

REFERENCES

- Anderson, J.A., and Garcia, M.F. (2005). Supported Metals in Catalysis. London: Imperial College Press.
- Beyer, H.K. (2002). Molecular Sieves Vol.3. Berlin: Springer-Verlag.
- Canizares, P., Lucas, A., Dorado, F., Duran, A., and Asencio, I. (1997). Characterization of Ni and Pd supported on H-mordenite catalysts: Influence of the metal loading method. Applied Catalysis A: General, 169, 137-150.
- Chankham, O. (2007). Hydrogen production via steam reforming of methane over Ni supported-NaY zeolite catalyst. M.S. Thesis, The Petroleum and Petrochemical College, Chulalongkorn University, Bangkok, Thailand.
- Chan, J.-S., Park, S.-E., and Chon, H. (1996). Catalytic activity and coke resistance in the CO₂ reforming of methane to synthesis gas over zeolite-supported Ni catalysts: Applied Catalysis A: General, 145, 111-124.
- Chen, Y.-G., Tomishige, K., Yokoyama, K., and Fujimoto, K. (1999). Catalytic performance and catalyst structure of Ni-Magnesia catalysts for CO₂ reforming of methane. Journal of Catalysis, 184, 479-490.
- Comas, J., Dieuzeide, M.L., Baronetti, G., Laborde, M., and Amadeo, N. (2006). Methane steam reforming and ethanol steam reforming using a Ni(II)-Al(III) catalyst prepared from lamellar double hydroxides. Chemical Engineering Journal, 118, 11-15.
- Craciun, R., Daniell, W., and Knozinger, H. (2002). The effect of CeO₂ structure on the activity of supported Pd catalysts used for methane steam reforming. Applied Catalysis A: General, 230, 153-168.
- Crisafulli, C., Scire, S., Minico, S., Solarino, L. (2002). Ni-Ru bimetallic catalysts for the CO₂ reforming of methane. Applied Catalysis A: General, 225, 1-9.
- Damyanova, S., Perez, C.A., Schmal, M., and Bueno, J.M.C. (2002). Characterization of ceria-coated alumina carrier. Applied Catalysis A: General, 234, 271-282.
- Dong, W.-S., Roh, H.-S., Jun, K.-W., Park, S.-E., and Oh, Y.-S. (2002). Methane reforming over Ni/Ce-ZrO₂ catalysts: effect of Ni content. Applied Catalysis A: General, 226, 63-72.

- Feio, L.S.F., Hori, C.E., Damyanova, S., Noronha, F.B., Cassinelli, W.H., Marques, C.M.P., and Bueno, J.M.C. (2007). The effect of ceria content on the properties of Pd/CeO₂/Al₂O₃ catalysts for steam reforming of methane. Applied Catalysis A: General, 316, 107-116.
- Gates, B.C. (1992). Catalytic Chemistry. New York: John Wiley & Sons, Inc.
- Gordeeva, L.G., Aristov, Y.I., Moroz, E.M., Rudina, N.A., Zaikovskii, V.I., Tanashev, Y.Y., and Parmon, V.N. (1994). Preparation and study of porous uranium oxides as supports for new catalysts of steam reforming of methane. Journal of Nuclear Materials, 218, 202-209.
- Goud, S.K., Whittenberger, W.A., Chattopadhyay, S., and Abraham, M.A. (2007). Steam reforming of n-hexadecane using a Pd/ZrO₂ catalyst : kinetics of catalyst deactivation. International Journal of Hydrogen Energy, 32, 2868-2874.
- Hegarty, M.E.S., O' Conner, A.M., and Ross, J.R.H. (1998). Syngas production from natural gas using ZrO₂-supported metals. Catalysis Today, 42, 225-232.
- Kusakabe, K., Sotowa, K.I., Eda, T., and Iwamoto, Y. (2004). Methane steam reforming over Ce-ZrO₂-supported noble metal catalysts at low temperature. Fuel Processing Technology, 86, 319-326.
- Levent, M., Gunn, D.J., Ali El-Bousiffi, M. (2002). Production of hydrogen-rich gases from steam reforming of methane in an automatic catalytic microreactor. International Journal of Hydrogen Energy, 1-15.
- Le Page, J.-F., Cosyns, J., Courty, P., Freund, E., Franck, J.-P., Jacquin, Y., Juguin, B., Marcilly, C., Martino, G., Miquel, J., Montarnal, R., Sugier, A., and Van Landegham, H. (1978). Applied Heterogeneous Catalysis. Paris: Editions Technip.
- Laosiripojana, N., Sutthisripok, W., Assabumrungrat, S. (2005). Synthesis gas production from dry reforming of methane over CeO₂ doped Ni/Al₂O₃ : Influence of the doping ceria on the resistance toward carbon formation. Chemical Engineering Journal, 112, 13-22.
- Matar, S., and Hatch, L. (1994). Chemistry of petrochemical processes. Texas: Gulf Publishing Company.

- Matsumura, Y., and Nakamori, T. (2004). Steam reforming of methane over Ni catalysts at low reaction temperature. Applied Catalyst A: General, 258, 107-114.
- Oh, Y.-S., Roh, H.-S., Jun, K.-W., and Baek, Y.-S. (2003). A highly active catalyst, Ni/Ce-ZrO₂/θ-Al₂O₃, for on-site H₂ generation by steam methane reforming: pretreatment effect. International Journal of Hydrogen Energy, 1-6.
- Querini, C.A. (2004). Catalysis, volume 17. The Royal Society of Chemistry.
- Quincoces, C.E., Vargas, S.P., Grange, P., Gonzalez, M.G. (2002). Role of Mo in CO₂ reforming of CH₄ over Mo promoted Ni/Al₂O₃ catalysts. Materials Letters, 56, 698-704.
- Roh, H.-S., Jun, K.-W., and Park, S.-E. (2003). Methane-reforming reactions over NiO/Ce-ZrO₂/θ-Al₂O₃ catalysts. Applied Catalysis A: General, 251, 275-283.
- Rostrup-nielsen. (1984). Catalysis science and technology. New York: Slinkger.
- Satterfield, C.N. (1991). Heterogeneous Catalysis in Industrial Practice. New York: McGraw-Hill, Inc.
- Senathipbodee, N. (2004). Hydrogen production from steam reforming of methane using Ni-supported KL zeolite catalysts. M.S. Thesis, The Petroleum and Petrochemical College, Chulalongkorn University, Bangkok, Thailand.
- Sturzenegger, M., D'Souza, L., Struis, R.P.W.J., and Stucki, S. (2006). Oxygen transfer and catalytic properties of Ni iron oxides for steam reforming of methane. Fuel, 85, 1599-1602.
- Tosheva, L. (1999). Zeolite macrostructure. Licentiate Thesis, Chemical technology department of chemical and metallurgical engineering, Lulea University, Sweden.
- Tosiri, S. (2006). Hydrogen production from CO₂ reforming of methane using Ni-supported KL zeolite catalysts. M.S. Thesis, The Petroleum and Petrochemical College, Chulalongkorn University, Bangkok, Thailand.
- Viswanathan, B., Sivasanker, S., and Ramaswamy, A.V. (2002). Catalysis Principles and Applications. New Delhi: Narosa Publishing House.

Wang, S., and Lu, G.Q.M. (1997). CO₂ reforming of methane on Ni catalysts: effects of the support phase and preparation technique. Applied Catalysis B: Environmental, 16, 269-277.

