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APPENDICES

Appendix A

Table A1 Raw data obtained from the screening of catalysts by using eight tubular flow reactors

No	Catalysts	Relative ratio of elemental loading (%)				Screening Data									
		Pd	Sn	Ti	Zr	T = 450°C		T = 500°C		T = 550°C		T = 600°C		T = 650°C	
						Time (min)	% Conv.	Time (min)	% Conv.	Time (min)	% Conv.	Time (min)	% Conv.	Time (min)	% Conv.
1	C1	4	0.00	1.00	0.00	32.0	8.87	32.0	19.6	32.0	33.7	32.0	41.9	32.0	37.5
						100	5.22	100	13.1	100	24.5	100	33.8	100	33.1
						168	7.68	168	13.0	168	32.9	168	32.2	168	31.9
						236	5.05	236	11.9	236	23.4	236	31.5	236	31.2
						304	0.00	304	11.2	304	20.3	304	30.7	304	30.5
						372	1.27	372	11.1	372	19.7	372	29.2	372	29.8
						440	0.00	440	10.8	440	19.0	440	28.3	440	32.1
2	C2	4	0.00	0.80	0.20	40.5	8.34	40.5	15.2	40.5	32.0	40.5	48.9	40.5	57.9
						108	6.10	108	12.2	108	27.7	108	46.0	108	56.7
						176	5.66	176	11.4	176	26.8	176	45.6	176	54.4
						244	5.53	244	11.2	244	26.7	244	45.3	244	52.9
						312	5.46	312	10.8	312	25.9	312	43.9	312	51.0
						380	5.45	380	10.7	380	25.1	380	41.1	380	49.9
						448	5.51	448	10.6	448	23.6	448	40.8	448	48.5
3	C3	4	0.00	0.60	0.40	32.0	11.7	32.0	20.3	32.0	30.1	32.0	35.4	32.0	32.0
						100	8.05	100	13.1	100	19.3	100	25.7	100	26.5
						168	6.65	168	11.0	168	17.3	168	23.6	168	21.6
						236	6.06	236	10.0	236	16.1	236	21.7	236	19.7
						304	5.64	304	9.46	304	15.3	304	20.9	304	18.6
						372	5.47	372	9.36	372	14.4	372	18.0	372	17.4
						440	5.31	440	9.00	440	13.8	440	18.3	440	16.6

Table A1 continued

4	C4	4	0.00	0.40	0.60	23.5	33.9	23.5	52.1	23.5	50.7	23.5	71.4	23.5	65.8
						91.5	22.3	91.5	27.0	91.5	35.7	91.5	43.3	91.5	55.0
						159	15.6	159	23.3	159	31.7	159	37.4	159	48.4
						227	14.3	227	21.4	227	28.5	227	34.6	227	45.9
						295	13.6	295	20.3	295	27.1	295	32.9	295	43.9
						363	12.7	363	19.6	363	26.1	363	31.4	363	45.3
						431	12.3	431	18.8	431	25.0	431	30.2	431	41.8
5	C5	4	0.00	0.20	0.80	15.0	28.7	15.0	31.3	15.0	39.5	15.0	0.00	15.0	31.8
						83.0	10.5	83.0	8.95	83.0	25.9	83.0	31.5	83.0	35.6
						151	11.6	151	15.7	151	21.9	151	26.2	151	35.0
						219	8.47	219	14.4	219	19.9	219	23.4	219	28.8
						287	7.77	287	13.6	287	18.9	287	22.0	287	26.6
						355	7.35	355	12.8	355	17.7	355	23.1	355	26.8
						423	7.15	423	12.2	423	16.7	423	20.1	423	24.7
6	C6	4	0.00	0.00	1.00	15.0	6.15	15.0	0.00	15.0	13.9	15.0	14.6	15.0	32.3
						83.0	0.00	83.0	6.93	83.0	13.2	83.0	18.4	83.0	25.1
						151	0.00	151	5.87	151	10.8	151	15.6	151	21.9
						219	0.00	219	5.10	219	9.62	219	13.8	219	20.6
						287	0.00	287	4.74	287	9.23	287	13.0	287	19.9
						355	0.00	355	5.30	355	8.45	355	12.3	355	19.2
						423	0.00	423	5.35	423	8.24	423	11.7	423	21.9
7	C7	4	0.20	0.80	0.00	74.5	5.42	74.5	17.7	74.5	17.6	74.5	31.5	74.5	39.5
						142	0.00	142	14.6	142	19.6	142	23.3	142	33.2
						210	0.249	210	13.9	210	17.4	210	20.6	210	30.2
						278	0.00	278	13.2	278	16.6	278	19.6	278	30.2
						346	0.00	346	11.9	346	14.8	346	18.4	346	25.8
						414	0.00	414	11.8	414	15.5	414	17.9	414	26.0
						482	0.273	482	11.5	482	14.6	482	17.5	482	25.2

