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## APPENDICES

### Appendix A Experimental Data of Gas Calibration of GC-8A

Condition: Detector Current 80 mA

Temperature	°C	Column	Model
Column	50	P/W HAYESEP DB	HP model 5890 SERIES 2
Detector	100		
TCD-T	100		
Pressure	kPa		
Carrier Pressure (Primary)	600		
Carrier Pressure (1)	550		
TCD-Ref	10		

#### 1. Methane

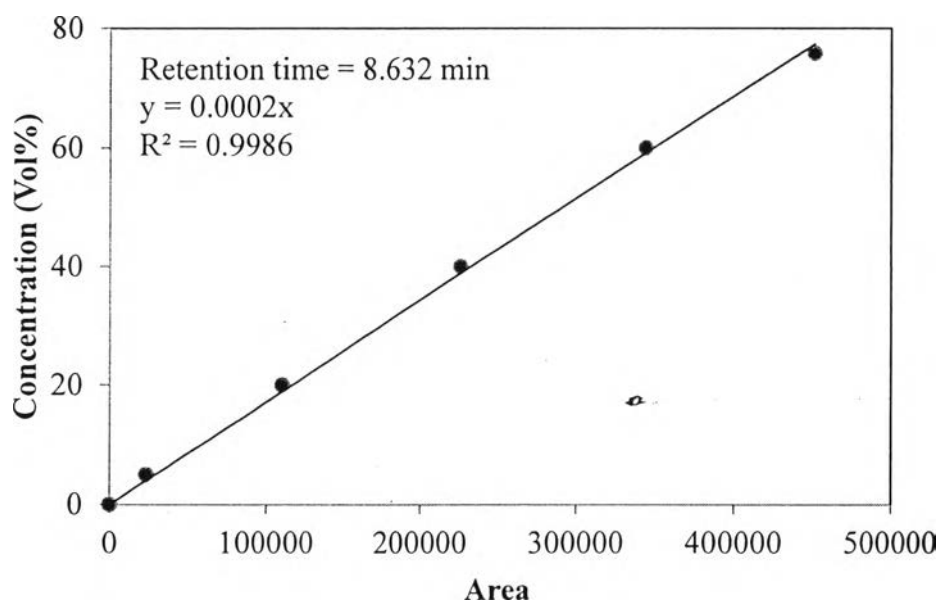


Figure A1 Relationship between area and concentration of methane.

## 2. Carbon dioxide

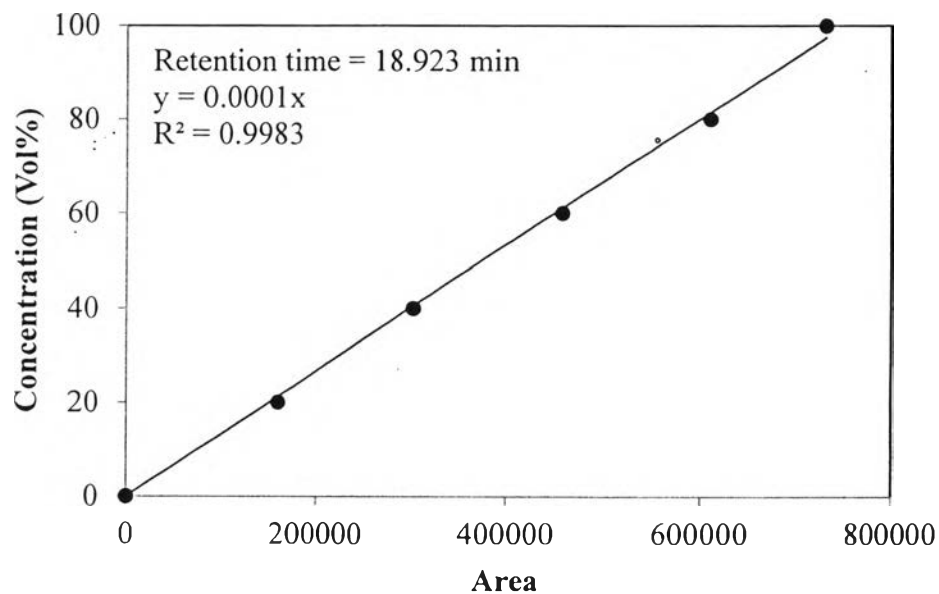


Figure A2 Relationship between area and concentration of carbon dioxide.