Yamazaki, O., Tomishige, K., and Fujimoto, K. (1996). Development of highly stable Ni catalyst for methane-steam reaction under low steam to carbon ratio. Applied Catalysis A: General, 136, 49-56.

http://ec.europa.eu/research/rtdinfo/42/01/article_1315_en.html

<http://www.3dchem.com/molecules.asp>

<http://www.uni-leipzig.de/~pdfhome/zsm1.html>

APPENDICES

Appendix A Calculations

1. Catalyst Preparation

1.1 Amount of Ni loading

Example Prepared 1 g of 3 wt% Ni/ZSM-5 catalyst;

- Amount of Ni (MW = 58.70 g/mole)

$$\begin{aligned}\text{Ni} &= 1 \cdot (3/100) \text{ g} \\ &= 0.03 \text{ g}\end{aligned}$$

- Amount of Ni (NO₃)₂·6H₂O (MW = 290.81 g/mole)

$$\begin{aligned}\text{Ni (NO}_3)_2 \cdot 6\text{H}_2\text{O} &= 0.03 \cdot (290.81/58.70) \text{ g} \\ &= 0.1486 \text{ g}\end{aligned}$$

- Amount of ZSM-5 Zeolite

$$\text{ZSM-5 Zeolite} = 1 - 0.03 = 0.97 \text{ g}$$

1.2 Amount of CeO₂ loading

Example Prepared 1g of 3%Ce-11% Ni/ZSM-5 catalyst;

- Amount of Ni (MW = 58.70 g/mole)

$$\begin{aligned}\text{Ni} &= 1 \cdot (11/100) \text{ g} \\ &= 0.11 \text{ g}\end{aligned}$$

- Amount of Ni (NO₃)₂·6H₂O (MW = 290.81 g/mole)

$$\begin{aligned}\text{Ni (NO}_3)_2 \cdot 6\text{H}_2\text{O} &= 0.11 \cdot (290.81/58.70) \text{ g} \\ &= 0.5450 \text{ g}\end{aligned}$$

- Amount of Ce (MW = 140.12 g/mole)

$$\begin{aligned}\text{Ce} &= 1 \cdot (3/100) \text{ g} \\ &= 0.03 \text{ g}\end{aligned}$$

- Amount of Ce(NO₃)₃·6H₂O (MW = 434.23 g/mole)

$$\begin{aligned}\text{Ce(NO}_3)_3 \cdot 6\text{H}_2\text{O} &= 0.03 \cdot (434.23/140.12) \text{ g} \\ &= 0.0930 \text{ g}\end{aligned}$$

- Amount of ZSM-5 Zeolite

$$\text{ZSM-5 Zeolite} = 1 - 0.11 - 0.03 = 0.86 \text{ g}$$

2. Steam- to-Methane Ratio for Feed

For mixture system;

Assume: 1:1 H₂O: CH₄ molar ratio;

Where Total flow = 100 ml/min

CH₄ flow = 20 ml/min

Steam flow = 20 ml/min

He balance = 60 ml/min

Finding the flow rate of water in feed;

From thermodynamic steam table;

At 160°C, V_g = 0.3068 m³/kg (D_g = 0.00326 g/cm³)

At 25°C, V_f = 0.001003 m³/kg (D_f = 0.997 g/cm³)

At evaporator (water feed is in the steam phase)

$$\begin{aligned} \text{Then, } M_g &= D_g * V_g \\ &= 0.00326 \text{ g/cm}^3 * 20 \text{ ml} \\ &= 0.0652 \text{ g} \end{aligned}$$

Thus, water in liquid phase as a feed is

$$\begin{aligned} V_l &= M_l / D_l \\ &= 0.0652 \text{ g} / 0.997 \text{ g/cm}^3 \\ &= 0.0654 \text{ ml/min.} = 3.9 \text{ ml/hr.} \end{aligned}$$

3. Metal Crystallite Size from XRD

The thickness of crystallite (L) calculated from Scherrer Equation;

$$L_{\text{hkl}} = k\lambda / (\beta \cos\theta_0)$$

Where λ = the x-ray wavelength

B = the peak width (expressed in radian)

θ_0 = the angle between the beam and the normal on the reflecting plane
(expressed in radian)

k = a constant (or shape factor) (often take as 1)

4. Average Particle Size from TEM

The average particle size (d_s) is calculated on the basis of counting the diameter of numerous particles, according to the equation;

$$d_s = \frac{\sum n_i d_i^3}{\sum n_i d_i^2}$$

Where n_i = number of particles in each size range

d_i = diameter of particle in each size range

Moreover, the particle size distribution is also calculated from:

$$\frac{\text{Number of particles in range}}{\text{Total number of particles}} \times 100\%$$

Appendix B Effect of Ni loading on reduced catalysts for XRD

These XRD patterns show that there are only Ni peaks for the reduced catalysts and the Ni peak intensity is increased with the increasing of Ni loading. It confirms the complete reduction of the catalysts before the reforming reaction was started.

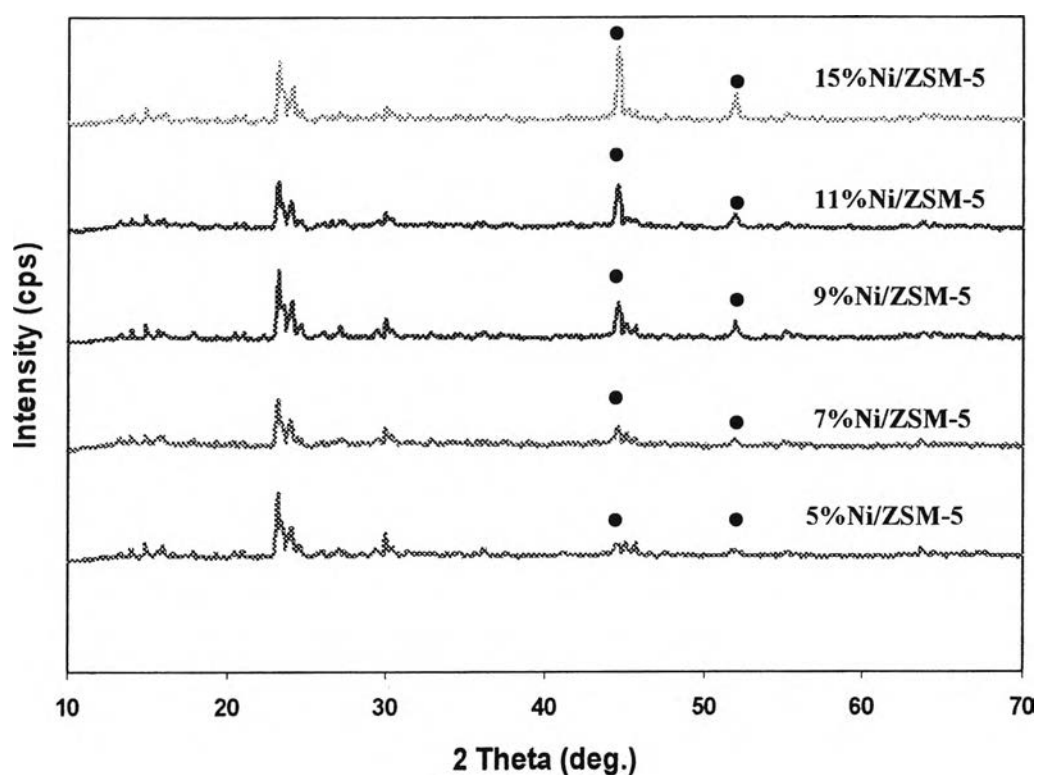


Figure B1 XRD patterns of the reduced Ni/ZSM-5 catalysts with various loadings of Ni, which operated on steam reforming reaction at 700°C and atmospheric pressure for 5 hours; (●), Ni metal phase.

Appendix C Particle size distribution from TEM

1. Effect of Ni loading

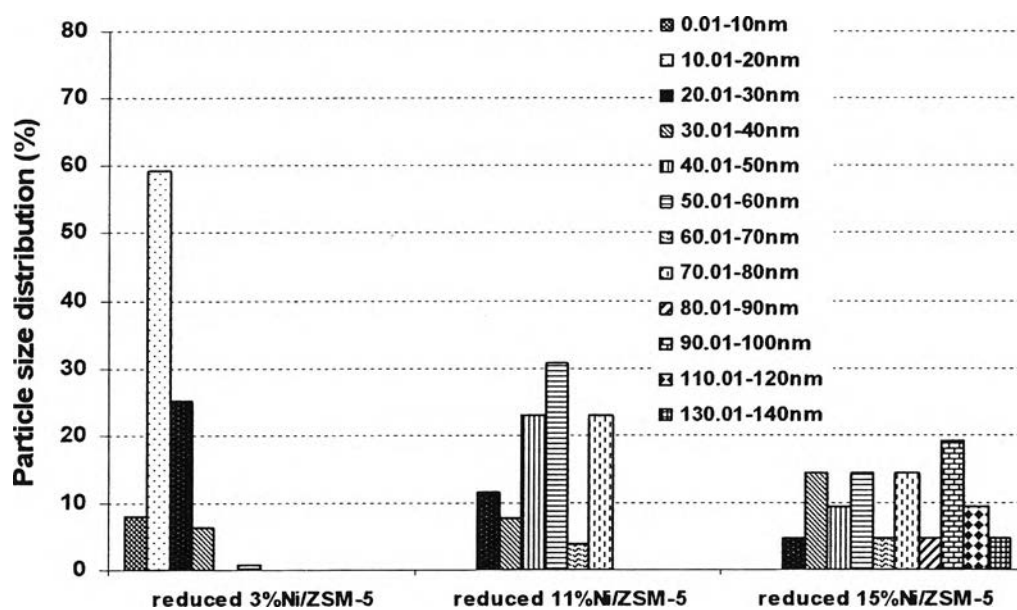


Figure C1 Particle size distribution of the reduced Ni/ZSM-5 catalysts with various loadings of Ni which were calculated from TEM results.