Table A1 continued

8	C8	4	0.20	0.60	0.20	15.0	17.3	15.0	34.8	15.0	64.6	15.0	74.1	15.0	66.5
						83.0	6.82	83.0	15.8	83.0	32.5	83.0	45.7	83.0	45.5
						151	6.73	151	14.6	151	39.0	151	39.7	151	41.4
						219	5.60	219	13.3	219	24.5	219	36.6	219	39.9
						287	5.27	287	12.8	287	21.8	287	34.8	287	38.3
						355	4.86	355	13.6	355	21.4	355	33.4	355	36.3
						423	4.88	423	12.7	423	20.5	423	31.5	423	35.7
9	C9	4	0.20	0.40	0.40	49.0	12.7	49.0	14.4	49.0	33.8	49.0	55.1	49.0	61.8
						117	9.55	117	10.9	117	27.4	117	43.0	117	56.4
						185	8.28	185	20.5	185	24.7	185	39.1	185	52.9
						253	8.01	253	19.4	253	23.8	253	36.6	253	50.5
						321	10.5	321	18.5	321	22.3	321	34.8	321	49.9
						389	7.80	389	17.9	389	21.3	389	33.4	389	47.0
						457	7.52	457	17.4	457	20.1	457	32.3	457	43.4
10	C10	4	0.20	0.20	0.60	32.0	11.8	32.0	20.8	32.0	28.7	32.0	48.7	32.0	51.5
						100	12.0	100	17.1	100	22.7	100	35.2	100	45.0
						168	7.72	168	13.2	168	20.4	168	31.3	168	40.5
						236	11.2	236	12.0	236	19.0	236	29.2	236	37.1
						304	7.11	304	11.4	304	18.0	304	27.5	304	36.1
						372	11.0	372	11.2	372	16.9	372	26.4	372	36.3
						440	6.67	440	10.8	440	16.4	440	25.7	440	35.7
11	C11	4	0.20	0.00	0.80	23.5	9.33	23.5	21.9	23.5	41.9	23.5	62.0	23.5	66.4
						91.5	8.32	91.5	15.6	91.5	32.0	91.5	45.8	91.5	45.6
						159	7.75	159	14.6	159	29.7	159	41.8	159	40.9
						227	7.51	227	13.9	227	27.8	227	39.2	227	37.7
						295	7.46	295	13.5	295	27.0	295	37.7	295	36.3
						363	7.21	363	14.2	363	25.7	363	35.[illegible]	363	35.0
						431	7.06	431	14.2	431	24.8	431	34.3	431	33.9