## 3. Hydrogen

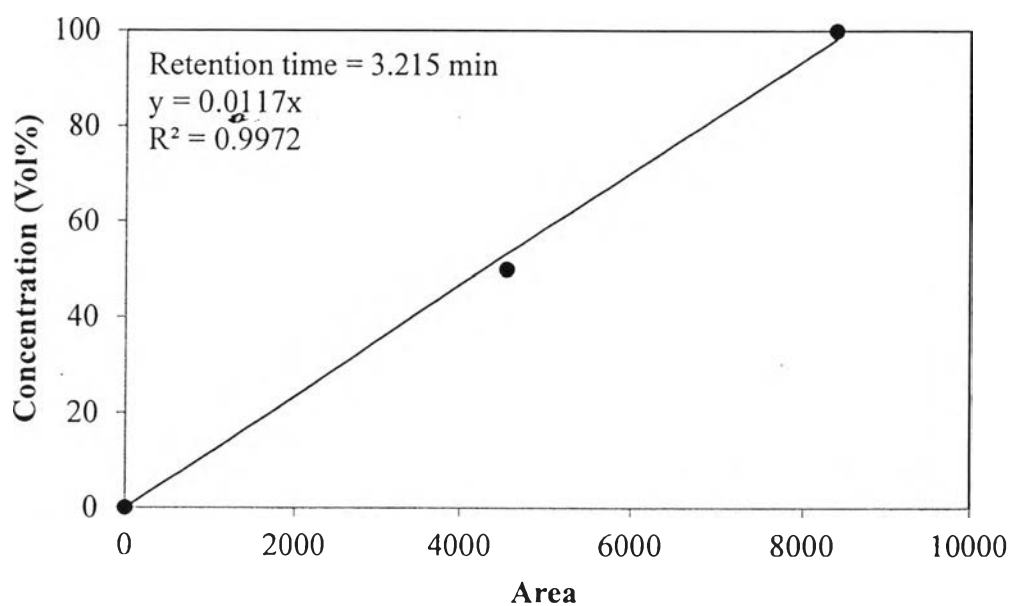


Figure A3 Relationship between area and concentration of hydrogen.