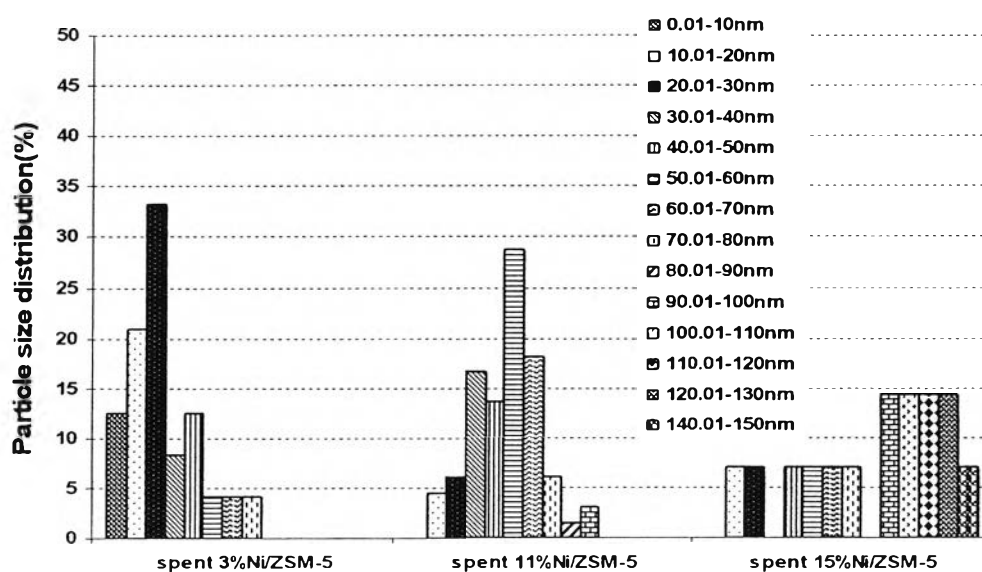


Figure C2 Particle size distribution of the spent Ni/ZSM-5 catalysts with various loadings of Ni which were calculated from TEM results.

2. Effect of CeO₂ addition

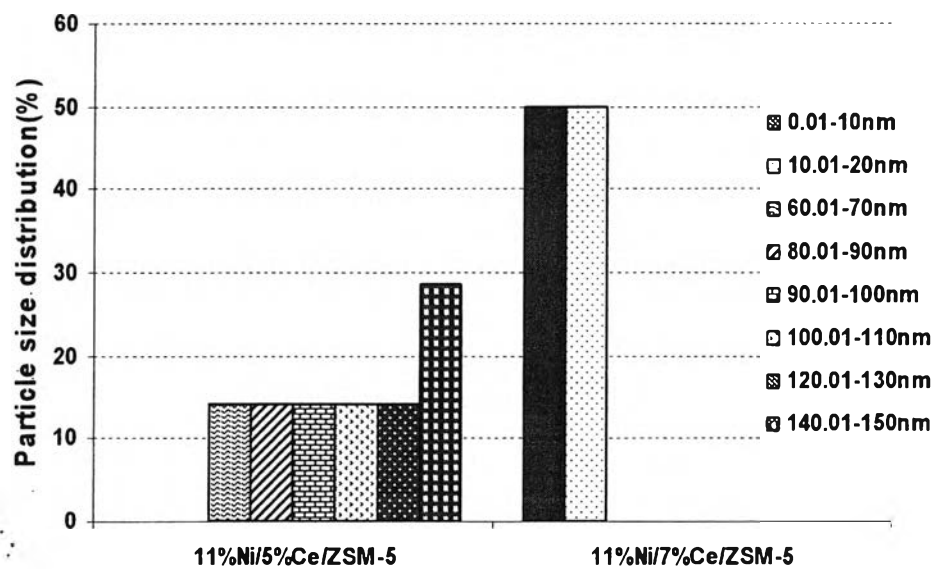


Figure C3 Particle size distribution of the spent Ni/ZSM-5 catalysts with various loadings of CeO₂ which were calculated from TEM results.

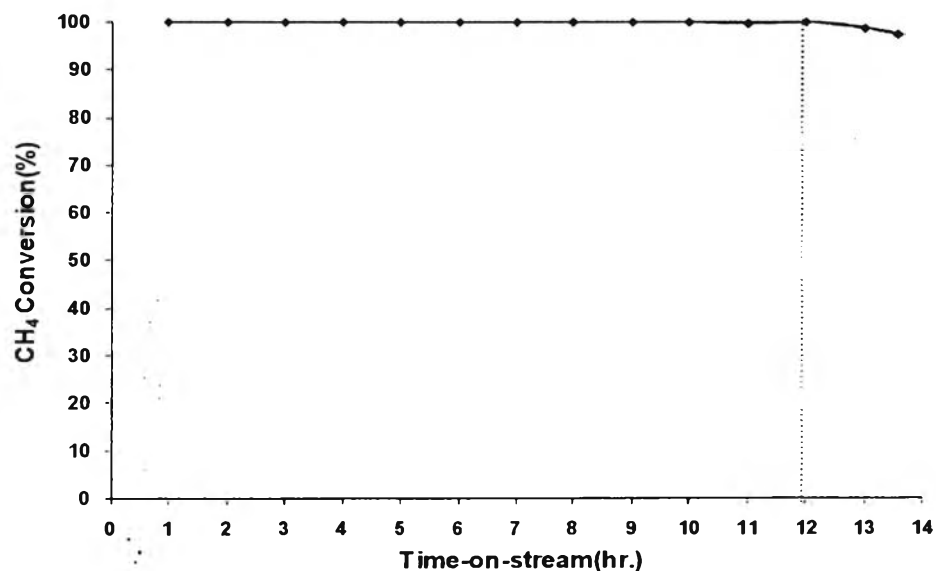
Appendix D Stability test of the 11%Ni/5%Ce/ZSM-5 catalyst

Figure D1 CH₄ conversion of the 11%Ni/5%Ce/ZSM-5 catalyst operated on the reforming reaction at 700°C and a H₂O/CH₄ ratio of 0.8 for the stability test.

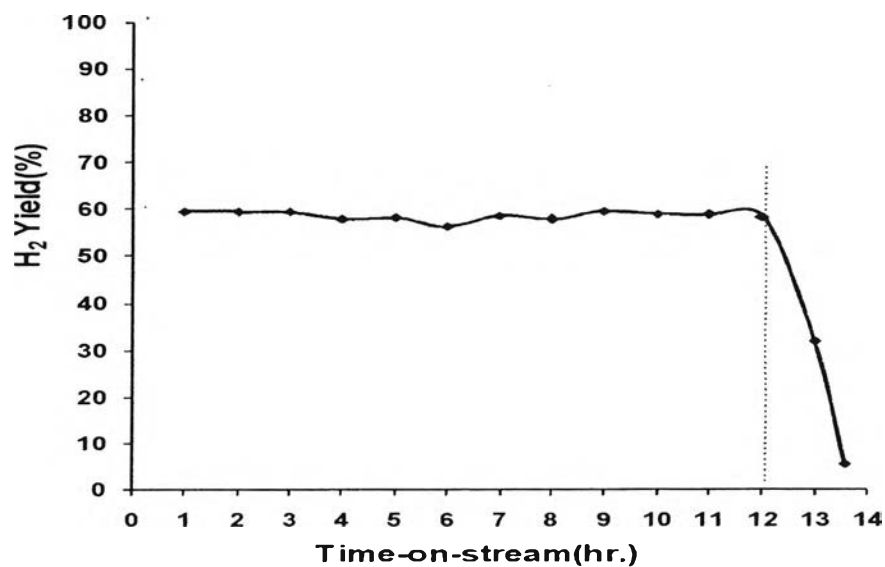


Figure D2 H₂ yield of the 11%Ni/5%Ce/ZSM-5 catalyst operated on the reforming reaction at 700°C and a H₂O/CH₄ ratio of 0.8 for the stability test.