Table A1 continued

12	C12	4	0.40	0.60	0.00	40.5	0.00	40.5	8.35	40.5	16.3	40.5	25.7	40.5	30.7
						108	0.30	108	7.16	108	13.9	108	22.5	108	27.6
						176	0.00	176	6.91	176	12.6	176	21.9	176	27.6
						244	0.106	244	6.49	244	12.2	244	21.6	244	26.5
						312	0.00	312	6.47	312	12.0	312	21.6	312	24.9
						380	0.00	380	6.23	380	11.7	380	20.1	380	24.2
						448	0.00	448	6.39	448	12.3	448	21.9	448	26.4
13	C13	4	0.40	0.40	0.20	57.5	0.00	57.5	14.5	57.5	23.9	57.5	36.8	57.5	42.5
						125	0.00	125	12.5	125	18.6	125	34.8	125	39.4
						193	0.00	193	11.3	193	16.9	193	32.7	193	37.1
						261	0.00	261	10.7	261	16.6	261	31.7	261	36.1
						329	0.00	329	10.4	329	15.9	329	31.0	329	34.3
						397	0.00	397	9.69	397	15.9	397	32.3	397	33.1
						465	0.00	465	9.65	465	15.4	465	31.5	465	31.7
14	C14	4	0.40	0.20	0.40	57.5	7.43	57.5	12.4	57.5	31.6	57.5	37.8	57.5	49.4
						125	5.98	125	10.6	125	26.4	125	30.5	125	44.9
						193	5.60	193	9.72	193	23.7	193	27.2	193	44.1
						261	5.60	261	9.29	261	22.4	261	26.0	261	41.3
						329	5.41	329	9.15	329	21.4	329	24.5	329	40.7
						397	5.34	397	8.91	397	20.5	397	23.7	397	38.7
						465	5.68	465	8.61	465	19.7	465	23.3	465	37.2
15	C15	4	0.40	0.00	0.60	40.5	4.61	40.5	18.6	40.5	39.1	40.5	68.8	40.5	89.6
						108	0.00	108	20.0	108	43.2	108	65.5	108	85.9
						176	5.65	176	21.6	176	44.4	176	63.6	176	83.7
						244	9.59	244	22.7	244	45.0	244	61.0	244	82.1
						312	9.36	312	23.1	312	44.9	312	59.3	312	82.1
						380	6.20	380	26.9	380	43.2	380	59.1	380	81.9
						448	10.1	448	24.1	448	42.8	448	58.0	448	80.6

Table A1 continued

						49.0	0.00	49.0	11.0	49.0	20.9	49.0	31.9	49.0	42.6
						117	0.00	117	8.32	117	16.3	117	27.8	117	38.4
						185	0.00	185	7.42	185	15.5	185	26.3	185	35.5
16	C16	4	0.60	0.40	0.00	253	0.00	253	7.70	253	14.9	253	23.6	253	32.3
						321	0.00	321	7.48	321	14.4	321	22.3	321	32.5
						[redacted]									
						457	0.00	457	6.63	457	13.5	457	20.2	457	30.2
						66.0	0.00	66.0	7.00	66.0	13.0	66.0	23.7	66.0	33.7
						134	0.00	134	6.66	134	11.1	134	23.5	134	29.6
						202	0.00	202	6.03	202	10.3	202	23.3	202	27.2
17	C17	4	0.60	0.20	0.20	270	0.00	270	5.74	270	10.1	270	23.1	270	25.8
						338	0.00	338	5.72	338	9.83	338	22.2	338	24.3
						[redacted]									
						474	0.00	474	4.85	474	9.57	474	21.45	474	22.0
						66.0	4.00	66.0	11.9	66.0	33.2	66.0	43.6	66.0	63.5
						134	0.00	134	11.7	134	31.3	134	39.8	134	60.3
						202	0.00	202	11.8	202	30.7	202	38.5	202	59.7
18	C18	4	0.60	0.00	0.40	270	0.228	270	12.1	270	29.8	270	36.2	270	57.5
						338	0.914	338	12.4	338	29.4	338	35.6	338	57.3
						[redacted]									
						474	0.00	474	12.5	474	28.0	474	33.8	474	54.7
						57.5	0.00	57.5	7.15	57.5	18.4	57.5	22.4	57.5	29.0
						125	0.00	125	6.08	125	15.6	125	18.6	125	25.0
						193	0.00	193	5.46	193	14.0	193	17.0	193	24.0
19	C19	4	0.80	0.20	0.00	261	0.00	261	5.35	261	13.1	261	15.6	261	23.1
						329	0.00	329	7.10	329	12.4	329	14.9	329	22.9
						[redacted]									
						465	0.00	465	4.77	465	11.6	465	13.5	465	21.5