#### 4. Carbon monoxide

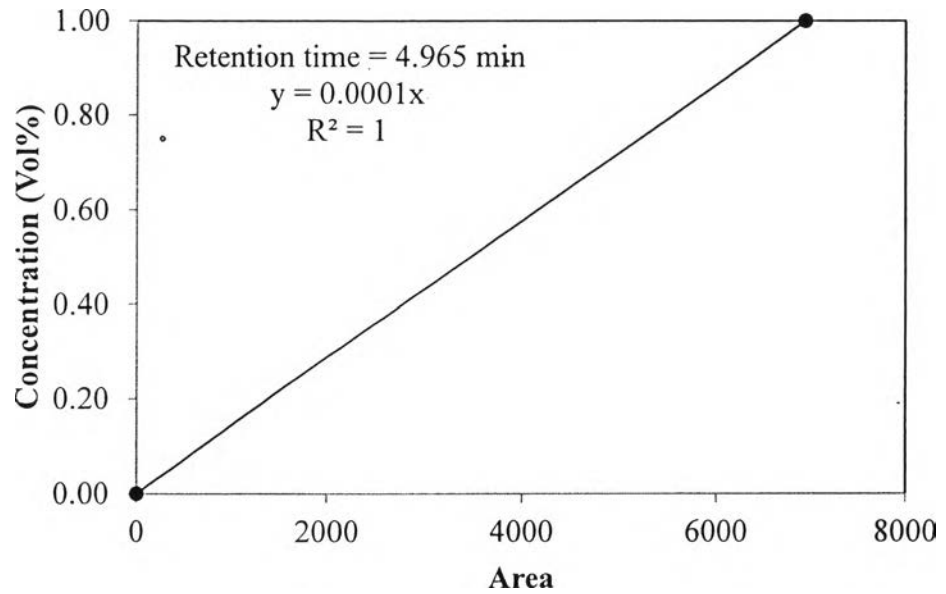


Figure A4 Relationship between area and concentration of carbon monoxide.

#### 5. Oxygen

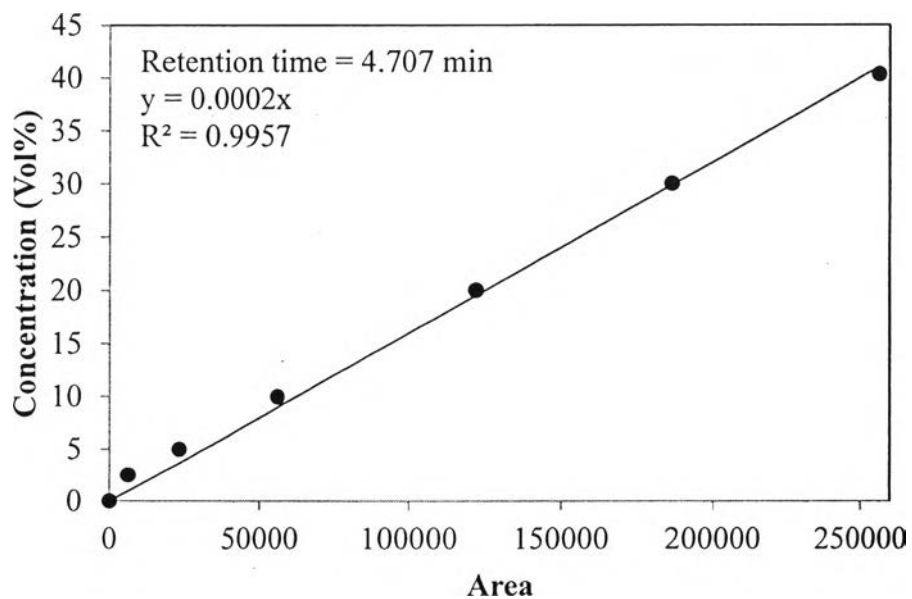


Figure A5 Relationship between area and concentration of oxygen.

## 6. Nitrogen

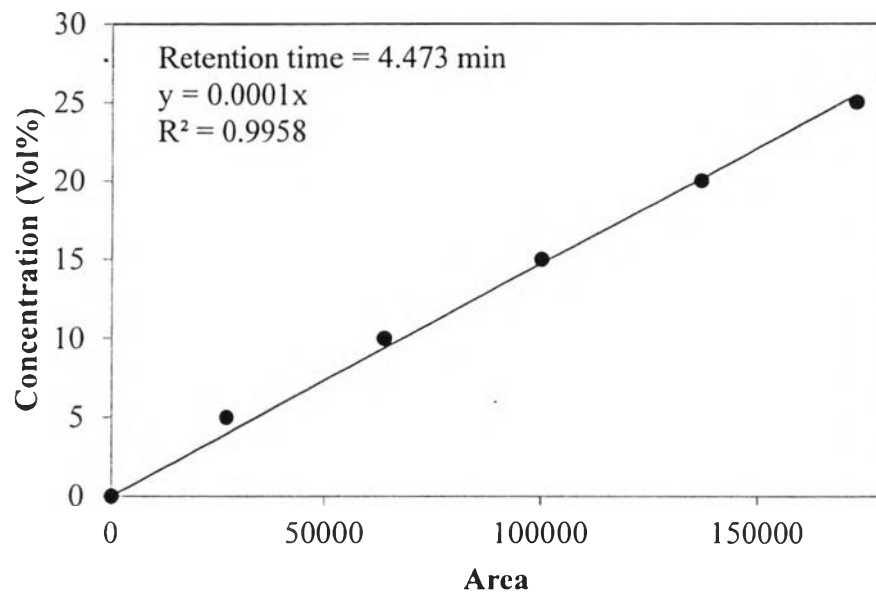


Figure A6 Relationship between area and concentration of nitrogen.