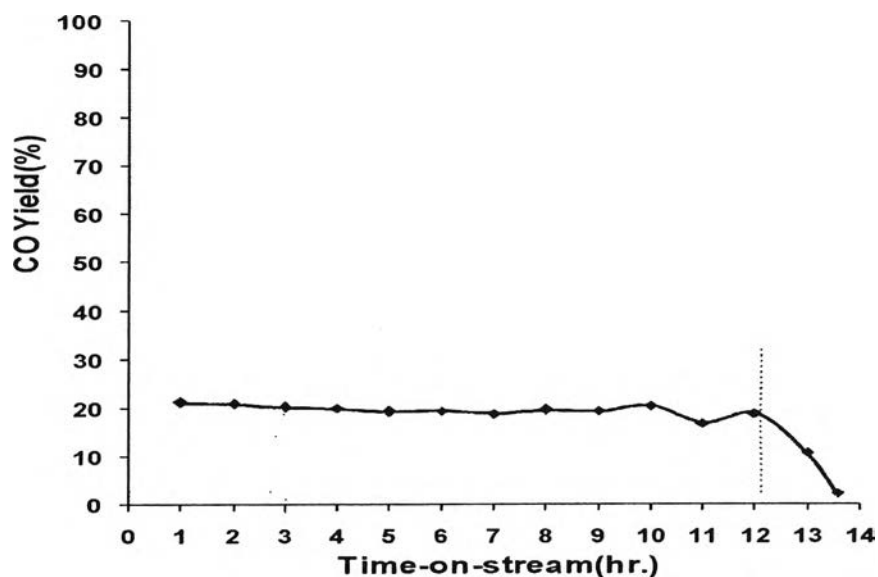


Figure D3 CO yield of the 11%Ni/5%Ce/ZSM-5 catalyst operated on the reforming reaction at 700°C and a H₂O/CH₄ ratio of 0.8 for the stability test.

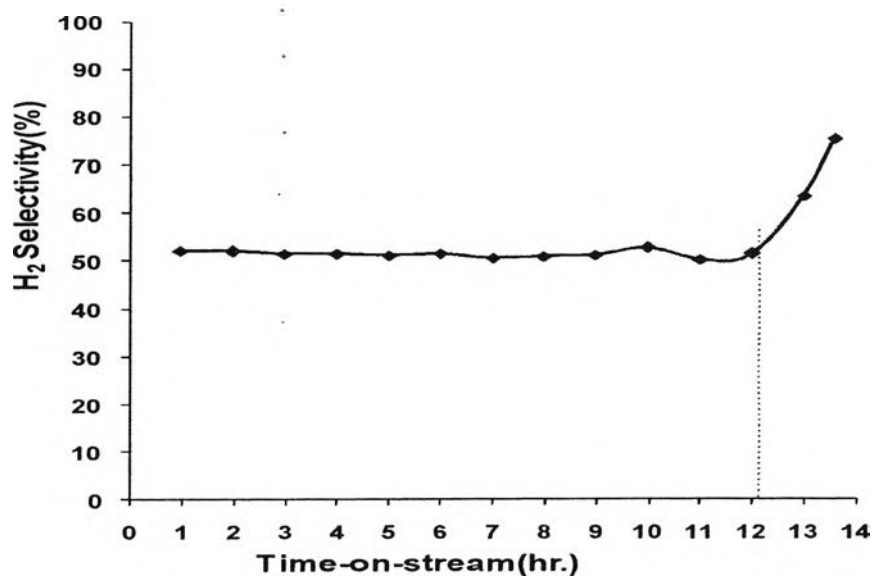


Figure D4 H₂ Selectivity of the 11%Ni/5%Ce/ZSM-5 catalyst operated on the reforming reaction at 700°C and a H₂O/CH₄ ratio of 0.8 for the stability test.

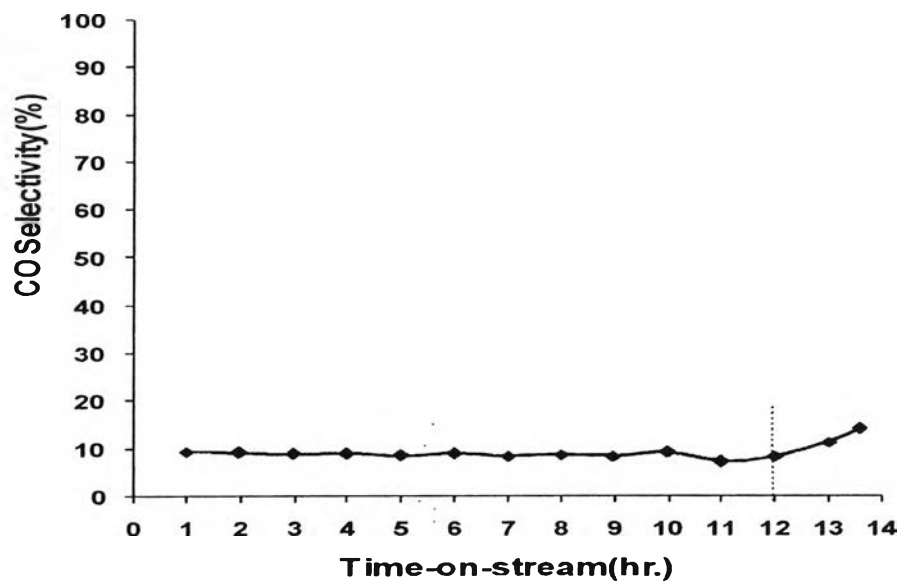


Figure D5 CO selectivity of the 11%Ni/5%Ce/ZSM-5 catalyst operated on the reforming reaction at 700°C and a H₂O/CH₄ ratio of 0.8 for the stability test.

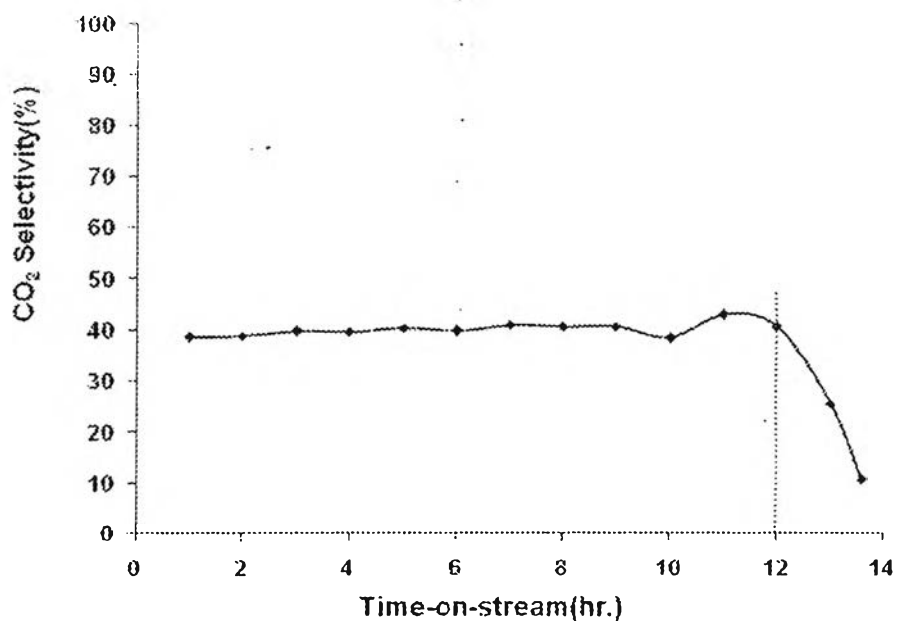


Figure D6 CO₂ selectivity of the 11%Ni/5%Ce/ZSM-5 catalyst operated on the reforming reaction at 700°C and a H₂O/CH₄ ratio of 0.8 for the stability test.