Table A1 continued

20	C20	4	0.80	0.00	0.20	23.5	0.00	23.5	0.00	23.5	12.5	23.5	21.4	23.5	35.7
						91.5	0.00	91.5	0.00	91.5	10.8	91.5	20.4	91.5	29.1
						159	0.00	159	0.00	159	19.3	159	16.5	159	27.1
						227	0.00	227	0.00	227	9.99	227	18.3	227	25.3
						295	0.00	295	0.210	295	9.91	295	17.7	295	24.0
						363	0.00	363	0.00	363	9.82	363	16.9	363	23.2
						431	0.00	431	0.00	431	9.59	431	16.3	431	22.5
21	C21	4	1.00	0.00	0.00	66.0	0.00	66.0	0.00	66.0	8.01	66.0	16.1	66.0	23.3
						134	0.00	134	0.00	134	7.61	134	15.5	134	20.6
						202	0.00	202	0.00	202	7.35	202	15.2	202	20.1
						270	0.00	270	0.00	270	7.46	270	14.6	270	19.9
						338	0.00	338	0.00	338	7.18	338	14.3	338	19.3
						406	1.49	406	0.00	406	7.18	406	13.8	406	19.0
						474	0.00	474	0.00	474	6.05	474	13.6	474	18.3
22	C22	4	0.00	0.00	0.00	74.5	14.1	74.5	24.8	74.5	39.7	74.5	48.2	74.5	49.6
						142	10.8	142	21.8	142	32.7	142	41.2	142	41.6
						210	11.1	210	20.1	210	29.3	210	35.8	210	39.7
						278	10.4	278	18.0	278	27.9	278	34.4	278	36.8
						346	9.74	346	17.2	346	26.3	346	31.8	346	32.9
						414	9.60	414	16.5	414	21.4	414	29.1	414	34.1
						482	9.08	482	14.3	482	23.9	482	27.7	482	31.9

Table A2 Raw data obtained from lead formulations selected from the activity screening results using the eight-tubular-flow reactors.

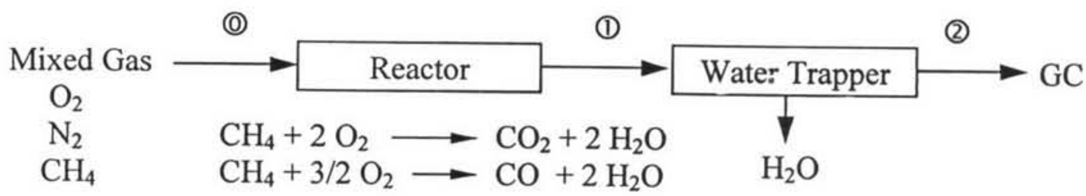
No	Catalysts	Relative ratio of elemental loading (%)				Screening Data									
		Pd	Sn	Ti	Zr	T = 450°C		T = 500°C		T = 550°C		T = 600°C		T = 650°C	
						Time (min)	% Conv.	Time (min)	% Conv.	Time (min)	% Conv.	Time (min)	% Conv.	Time (min)	% Conv.
1	C02	4	0.00	0.80	0.20	40.5	0.00	40.5	1.20	40.5	0.00	40.5	0.00	40.5	0.00
						108	0.00	108	2.00	108	3.60	108	14.0	108	6.40
						176	2.60	176	1.00	176	3.40	176	3.30	176	6.00
						244	3.10	244	3.30	244	1.20	244	0.00	244	7.40
						312	1.00	312	0.00	312	0.00	312	0.00	312	4.20
						380	0.000	380	0.00	380	0.00	380	0.00	380	5.90
2	C03	4	0.00	0.60	0.40	32.0	3.30	32.0	8.90	32.0	11.1	32.0	14.5	32.0	22.7
						100	4.10	100	0.00	100	7.10	100	11.5	100	16.2
						168	0.00	168	2.40	168	1.30	168	6.40	168	15.8
						236	0.00	236	6.80	236	3.50	236	3.30	236	10.1
						304	2.30	304	0.20	304	8.30	304	3.40	304	10.9
						372	0.00	372	1.90	372	1.30	372	10.9	372	12.9
3	C04	4	0.00	0.4	0.6	15.0	6.70	15.0	14.3	15.0	14.0	15.0	26.0	15.0	39.9
						83.0	2.40	83.0	10.5	83.0	10.1	83.0	16.9	83.0	43.7
						151	3.50	151	10.3	151	4.20	151	17.6	151	50.2
						273	1.10	273	3.90	273	7.80	273	18.8	273	51.1
						287	0.40	287	3.50	287	7.30	287	23.9	287	51.2
						355	0.00	355	2.40	355	0.90	355	19.9	355	47.0