## Appendix B Calibration of Brooks 5850E Mass Flow Controllers

### 1. Methane

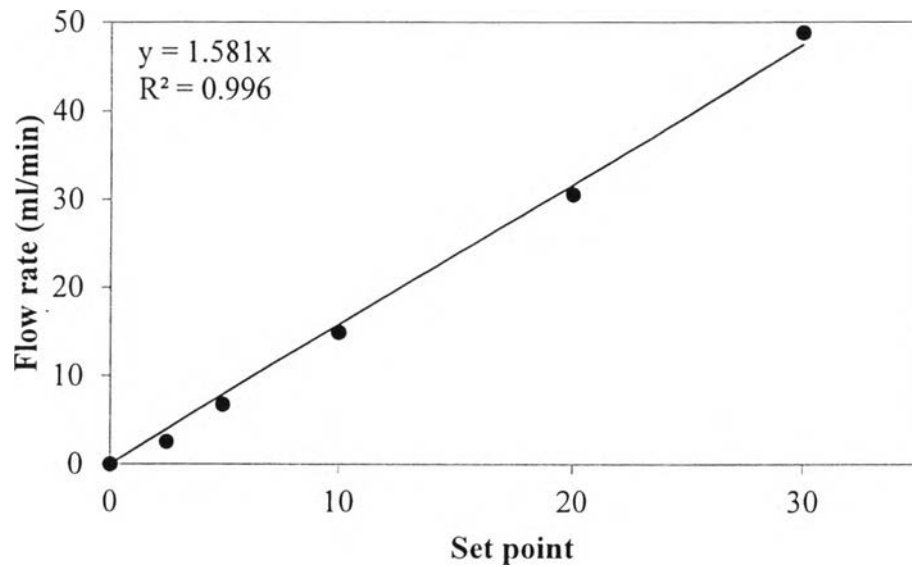


Figure B1 Relationship between SP and flow rate of methane.

### 2. Air

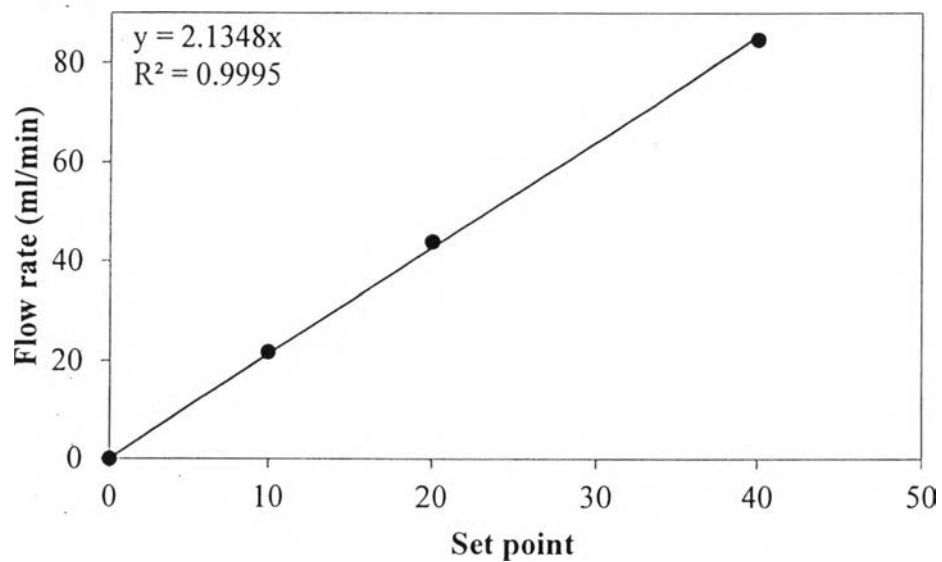


Figure B2 Relationship between SP and flow rate of air.