Appendix E CO₂ selectivity of the catalysts in all effect studies

1. Effect of Ni loading

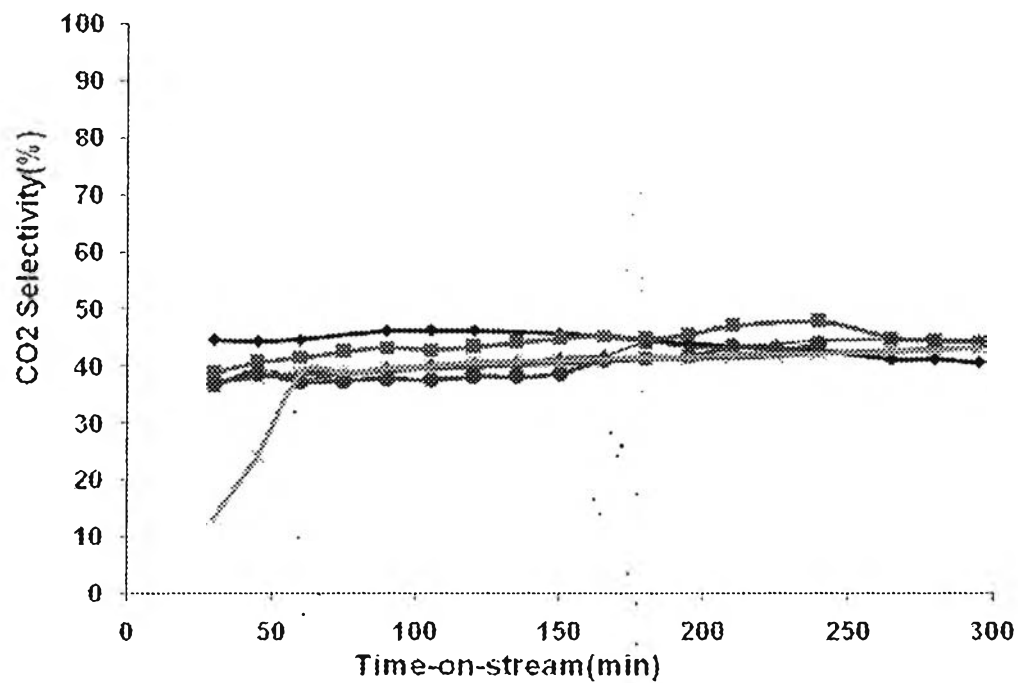


Figure E1 CO₂ selectivity of the Ni/ZSM-5 catalysts with various loadings of Ni, which operated on steam reforming reaction at 700°C and atmospheric pressure for 5 hours; (-◆-), 3%Ni/ZSM-5; (-■-), 5%Ni/ZSM-5; (-▲-), 7%Ni/ZSM-5; (-×-), 9%Ni/ZSM-5; (-●-), 11%Ni/ZSM-5; (-ж-), 15%Ni/ZSM-5.

2. Effect of H₂O/CH₄ ratio

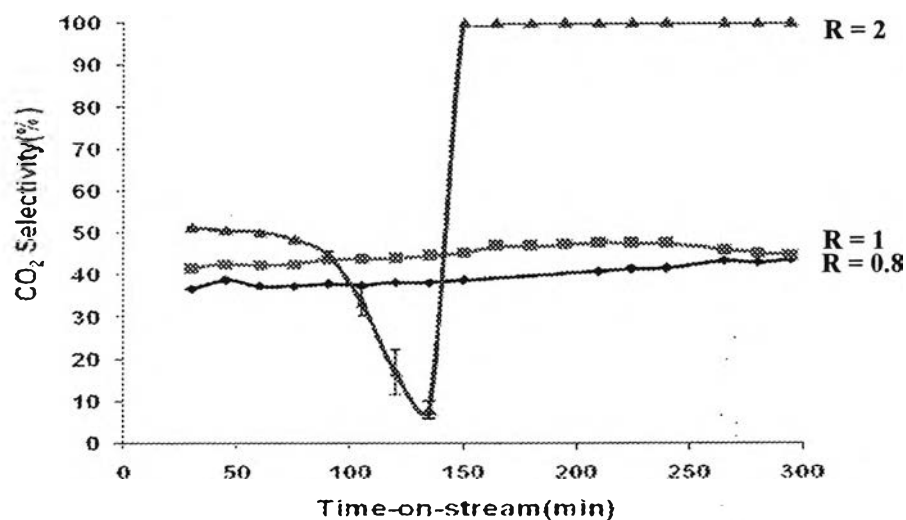


Figure E2 CO₂ selectivity of the 11%Ni/ZSM-5 catalysts with different H₂O/CH₄ ratios, which operated on steam reforming reaction at 700°C and atmospheric pressure for 5 hours.

3. Effect of CeO₂ loading

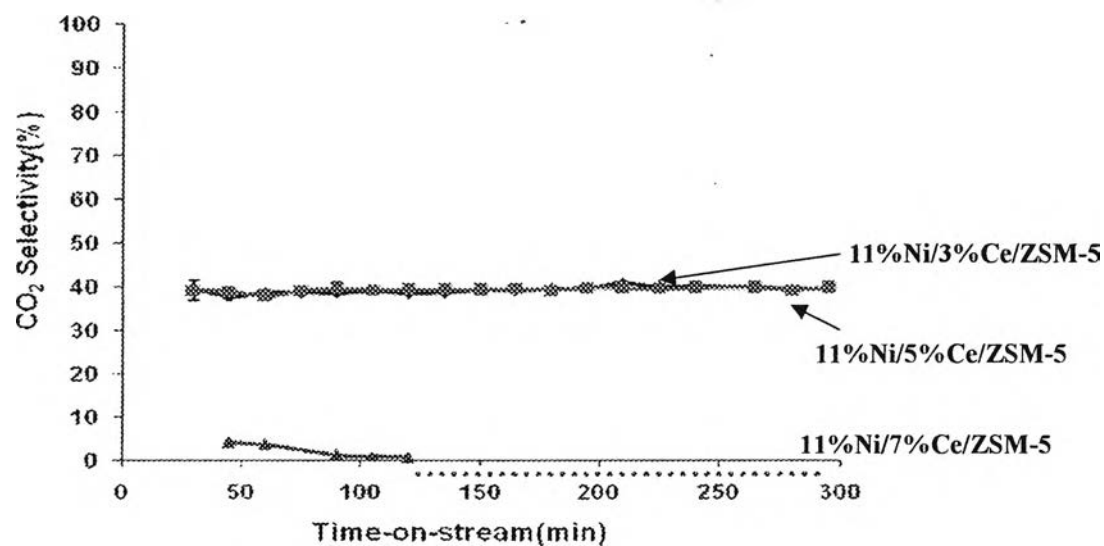


Figure E3 CO₂ selectivity of the 11%Ni/ZSM-5 catalysts with various CeO₂ loadings, which operated on steam reforming reaction at 700°C, H₂O/CH₄ ratio of 0.8, and atmospheric pressure for 5 hours.

Appendix F Experimental data

Table F1 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

3%Ni loading

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
15	H ₂	0.5973	0.0075	50.37	4.43	45.21	0.0123	43.91	3.86	87.17
	CO ₂	0.5360	0.0067							
	CO	0.0525	0.0007							
	CH ₄	0.1256	0.0016							
30	H ₂	1.0877	0.0136	50.79	4.68	44.53	0.0245	42.62	3.92	83.91
	CO ₂	0.9537	0.0119							
	CO	0.1002	0.0013							
	CH ₄	0.3151	0.0039							
45	H ₂	1.4391	0.0180	50.87	4.82	44.31	0.0368	42.18	4.00	82.90
	CO ₂	1.2533	0.0157							
	CO	0.1363	0.0017							
	CH ₄	0.5022	0.0063							
60	H ₂	1.6617	0.0208	50.71	4.64	44.65	0.0490	41.48	3.79	81.81
	CO ₂	1.4632	0.0183							
	CO	0.1519	0.0019							
	CH ₄	0.7126	0.0089							
90	H ₂	2.0331	0.0255	49.73	4.10	46.17	0.0736	38.99	3.21	78.40
	CO ₂	1.8875	0.0236							
	CO	0.1676	0.0021							
	CH ₄	1.2692	0.0159							
105	H ₂	2.0510	0.0257	49.88	3.92	46.20	0.0858	38.83	3.05	77.86
	CO ₂	1.8998	0.0238							
	CO	0.1612	0.0020							
	CH ₄	1.5178	0.0190							
120	H ₂	2.1566	0.0270	50.06	3.80	46.13	0.0981	38.21	2.90	76.33
	CO ₂	1.9872	0.0249							
	CO	0.1638	0.0021							
	CH ₄	1.8542	0.0232							
150	H ₂	2.4308	0.0304	50.74	3.55	45.71	0.1226	37.57	2.63	74.05
	CO ₂	2.1901	0.0274							
	CO	0.1699	0.0021							
	CH ₄	2.5414	0.0318							
180	H ₂	2.5408	0.0318	51.96	3.47	44.56	0.1471	38.09	2.55	73.31
	CO ₂	2.1790	0.0273							
	CO	0.1699	0.0021							
	CH ₄	3.1361	0.0393							