Table A2 continued

4	C09	4	0.2	0.4	0.4	23.5	0.00	23.5	4.60	23.5	1.50	23.5	4.10	23.5	8.30
						91.5	0.00	91.5	1.80	91.5	0.00	91.5	2.50	91.5	9.80
						159	0.00	159	0.00	159	0.00	159	0.00	159	10.4
						227	0.00	227	0.00	227	0.00	227	2.30	227	9.70
						295	0.00	295	0.00	295	0.00	295	0.00	295	3.60
						363	0.00	363	0.00	363	0.00	363	0.00	363	6.90
5	C10	4	0.2	0.2	0.6	40.5	17.9	40.5	16.8	40.5	11.7	40.5	12.6	40.5	17.2
						108	15.2	108	20.1	108	11.2	108	12.5	108	25.5
						176	17.3	176	22.0	176	9.30	176	10.9	176	30.7
						244	17.4	244	19.0	244	10.2	244	9.90	244	36.3
						312	16.5	312	16.5	312	6.40	312	7.60	312	39.5
						380	14.4	380	14.2	380	5.80	380	11.90	380	37.3
6	C14	4	0.4	0.2	0.4	57.5	0.00	57.5	0.80	57.5	3.10	57.5	1.80	57.5	1.60
						125	0.00	125	0.00	125	0.20	125	0.30	125	8.00
						193	0.00	193	1.60	193	0.00	193	0.30	193	10.0
						261	0.00	261	0.00	261	0.00	261	0.00	261	13.1
						329	0.00	329	0.00	329	0.00	329	0.00	329	9.90
						397	0.00	397	0.00	397	0.00	397	0.00	397	15.7
7	C15	4	0.4	0.0	0.6	57.5	0.00	57.5	3.40	57.5	6.90	57.5	9.90	57.5	28.9
						125	0.00	125	3.50	125	6.60	125	9.60	125	39.8
						193	0.00	193	4.00	193	3.90	193	10.0	193	44.3
						261	0.00	261	2.80	261	3.50	261	7.90	261	51.5
						329	0.00	329	0.00	329	1.70	329	5.60	329	56.1
						397	0.00	397	0.00	397	6.60	397	7.80	397	57.4
8	C18	4	0.6	0.00	0.4	40.5	9.60	40.5	10.9	40.5	13.1	40.5	20.3	40.5	84.5
						108	7.00	108	10.2	108	14.1	108	22.5	108	90.9
						176	9.00	176	9.70	176	13.7	176	23.9	176	91.4
						244	10.3	244	9.00	244	16.5	244	24.9	244	89.3
						312	10.8	312	7.50	312	14.3	312	23.9	312	91.5
						380	10.6	380	4.80	380	16.4	380	24.4	380	90.5

Appendix B

Calculations:



Streams Details:

- Stream #0: Volumetric flow rate: F_0 cm^3/min

  Compositions:
  - Methane fraction: $y_{Me,0}$
  - Oxygen fraction: $y_{O2,0}$
  - Nitrogen fraction: $y_{N2,0}$