### 3. Helium

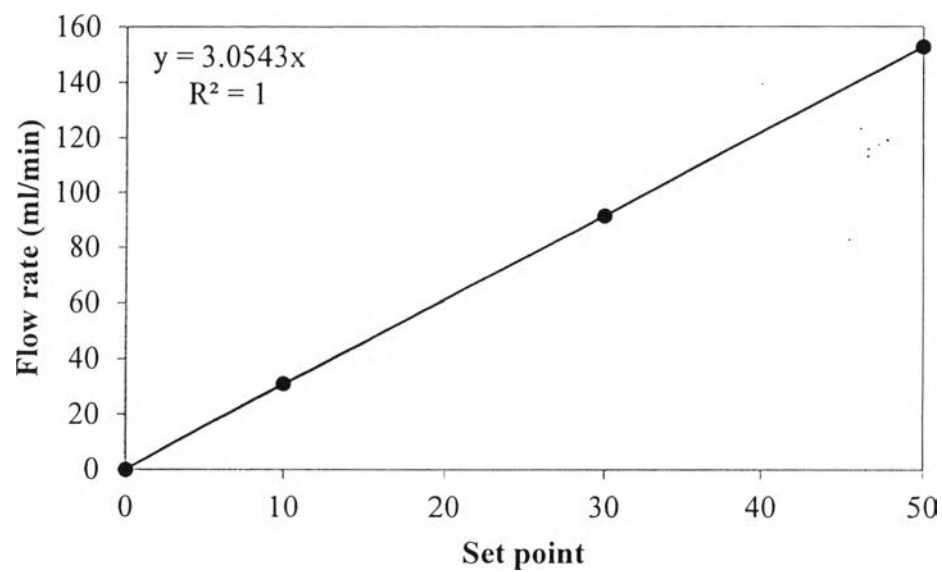


Figure B3 Relationship between SP and flow rate of helium.

### Appendix C Experimental Data of Catalytic Activity Tests for MPO

**Table C1** Catalytic activity test of 5Ni/CZOi catalyst ( $\text{CH}_4/\text{O}_2$  ratio = 2:1)

Temperature (°C)	$X_{\text{CH}_4}$ (%)	$X_{\text{O}_2}$ (%)	$S_{\text{H}_2}$ (%)	$S_{\text{CO}}$ (%)	$Y_{\text{H}_2}$ (%)	$Y_{\text{CO}}$ (%)
401	0.15	1.64	0.00	0.00	0.00	0.00
452	1.21	9.05	0.00	0.00	0.00	0.00
503	6.01	37.26	0.00	0.00	0.00	0.00
559	9.32	52.41	0.00	0.00	0.00	0.00
602	63.77	100.00	87.24	77.67	27.54	35.02
658	64.36	100.00	88.10	82.17	29.05	38.25
705	65.74	100.00	89.50	85.03	31.97	41.95
753	66.79	100.00	90.36	88.48	34.08	43.03
809	67.75	100.00	91.59	90.43	37.10	45.79

**Table C2** Catalytic activity test of 5Ni/CZOp catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 2:1)

Temperature (°C)	X <sub>CH<sub>4</sub></sub> (%)	X <sub>O<sub>2</sub></sub> (%)	S <sub>H<sub>2</sub></sub> (%)	S <sub>CO</sub> (%)	Y <sub>H<sub>2</sub></sub> (%)	Y <sub>CO</sub> (%)
400	2.77	2.42	0.00	0.00	0.00	0.00
450	3.99	10.62	0.00	0.00	0.00	0.00
501	10.72	40.40	0.00	0.00	0.00	0.00
553	17.18	53.19	0.00	0.00	0.00	0.00
603	67.02	100.00	89.94	83.43	33.40	46.01
653	68.55	100.00	89.78	86.09	33.85	46.58
705	69.40	100.00	90.81	88.22	36.32	46.95
753	70.46	100.00	91.29	90.79	37.90	47.02
808	71.41	100.00	91.57	91.33	39.06	47.45