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
195	H ₂	2.6662	0.0334	52.47	3.62	43.91	0.1594	38.07	2.63	72.56
	CO ₂	2.2310	0.0279							
	CO	0.1838	0.0023							
	CH ₄	3.4931	0.0437							
210	H ₂	2.9080	0.0364	53.10	3.51	43.39	0.1717	37.08	2.45	69.83
	CO ₂	2.3766	0.0298							
	CO	0.1921	0.0024							
	CH ₄	4.1352	0.0518							
240	H ₂	2.8459	0.0356	53.98	3.35	42.67	0.1962	38.52	2.39	71.37
	CO ₂	2.2500	0.0282							
	CO	0.1768	0.0022							
	CH ₄	4.4852	0.0562							
265	H ₂	2.8742	0.0360	55.52	3.30	41.17	0.2166	40.59	2.42	73.10
	CO ₂	2.1315	0.0267							
	CO	0.1710	0.0021							
	CH ₄	4.6527	0.0583							
280	H ₂	2.9320	0.0367	55.24	3.53	41.23	0.2289	40.36	2.58	73.07
	CO ₂	2.1888	0.0274							
	CO	0.1873	0.0023							
	CH ₄	4.9222	0.0616							
295	H ₂	3.0694	0.0384	55.88	3.47	40.65	0.2411	40.35	2.50	72.21
	CO ₂	2.2329	0.0280							
	CO	0.1904	0.0024							
	CH ₄	5.3523	0.0670							

Table F2 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

5%Ni loading

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
15	H2	0.8279	0.0104	56.72	11.69	31.59	0.0123	52.36	10.79	92.31
	CO2	0.4612	0.0058							
	CO	0.1706	0.0021							
	CH4	0.0753	0.0009							
30	H2	1.7872	0.0224	54.24	6.95	38.82	0.0245	53.43	6.84	98.51
	CO2	1.2792	0.0160							
	CO	0.2289	0.0029							
	CH4	0.0292	0.0004							
45	H2	2.5562	0.0320	53.24	6.15	40.61	0.0368	51.35	5.93	96.44
	CO2	1.9494	0.0244							
	CO	0.2952	0.0037							
	CH4	0.1045	0.0013							
60	H2	3.2741	0.0410	52.55	5.94	41.51	0.0490	49.53	5.60	94.25
	CO2	2.5865	0.0324							
	CO	0.3699	0.0046							
	CH4	0.2253	0.0028							
75	H2	3.7741	0.0473	52.15	5.39	42.46	0.0613	48.60	5.02	93.18
	CO2	3.0726	0.0385							
	CO	0.3902	0.0049							
	CH4	0.3338	0.0042							
90	H2	4.2356	0.0530	51.50	5.35	43.16	0.0736	46.79	4.86	90.86
	CO2	3.5498	0.0445							
	CO	0.4398	0.0055							
	CH4	0.5372	0.0067							
105	H2	4.6082	0.0577	51.72	5.47	42.81	0.0858	46.75	4.94	90.40
	CO2	3.8145	0.0478							
	CO	0.4872	0.0061							
	CH4	0.6583	0.0082							
120	H2	4.9539	0.0620	51.43	5.28	43.29	0.0981	45.63	4.68	88.73
	CO2	4.1705	0.0522							
	CO	0.5084	0.0064							
	CH4	0.8829	0.0111							
135	H2	5.4288	0.0680	50.91	4.96	44.13	0.1104	43.86	4.27	86.15
	CO2	4.7062	0.0589							
	CO	0.5289	0.0066							
	CH4	1.2210	0.0153							
150	H2	5.2162	0.0653	50.30	4.93	44.77	0.1226	42.92	4.21	85.33
	CO2	4.6430	0.0581							
	CO	0.5112	0.0064							
	CH4	1.4360	0.0180							

rxn. time (min)	Gas	V (L)	n _P (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
165	H ₂	5.1600	0.0646	49.98	4.77	45.25	0.1349	42.26	4.04	84.55
	CO ₂	4.6718	0.0585							
	CO	0.4929	0.0062							
	CH ₄	1.6637	0.0208							
180	H ₂	5.4539	0.0683	50.65	4.60	44.75	0.1471	42.13	3.83	83.18
	CO ₂	4.8190	0.0603							
	CO	0.4954	0.0062							
	CH ₄	1.9765	0.0248							
195	H ₂	5.3826	0.0674	49.81	4.58	45.61	0.1594	40.64	3.74	81.60
	CO ₂	4.9291	0.0617							
	CO	0.4951	0.0062							
	CH ₄	2.3423	0.0293							
210	H ₂	5.1761	0.0648	48.30	4.69	47.01	0.1717	38.87	3.78	80.47
	CO ₂	5.0382	0.0631							
	CO	0.5028	0.0063							
	CH ₄	2.6767	0.0335							
240	H ₂	4.5985	0.0576	47.61	4.59	47.79	0.1962	37.61	3.63	78.98
	CO ₂	4.6158	0.0578							
	CO	0.4438	0.0056							
	CH ₄	3.2929	0.0412							
265	H ₂	5.1473	0.0645	50.99	4.15	44.86	0.2166	39.72	3.23	77.90
	CO ₂	4.5288	0.0567							
	CO	0.4185	0.0052							
	CH ₄	3.8236	0.0479							
280	H ₂	4.9673	0.0622	51.29	4.34	44.36	0.2289	39.90	3.38	77.79
	CO ₂	4.2965	0.0538							
	CO	0.4207	0.0053							
	CH ₄	4.0589	0.0508							
295	H ₂	5.2034	0.0652	51.43	4.32	44.25	0.2411	39.18	3.29	76.19
	CO ₂	4.4769	0.0561							
	CO	0.4370	0.0055							
	CH ₄	4.5859	0.0574							

Table F3 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

7%Ni loading

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H2	1.9838	0.0248	55.26	8.04	36.70	0.0245	55.26	8.04	99.99
	CO2	1.3175	0.0165							
	CO	0.2885	0.0036							
	CH4	0.0001	0.0000							
45	H2	2.8165	0.0353	53.98	6.67	39.35	0.0368	53.80	6.65	99.67
	CO2	2.0532	0.0257							
	CO	0.3483	0.0044							
	CH4	0.0097	0.0001							
60	H2	3.3914	0.0425	54.78	7.35	37.87	0.0490	54.23	7.28	98.99
	CO2	2.3443	0.0294							
	CO	0.4550	0.0057							
	CH4	0.0397	0.0005							
75	H2	4.1649	0.0522	54.39	7.11	38.49	0.0613	53.48	6.99	98.32
	CO2	2.9475	0.0369							
	CO	0.5447	0.0068							
	CH4	0.0820	0.0010							
90	H2	5.2650	0.0659	53.53	6.71	39.75	0.0736	52.00	6.52	97.14
	CO2	3.9095	0.0490							
	CO	0.6603	0.0083							
	CH4	0.1681	0.0021							
105	H2	5.8059	0.0727	53.23	6.52	40.25	0.0858	51.33	6.28	96.44
	CO2	4.3905	0.0550							
	CO	0.7107	0.0089							
	CH4	0.2442	0.0031							
120	H2	6.6950	0.0838	53.13	6.19	40.68	0.0981	50.73	5.91	95.49
	CO2	5.1255	0.0642							
	CO	0.7804	0.0098							
	CH4	0.3535	0.0044							
135	H2	7.1055	0.0890	53.10	6.16	40.74	0.1104	50.26	5.83	94.66
	CO2	5.4516	0.0683							
	CO	0.8246	0.0103							
	CH4	0.4706	0.0059							
150	H2	7.7021	0.0964	52.53	5.95	41.52	0.1226	49.16	5.57	93.59
	CO2	6.0883	0.0762							
	CO	0.8727	0.0109							
	CH4	0.6278	0.0079							
165	H2	8.4990	0.1064	52.34	5.82	41.84	0.1349	48.32	5.38	92.33
	CO2	6.7937	0.0851							
	CO	0.9457	0.0118							
	CH4	0.8266	0.0104							

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
180	H ₂	9.2382	0.1157	50.88	5.02	44.10	0.1471	46.15	4.56	90.71
	CO ₂	8.0079	0.1003							
	CO	0.9122	0.0114							
	CH ₄	1.0911	0.0137							
195	H ₂	8.9797	0.1124	51.35	5.49	43.16	0.1594	46.59	4.98	90.72
	CO ₂	7.5467	0.0945							
	CO	0.9607	0.0120							
	CH ₄	1.1812	0.0148							
210	H ₂	9.0225	0.1130	51.14	5.30	43.55	0.1717	46.00	4.77	89.93
	CO ₂	7.6831	0.0962							
	CO	0.9354	0.0117							
	CH ₄	1.3801	0.0173							
225	H ₂	9.1172	0.1142	50.92	5.26	43.82	0.1839	45.40	4.69	89.17
	CO ₂	7.8460	0.0983							
	CO	0.9415	0.0118							
	CH ₄	1.5913	0.0199							
240	H ₂	9.1529	0.1146	50.52	4.95	44.53	0.1962	44.34	4.35	87.76
	CO ₂	8.0679	0.1010							
	CO	0.8970	0.0112							
	CH ₄	1.9171	0.0240							
265	H ₂	9.5642	0.1198	50.72	4.56	44.71	0.2166	43.63	3.93	86.02
	CO ₂	8.4304	0.1056							
	CO	0.8607	0.0108							
	CH ₄	2.4181	0.0303							
280	H ₂	9.5723	0.1199	50.79	4.61	44.60	0.2289	43.29	3.93	85.23
	CO ₂	8.4051	0.1053							
	CO	0.8692	0.0109							
	CH ₄	2.6994	0.0338							
295	H ₂	9.8589	0.1235	50.86	4.65	44.49	0.2411	42.83	3.91	84.20
	CO ₂	8.6244	0.1080							
	CO	0.9006	0.0113							
	CH ₄	3.0422	0.0381							

