxygen fraction: $y_{\text{O}_2,1}$
	- Nitrogen fraction: $y_{\text{N}_2,1}$
	- Carbon dioxide: $y_{\text{CO}_2,1}$
	- Carbon monoxide: $y_{\text{CO},1}$
	- Water: $y_{\text{H}_2\text{O},1}$

◆ Stream #2: Volumetric flow rate:  $F_2 \text{ cm}^3/\text{min}$

Compositions:	- Methane fraction: $y_{\text{Me},2}$
	- Oxygen fraction: $y_{\text{O}_2,2}$
	- Nitrogen fraction: $y_{\text{N}_2,2}$
	- Carbon dioxide: $y_{\text{CO}_2,2}$
	- Carbon monoxide: $y_{\text{CO},2}$

**Assumptions:**

- no NO<sub>x</sub> formation during the combustion since the combustion is occurred below 1500°C
- the analyzed compositions received from GC are the compositions of the stream #2

**Conversion:**

$$\text{Conversion} = \frac{\text{Initial methane fed} - \text{Residual methane}}{\text{Initial methane fed}} \times 100\%$$

$$= \frac{\frac{P_0 F_0}{R T_0} \cdot y_{Me,0} - \frac{P_1 F_1}{R T_1} \cdot y_{Me,1}}{\frac{P_0 F_0}{R T_0} \cdot y_{Me,0}} \times 100\%$$

**Assume:** Pressure remains constant along the combustion reaction, thus

$$\text{Conversion} = \frac{\frac{F_0}{T_0} \cdot y_{Me,0} - \frac{F_1}{T_1} \cdot y_{Me,1}}{\frac{F_0}{T_0} \cdot y_{Me,0}} \times 100\%$$

Carbon balanced around the reactor

$$\frac{P_0 F_0}{R T_0} \cdot y_{Me,0} = \frac{P_1 F_1}{R T_1} (y_{Me,1} + y_{CO_2,1} + y_{CO,1})$$

**Assume:** Pressure remains constant along the combustion reaction, so

$$\frac{F_0}{T_0} \cdot y_{Me,0} = \frac{F_1}{T_1} (y_{Me,1} + y_{CO_2,1} + y_{CO,1})$$

Therefore, the conversion can be written in the new form of evaluated data as below,

$$\text{Conversion} = \frac{\frac{F_1}{T_1}(y_{Me,1} + y_{CO_2,1} + y_{CO,1}) - \frac{F_1}{T_1} \cdot y_{Me,1}}{\frac{F_1}{T_1}(y_{Me,1} + y_{CO_2,1} + y_{CO,1})} \times 100\%$$

But, the evaluated compositions from GC are the compositions of stream #2. Therefore, carbon and methane balances are needed in order to convert these compositions to the flue composition from the reactor.

Carbon balanced around the water trapper

$$\frac{F_2}{T_2}(y_{Me,2} + y_{CO_2,2} + y_{CO,2}) = \frac{F_1}{T_1}(y_{Me,1} + y_{CO_2,1} + y_{CO,1})$$

Methane Balanced around Water Trapper:

$$\frac{F_2}{T_2} \cdot y_{Me,2} = \frac{F_1}{T_1} \cdot y_{Me,1}$$

Thus,

$$\begin{aligned} \text{Conversion} &= \frac{\frac{F_2}{T_2}(y_{Me,2} + y_{CO_2,2} + y_{CO,2}) - \frac{F_2}{T_2} \cdot y_{Me,2}}{\frac{F_2}{T_2}(y_{Me,2} + y_{CO_2,2} + y_{CO,2})} \times 100\% \\ &= \frac{y_{CO_2,2} + y_{CO,2}}{y_{Me,2} + y_{CO_2,2} + y_{CO,2}} \times 100\% \end{aligned}$$

**Selectivity:**

$$\text{Selectivity}_{\text{CO}_2} = \frac{\text{Mole of CO}_2 \text{ generated}}{\text{Mole of CH}_4 \text{ consumed}}$$

$$= \frac{\frac{P_1 F_1}{R T_1} \cdot y_{\text{CO}_2,1}}{\frac{P_0 F_0}{R T_0} \cdot y_{\text{Me},0} - \frac{P_1 F_1}{R T_1} \cdot y_{\text{Me},1}}, \text{ and}$$

$$\text{Selectivity}_{\text{CO}} = \frac{\frac{P_1 F_1}{R T_1} \cdot y_{\text{CO},1}}{\frac{P_0 F_0}{R T_0} \cdot y_{\text{Me},0} - \frac{P_1 F_1}{R T_1} \cdot y_{\text{Me},1}}$$

Thus,

$$\% \text{ Selectivity}_{\text{CO}_2/\text{CO}} = \frac{\text{Selectivity to CO}_2}{\text{Selectivity to CO} + \text{Selectivity to CO}_2} \times 100\%$$

$$= \frac{y_{\text{CO}_2,1}}{y_{\text{CO},1} + y_{\text{CO}_2,1}} \times 100\%$$

From CO and CO<sub>2</sub> balanced around the water trapper, we can rewrite the above equation as,

$$\text{Selectivity} = \frac{y_{\text{CO}_2,2}}{y_{\text{CO},2} + y_{\text{CO}_2,2}} \times 100\%$$

## CURRICULUM VITAE

**Name:** Mr. Natthakorn Kraikul

**Date of Birth:** 20<sup>th</sup> October, 1980

**Nationality:** Thai

**University Education:**

1998-2001 Bachelor Degree of Science in Industrial Chemistry, Faculty of Applied Science, King Mongkut's Institute of Technology North Bangkok, Bangkok, Thailand.