**Table C3** Catalytic activity test of 10Ni/CZOi catalyst ( $\text{CH}_4/\text{O}_2$  ratio = 2:1)

Temperature (°C)	$X_{\text{CH}_4}$ (%)	$X_{\text{O}_2}$ (%)	$S_{\text{H}_2}$ (%)	$S_{\text{CO}}$ (%)	$Y_{\text{H}_2}$ (%)	$Y_{\text{CO}}$ (%)
402	0.31	2.66	0.00	0.00	0.00	0.00
453	2.26	9.91	0.00	0.00	0.00	0.00
504	8.10	38.04	0.00	0.00	0.00	0.00
557	11.00	52.95	0.00	0.00	0.00	0.00
603	65.24	100.00	87.03	76.29	27.87	39.73
657	66.49	100.00	88.64	79.03	30.88	41.18
708	67.83	100.00	90.04	83.69	33.99	43.27
757	68.45	100.00	91.37	85.14	36.99	45.02
811	68.78	100.00	92.76	88.04	40.39	46.79

**Table C4** Catalytic activity test of 10Ni/CZOp catalyst ( $\text{CH}_4/\text{O}_2$  ratio = 2:1)

Temperature (°C)	$X_{\text{CH}_4}$ (%)	$X_{\text{O}_2}$ (%)	$S_{\text{H}_2}$ (%)	$S_{\text{CO}}$ (%)	$Y_{\text{H}_2}$ (%)	$Y_{\text{CO}}$ (%)
400	5.39	3.21	0.00	0.00	0.00	0.00
452	6.24	10.23	0.00	0.00	0.00	0.00
503	12.60	41.97	0.00	0.00	0.00	0.00
554	18.65	53.98	0.00	0.00	0.00	0.00
604	68.33	100.00	89.59	82.66	33.40	46.01
654	69.07	100.00	89.97	86.31	34.48	48.24
706	70.45	100.00	91.16	89.30	37.62	48.94
756	72.55	100.00	91.48	91.28	39.48	50.09
809	72.83	100.00	92.03	91.60	40.94	50.45



**Table C5** Catalytic activity test of 15Ni/CZOi catalyst ( $\text{CH}_4/\text{O}_2$  ratio = 2:1)

Temperature (°C)	$X_{\text{CH}_4}$ (%)	$X_{\text{O}_2}$ (%)	$S_{\text{H}_2}$ (%)	$S_{\text{CO}}$ (%)	$Y_{\text{H}_2}$ (%)	$Y_{\text{CO}}$ (%)
402	66.00	5.63	0.00	0.00	0.00	0.00
455	4.64	18.05	0.00	0.00	0.00	0.00
507	12.51	46.96	0.00	0.00	0.00	0.00
560	18.91	76.17	0.00	0.00	0.00	0.00
608	77.58	100.00	90.34	72.76	39.54	47.93
659	80.75	100.00	91.25	77.32	43.34	53.12
709	84.72	100.00	90.94	78.95	44.67	55.47
760	85.77	100.00	91.64	81.99	47.10	59.17
812	85.83	100.00	90.77	83.60	44.82	57.29

**Table C6** Catalytic activity test of 15Ni/CZOp catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 2:1)

Temperature (°C)	X <sub>CH<sub>4</sub></sub> (%)	X <sub>O<sub>2</sub></sub> (%)	S <sub>H<sub>2</sub></sub> (%)	S <sub>CO</sub> (%)	Y <sub>H<sub>2</sub></sub> (%)	Y <sub>CO</sub> (%)
401	7.53	14.64	0.00	0.00	0.00	0.00
453	8.90	23.20	0.00	0.00	0.00	0.00
504	16.39	48.79	0.00	0.00	0.00	0.00
555	23.13	82.41	0.00	0.00	0.00	0.00
604	80.40	100.00	90.37	75.60	41.05	53.33
655	83.20	100.00	91.41	79.27	45.07	57.62
705	87.03	100.00	92.15	82.16	49.27	57.95
757	87.88	100.00	92.05	85.80	49.44	62.41
809	89.16	100.00	92.00	86.18	50.01	63.22