Table F4 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

9%Ni loading

rxn. time (min)	Gas	V (L)	n _P (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
15	H2	1.0214	0.0128	56.09	7.92	35.99	0.0123	56.09	7.92	100.00
	CO2	0.6554	0.0082							
	CO	0.1443	0.0018							
	CH4	0.0000	0.0000							
30	H2	2.0605	0.0258	55.14	7.46	37.40	0.0245	55.14	7.46	100.00
	CO2	1.3975	0.0175							
	CO	0.2788	0.0035							
	CH4	0.0000	0.0000							
45	H2	2.9026	0.0363	54.70	7.20	38.10	0.0368	54.60	7.19	99.82
	CO2	2.0217	0.0253							
	CO	0.3822	0.0048							
	CH4	0.0053	0.0001							
60	H2	3.8568	0.0483	54.15	7.12	38.73	0.0490	53.65	7.05	99.07
	CO2	2.7583	0.0345							
	CO	0.5071	0.0063							
	CH4	0.0363	0.0005							
75	H2	4.7141	0.0590	54.27	6.94	38.80	0.0613	53.40	6.82	98.40
	CO2	3.3702	0.0422							
	CO	0.6025	0.0075							
	CH4	0.0784	0.0010							
90	H2	5.4163	0.0678	53.96	6.85	39.18	0.0736	52.76	6.70	97.77
	CO2	3.9327	0.0492							
	CO	0.6878	0.0086							
	CH4	0.1312	0.0016							
105	H2	6.1354	0.0768	53.74	6.75	39.50	0.0858	52.16	6.55	97.05
	CO2	4.5095	0.0565							
	CO	0.7710	0.0097							
	CH4	0.2023	0.0025							
120	H2	6.8530	0.0858	53.60	6.58	39.81	0.0981	51.62	6.34	96.29
	CO2	5.0896	0.0637							
	CO	0.8418	0.0105							
	CH4	0.2906	0.0036							
135	H2	7.4243	0.0930	53.42	6.47	40.10	0.1104	51.06	6.19	95.58
	CO2	5.5738	0.0698							
	CO	0.8999	0.0113							
	CH4	0.3895	0.0049							
150	H2	7.9355	0.0994	53.17	6.38	40.45	0.1226	50.41	6.05	94.81
	CO2	6.0363	0.0756							
	CO	0.9520	0.0119							
	CH4	0.5080	0.0064							

rxn. time (min)	Gas	V (L)	$n_{i,P}$ (mol)	S_{H_2} (%)	S_{CO} (%)	S_{CO_2} (%)	$n_{CH_4,F}$ (mol)	H_2 Yield (%)	CO Yield (%)	CH_4 Conversion
165	H2	8.5188	0.1067	52.95	6.17	40.88	0.1349	49.73	5.79	93.92
	CO2	6.5772	0.0824							
	CO	0.9926	0.0124							
	CH4	0.6546	0.0082							
180	H2	9.0772	0.1137	52.72	6.05	41.23	0.1471	49.10	5.63	93.13
	CO2	7.1002	0.0889							
	CO	1.0418	0.0130							
	CH4	0.8068	0.0101							
195	H2	9.5824	0.1200	52.72	6.03	41.26	0.1594	48.70	5.57	92.38
	CO2	7.4992	0.0939							
	CO	1.0955	0.0137							
	CH4	0.9696	0.0121							
210	H2	9.9914	0.1251	52.59	5.99	41.42	0.1717	48.32	5.50	91.87
	CO2	7.8694	0.0985							
	CO	1.1370	0.0142							
	CH4	1.1142	0.0140							
225	H2	10.1359	0.1269	52.35	5.89	41.76	0.1839	47.66	5.36	91.05
	CO2	8.0867	0.1013							
	CO	1.1399	0.0143							
	CH4	1.3152	0.0165							
240	H2	10.6161	0.1329	52.08	5.70	42.22	0.1962	46.95	5.14	90.16
	CO2	8.6070	0.1078							
	CO	1.1612	0.0145							
	CH4	1.5421	0.0193							
265	H2	11.4100	0.1429	52.09	5.25	42.65	0.2166	46.32	4.67	88.93
	CO2	9.3428	0.1170							
	CO	1.1506	0.0144							
	CH4	1.9156	0.0240							
280	H2	11.3235	0.1418	51.73	5.27	43.00	0.2289	45.47	4.63	87.89
	CO2	9.4127	0.1179							
	CO	1.1529	0.0144							
	CH4	2.2127	0.0277							
295	H2	11.5276	0.1444	51.70	5.26	43.05	0.2411	45.23	4.60	87.48
	CO2	9.5984	0.1202							
	CO	1.1725	0.0147							
	CH4	2.4105	0.0302							

Table F5 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni loading

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H ₂	2.0760	0.0260	55.48	7.80	36.72	0.0245	55.48	7.80	100.00
	CO ₂	1.3740	0.0172							
	CO	0.2917	0.0037							
	CH ₄	0.0000	0.0000							
45	H ₂	3.0704	0.0384	54.22	7.02	38.76	0.0368	54.22	7.02	100.00
	CO ₂	2.1949	0.0275							
	CO	0.3974	0.0050							
	CH ₄	0.0000	0.0000							
60	H ₂	4.1379	0.0518	55.12	7.56	37.32	0.0490	55.12	7.56	100.00
	CO ₂	2.8016	0.0351							
	CO	0.5677	0.0071							
	CH ₄	0.0000	0.0000							
75	H ₂	5.0568	0.0633	54.91	7.65	37.44	0.0613	54.91	7.65	100.00
	CO ₂	3.4482	0.0432							
	CO	0.7041	0.0088							
	CH ₄	0.0000	0.0000							
90	H ₂	6.1093	0.0765	54.74	7.35	37.91	0.0736	54.74	7.35	100.00
	CO ₂	4.2302	0.0530							
	CO	0.8203	0.0103							
	CH ₄	0.0000	0.0000							
105	H ₂	7.1432	0.0894	54.87	7.48	37.65	0.0858	54.87	7.48	100.00
	CO ₂	4.9021	0.0614							
	CO	0.9738	0.0122							
	CH ₄	0.0000	0.0000							
120	H ₂	8.1520	0.1021	54.55	7.23	38.22	0.0981	54.55	7.23	100.00
	CO ₂	5.7121	0.0715							
	CO	1.0810	0.0135							
	CH ₄	0.0000	0.0000							
135	H ₂	8.9928	0.1126	54.57	7.22	38.21	0.1104	54.57	7.22	100.00
	CO ₂	6.2970	0.0789							
	CO	1.1892	0.0149							
	CH ₄	0.0000	0.0000							
150	H ₂	10.0226	0.1255	54.18	7.08	38.74	0.1226	54.09	7.07	99.84
	CO ₂	7.1653	0.0897							
	CO	1.3103	0.0164							
	CH ₄	0.0158	0.0002							
210	H ₂	11.2544	0.1409	52.77	6.35	40.88	0.1717	50.66	6.09	96.00
	CO ₂	8.7185	0.1092							
	CO	1.3537	0.0170							
	CH ₄	0.5482	0.0069							

rxn. time (min)	Gas	V (L)	$n_{i,P}$ (mol)	S_{H_2} (%)	S_{CO} (%)	S_{CO_2} (%)	$n_{CH_4,F}$ (mol)	H_2 Yield (%)	CO Yield (%)	CH_4 Conversion
225	H2	11.6087	0.1454	52.48	6.10	41.41	0.1839	49.60	5.77	94.50
	CO2	9.1601	0.1147							
	CO	1.3503	0.0169							
	CH4	0.8081	0.0101							
240	H2	12.2652	0.1536	52.52	5.85	41.63	0.1962	48.83	5.43	92.97
	CO2	9.7209	0.1217							
	CO	1.3651	0.0171							
	CH4	1.1012	0.0138							
265	H2	12.4142	0.1555	51.58	5.07	43.35	0.2166	47.29	4.65	91.67
	CO2	10.4322	0.1306							
	CO	1.2199	0.0153							
	CH4	1.4402	0.0180							
280	H2	12.6704	0.1587	51.79	5.30	42.91	0.2289	47.31	4.84	91.36
	CO2	10.4977	0.1315							
	CO	1.2973	0.0162							
	CH4	1.5793	0.0198							
295	H2	12.4740	0.1562	51.16	5.15	43.69	0.2289	46.28	4.66	90.47
	CO2	10.6525	0.1334							
	CO	1.2567	0.0157							
	CH4	1.7414	0.0218							