- Stream #1: Volumetric flow rate: F_1 cm^3/min

  Compositions:
  - Methane fraction: $y_{Me,1}$
  - Oxygen fraction: $y_{O2,1}$
  - Nitrogen fraction: $y_{N2,1}$
  - Carbon dioxide: $y_{CO2,1}$
  - Carbon monoxide: $y_{CO,1}$
  - Water: $y_{H2O,1}$

- Stream #2: Volumetric flow rate: F_2 cm^3/min

  Compositions:
  - Methane fraction: $y_{Me,2}$
  - Oxygen fraction: $y_{O2,2}$
  - Nitrogen fraction: $y_{N2,2}$
  - Carbon dioxide: $y_{CO2,2}$
  - Carbon monoxide: $y_{CO,2}$

Assumptions:

- no NO_X 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:

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

$$= \frac{\frac{P_0F_0}{RT_0} \cdot y_{Me,0} - \frac{P_1F_1}{RT_1} \cdot y_{Me,1}}{\frac{P_0F_0}{RT_0} \cdot y_{Me,0}} \times 100\%$$

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

$$\frac{P_0F_0}{RT_0} \cdot y_{Me,0} = \frac{P_1F_1}{RT_1}(y_{Me,1} + y_{CO2,1} + y_{CO,1})$$

Assume: Pressure remains constant along the combustion reaction, so

$$\frac{F_0}{T_0} \cdot y_{Me,0} = \frac{F_1}{T_1}(y_{Me,1} + y_{CO2,1} + y_{CO,1})$$

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

$$\text{Conversion} = \frac{\frac{F_1}{T_1}(y_{Me,1} + y_{CO2,1} + y_{CO,1}) - \frac{F_1}{T_1} \cdot y_{Me,1}}{\frac{F_1}{T_1}(y_{Me,1} + y_{CO2,1} + y_{CO,1})} \times 100\%$$

But, the evaluated compositions from GC are the compositions for 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

$$\frac{F_2}{T_2}(y_{Me,2} + y_{CO2,2} + y_{CO,2}) = \frac{F_1}{T_1}(y_{Me,1} + y_{CO2,1} + y_{CO,1})$$

Methane Balanced around Water Trapper:

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

Thus,

$$\text{Conversion} = \frac{\frac{F_2}{T_2}(y_{Me,2} + y_{CO2,2} + y_{CO,2}) - \frac{F_2}{T_2} \cdot y_{Me,2}}{\frac{F_2}{T_2}(y_{Me,2} + y_{CO2,2} + y_{CO,2})} \times 100\%$$

$$= \frac{y_{CO2,2} + y_{CO,2}}{y_{Me,2} + y_{CO2,2} + y_{CO,2}} \times 100\%$$

Selectivity:

$$\text{Selectivity}_{CO_2} = \frac{\text{Mole of } CO_2 \text{ generated}}{\text{Mole of } CH_4 \text{ consumed}}$$

$$= \frac{\frac{P_1F_1}{RT_1}\cdot y_{CO2,1}}{\frac{P_0F_0}{RT_0}\cdot y_{Me,0} - \frac{P_1F_1}{RT_1}\cdot y_{Me,1}}$$, and

$$\text{Selectivity}_{CO} = \frac{\frac{P_1F_1}{RT_1}\cdot y_{CO,1}}{\frac{P_0F_0}{RT_0}\cdot y_{Me,0} - \frac{P_1F_1}{RT_1}\cdot y_{Me,1}}$$

Thus,

$$\%\text{Selectivity}_{CO_2/CO} = \frac{\text{Selectivity to } CO_2}{\text{Selectivity to CO} + \text{Selectivity to } CO_2}\times 100\%$$

$$= \frac{y_{CO2,1}}{y_{CO,1} + y_{CO2,1}}\times 100\%$$

From CO and CO_2 balanced around the water trapper, we can rewrite the above equation as,

$$\text{Selectivity} = \frac{y_{CO2,2}}{y_{CO,2} + y_{CO2,2}}\times 100\%$$

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