**Table C7** Catalytic activity test of 25Ni/CZOi catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 2:1)

Temperature (°C)	X <sub>CH<sub>4</sub></sub> (%)	X <sub>O<sub>2</sub></sub> (%)	S <sub>H<sub>2</sub></sub> (%)	S <sub>CO</sub> (%)	Y <sub>H<sub>2</sub></sub> (%)	Y <sub>CO</sub> (%)
405	7.53	4.86	0.00	0.00	0.00	0.00
457	8.90	14.14	0.00	0.00	0.00	0.00
509	16.39	43.29	0.00	0.00	0.00	0.00
562	23.13	64.62	0.00	0.00	0.00	0.00
609	80.40	100.00	87.99	70.83	33.35	41.54
660	83.20	100.00	88.88	74.46	34.99	44.77
710	87.03	100.00	88.29	78.56	36.20	45.47
763	87.88	100.00	88.84	79.73	37.77	46.54
814	89.16	100.00	89.61	82.95	39.59	48.96

**Table C8** Catalytic activity test of 25Ni/CZOp catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 2:1)

Temperature (°C)	X <sub>CH<sub>4</sub></sub> (%)	X <sub>O<sub>2</sub></sub> (%)	S <sub>H<sub>2</sub></sub> (%)	S <sub>CO</sub> (%)	Y <sub>H<sub>2</sub></sub> (%)	Y <sub>CO</sub> (%)
402	7.53	4.55	0.00	0.00	0.00	0.00
454	8.90	15.66	0.00	0.00	0.00	0.00
505	16.39	36.12	0.00	0.00	0.00	0.00
556	23.13	67.66	0.00	0.00	0.00	0.00
605	80.40	100.00	89.47	77.81	38.34	50.33
656	83.20	100.00	89.21	79.30	38.86	51.06
705	87.03	100.00	89.56	81.16	41.31	54.11
758	87.88	100.00	90.71	85.00	44.68	57.57
808	89.16	100.00	90.66	87.40	45.43	58.36

**Table C9** Catalytic activity test of 15Ni/CZOi catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 4:1)

Temperature (°C)	X <sub>CH<sub>4</sub></sub> (%)	X <sub>O<sub>2</sub></sub> (%)	S <sub>H<sub>2</sub></sub> (%)	S <sub>CO</sub> (%)	Y <sub>H<sub>2</sub></sub> (%)	Y <sub>CO</sub> (%)
405	0.32	5.11	0.00	0.00	0.00	0.00
455	2.14	12.50	0.00	0.00	0.00	0.00
507	6.15	35.36	0.00	0.00	0.00	0.00
556	10.63	63.02	0.00	0.00	0.00	0.00
606	53.37	100.00	83.57	83.57	19.27	25.61
655	57.22	100.00	88.02	88.02	25.72	30.45
707	61.47	100.00	89.07	89.07	29.20	34.44
757	66.17	100.00	88.68	88.68	30.79	37.01
808	67.49	100.00	87.28	87.28	29.20	36.41

**Table C10** Catalytic activity test of 15Ni/CZOp catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 4:1)

Temperature (°C)	X <sub>CH4</sub> (%)	X <sub>O2</sub> (%)	S <sub>H2</sub> (%)	S <sub>CO</sub> (%)	Y <sub>H2</sub> (%)	Y <sub>CO</sub> (%)
401	2.48	12.44	0.00	0.00	0.00	0.00
453	6.63	21.19	0.00	0.00	0.00	0.00
504	13.57	44.70	0.00	0.00	0.00	0.00
555	19.57	62.44	0.00	0.00	0.00	0.00
604	69.01	100.00	86.35	90.61	28.49	33.05
655	70.99	100.00	89.23	94.82	34.04	37.95
705	73.24	100.00	89.27	94.23	35.18	39.06
757	75.12	100.00	88.67	93.81	34.93	39.80
809	75.87	100.00	88.21	93.45	34.44	40.41

### Appendix D Experimental Data of Stability Tests for MPO

**Table D1** Stability test of 15Ni/CZOi catalyst ( $\text{CH}_4/\text{O}_2$  ratio = 2:1)

Time (hr)	$X_{\text{CH}_4}$ (%)	$X_{\text{O}_2}$ (%)	$S_{\text{H}_2}$ (%)	$S_{\text{CO}}$ (%)
1	85.84	100.00	92.05	81.16
2	85.60	100.00	92.14	81.24
3	85.30	100.00	91.89	81.10
4	85.84	100.00	91.86	81.31
5	85.86	100.00	92.08	80.93
6	85.58	100.00	91.58	80.73

**Table D2** Stability test of 15Ni/CZO<sub>p</sub> catalyst ( $\text{CH}_4/\text{O}_2$  ratio = 2:1)

Time (hr)	$X_{\text{CH}_4}$ (%)	$X_{\text{O}_2}$ (%)	$S_{\text{H}_2}$ (%)	$S_{\text{CO}}$ (%)
1	87.67	100.00	92.16	85.81
2	87.52	100.00	92.18	86.72
3	87.80	100.00	92.22	86.07
4	87.81	100.00	92.16	85.98
5	87.76	100.00	92.07	85.49
6	87.68	100.00	92.18	84.48

**Table D3** Stability test of 15Ni/CZOi catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 4:1)

Time (hr)	X <sub>CH<sub>4</sub></sub> (%)	X <sub>O<sub>2</sub></sub> (%)	S <sub>H<sub>2</sub></sub> (%)	S <sub>CO</sub> (%)
1	64.96	100.00	89.67	89.02
2	64.94	100.00	89.43	88.42
3	64.35	100.00	89.36	87.56
4	64.10	100.00	88.63	86.85
5	64.32	100.00	89.67	85.03
6	62.77	100.00	89.43	84.17

**Table D4** Stability test of 15Ni/CZOp catalyst (CH<sub>4</sub>/O<sub>2</sub> ratio = 4:1)

Time (hr)	X <sub>CH<sub>4</sub></sub> (%)	X <sub>O<sub>2</sub></sub> (%)	S <sub>H<sub>2</sub></sub> (%)	S <sub>CO</sub> (%)
1	75.12	100.00	89.63	93.81
2	75.12	100.00	89.58	93.78
3	75.12	100.00	89.49	93.62
4	74.87	100.00	89.51	91.46
5	74.83	100.00	89.31	89.80
6	74.78	100.00	89.30	88.97



## Appendix E Calculation of Metal Surface Area

The calculation of the metal surface area is based on the following relation:

$$MSS = (V_m N_a) / (S_f S_d)$$

MSS = metal surface area ( $m^2/g$  of sample)

$V_m$  = gas adsorbed at monolayer (moles/gram of sample)

$N_a$  = Avogadro number (molecule/mole of gas) =  $6.02 \times 10^{23}$  molecule/mole

$S_f$  = stoichiometric factor of the reaction (number of molecule of adsorbate/surface metal atom) = 0.5

$S_d$  = Metal surface density (number of metal atoms/ $m^2$ ) =  $0.154 \times 10^{20}$  atoms/ $m^2$

For example

Metal Surface Area of 5Ni/CZOi

$$\begin{aligned} MSS &= [(3.63223 \times 10^{-5}) \times (6.02 \times 10^{23})] / [0.5 \times (0.154 \times 10^{20})] \\ &= 2.84 \text{ m}^2/\text{g} \end{aligned}$$

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