Table F6 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

15%Ni loading

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H2	0.7994	0.0100	69.69	16.99	13.32	0.0245	50.19	12.24	72.03
	CO2	0.1528	0.0019							
	CO	0.1949	0.0024							
	CH4	0.5479	0.0069							
45	H2	1.9340	0.0242	62.16	13.56	24.28	0.0368	52.89	11.54	85.08
	CO2	0.7555	0.0095							
	CO	0.4219	0.0053							
	CH4	0.4382	0.0055							
60	H2	3.7346	0.0468	54.24	7.11	38.66	0.0490	54.17	7.10	99.88
	CO2	2.6620	0.0333							
	CO	0.4893	0.0061							
	CH4	0.0046	0.0001							
75	H2	4.7155	0.0590	54.29	6.88	38.83	0.0613	53.90	6.83	99.28
	CO2	3.3729	0.0422							
	CO	0.5974	0.0075							
	CH4	0.0351	0.0004							
90	H2	5.3193	0.0666	53.82	6.64	39.54	0.0736	52.87	6.53	98.25
	CO2	3.9082	0.0489							
	CO	0.6565	0.0082							
	CH4	0.1030	0.0013							
105	H2	6.1694	0.0773	53.59	6.48	39.92	0.0858	52.24	6.32	97.48
	CO2	4.5960	0.0576							
	CO	0.7462	0.0093							
	CH4	0.1727	0.0022							
120	H2	6.7298	0.0843	53.13	6.20	40.67	0.0981	51.30	5.98	96.56
	CO2	5.1509	0.0645							
	CO	0.7849	0.0098							
	CH4	0.2698	0.0034							
135	H2	7.7331	0.0968	53.00	6.16	40.84	0.1104	50.74	5.89	95.73
	CO2	5.9590	0.0746							
	CO	0.8984	0.0112							
	CH4	0.3765	0.0047							
150	H2	8.4185	0.1054	52.85	6.23	40.91	0.1226	50.37	5.94	95.30
	CO2	6.5168	0.0816							
	CO	0.9926	0.0124							
	CH4	0.4603	0.0058							
165	H2	8.5323	0.1068	52.69	6.10	41.21	0.1349	50.14	5.81	95.16
	CO2	6.6724	0.0836							
	CO	0.9878	0.0124							
	CH4	0.5209	0.0065							

rxn. time (min)	Gas	V (L)	n _P (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH_{4,F}} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
180	H ₂	9.3822	0.1175	52.39	5.99	41.62	0.1471	49.44	5.65	94.38
	CO ₂	7.4542	0.0933							
	CO	1.0725	0.0134							
	CH ₄	0.6605	0.0083							
195	H ₂	10.0526	0.1259	52.47	6.00	41.53	0.1594	49.25	5.64	93.86
	CO ₂	7.9557	0.0996							
	CO	1.1505	0.0144							
	CH ₄	0.7816	0.0098							
210	H ₂	10.2361	0.1282	52.20	5.83	41.97	0.1717	48.85	5.46	93.58
	CO ₂	8.2288	0.1030							
	CO	1.1436	0.0143							
	CH ₄	0.8802	0.0110							
265	H ₂	12.1037	0.1516	52.11	5.44	42.45	0.2166	47.46	4.95	91.07
	CO ₂	9.8592	0.1235							
	CO	1.2632	0.0158							
	CH ₄	1.5450	0.0193							
295	H ₂	12.8086	0.1604	51.72	5.10	43.18	0.2411	46.09	4.55	89.13
	CO ₂	10.6952	0.1339							
	CO	1.2639	0.0158							
	CH ₄	2.0941	0.0262							

Table F7 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni loading at R = 1

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H2	2.0609	0.0258	52.72	5.54	41.74	0.0245	52.72	5.54	100.00
	CO2	1.6314	0.0204							
	CO	0.2167	0.0027							
	CH4	0.0000	0.0000							
45	H2	2.9952	0.0375	52.17	5.25	42.59	0.0368	51.82	5.21	99.33
	CO2	2.4451	0.0306							
	CO	0.3012	0.0038							
	CH4	0.0195	0.0002							
60	H2	3.8887	0.0487	52.22	5.48	42.30	0.0490	51.13	5.36	97.91
	CO2	3.1498	0.0394							
	CO	0.4080	0.0051							
	CH4	0.0818	0.0010							
75	H2	4.5779	0.0573	52.05	5.36	42.60	0.0613	50.55	5.20	97.12
	CO2	3.7467	0.0469							
	CO	0.4714	0.0059							
	CH4	0.1408	0.0018							
90	H2	5.1279	0.0642	51.41	4.96	43.64	0.0736	49.06	4.73	95.43
	CO2	4.3532	0.0545							
	CO	0.4943	0.0062							
	CH4	0.2684	0.0034							
105	H2	5.6597	0.0709	51.28	4.81	43.91	0.0858	48.22	4.52	94.02
	CO2	4.8457	0.0607							
	CO	0.5304	0.0066							
	CH4	0.4100	0.0051							
120	H2	5.9662	0.0747	51.20	4.79	44.01	0.0981	47.11	4.41	92.02
	CO2	5.1277	0.0642							
	CO	0.5585	0.0070							
	CH4	0.6253	0.0078							
135	H2	6.3259	0.0792	50.82	4.53	44.65	0.1104	45.32	4.04	89.19
	CO2	5.5585	0.0696							
	CO	0.5639	0.0071							
	CH4	0.9529	0.0119							
150	H2	6.0843	0.0762	50.43	4.23	45.34	0.1226	44.35	3.72	87.94
	CO2	5.4701	0.0685							
	CO	0.5102	0.0064							
	CH4	1.1806	0.0148							
165	H2	5.1954	0.0651	49.58	3.68	46.75	0.1349	41.61	3.08	83.92
	CO2	4.8984	0.0613							
	CO	0.3852	0.0048							
	CH4	1.7317	0.0217							

rxn. time (min)	Gas	V (L)	n _{i,P} (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH_{4,F}} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
180	H ₂	5.2611	0.0659	49.53	3.70	46.77	0.1471	40.18	3.00	81.12
	CO ₂	4.9681	0.0622							
	CO	0.3929	0.0049							
	CH ₄	2.2190	0.0278							
195	H ₂	4.9613	0.0621	49.25	3.53	47.22	0.1594	38.83	2.78	78.83
	CO ₂	4.7560	0.0596							
	CO	0.3556	0.0045							
	CH ₄	2.6942	0.0337							
210	H ₂	4.7417	0.0594	48.84	3.45	47.71	0.1717	37.55	2.65	76.88
	CO ₂	4.6318	0.0580							
	CO	0.3345	0.0042							
	CH ₄	3.1701	0.0397							
225	H ₂	4.3515	0.0545	49.28	3.30	47.42	0.1839	37.87	2.54	76.84
	CO ₂	4.1868	0.0524							
	CO	0.2914	0.0036							
	CH ₄	3.4024	0.0426							
240	H ₂	4.3492	0.0545	49.23	3.34	47.43	0.1962	37.03	2.51	75.22
	CO ₂	4.1905	0.0525							
	CO	0.2950	0.0037							
	CH ₄	3.8826	0.0486							
265	H ₂	4.2348	0.0530	50.81	3.12	46.07	0.2166	37.74	2.32	74.27
	CO ₂	3.8400	0.0481							
	CO	0.2604	0.0033							
	CH ₄	4.4506	0.0557							
280	H ₂	4.1036	0.0514	51.80	3.07	45.14	0.2289	38.52	2.28	74.36
	CO ₂	3.5762	0.0448							
	CO	0.2428	0.0030							
	CH ₄	4.6858	0.0587							
295	H ₂	4.1306	0.0517	52.10	3.14	44.76	0.2411	38.58	2.32	74.05
	CO ₂	3.5488	0.0444							
	CO	0.2486	0.0031							
	CH ₄	4.9972	0.0626							

Table F8 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni loading at R = 2

rxn. time (min)	Gas	V (L)	n _{r,P} (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H2	0.6467	0.0081	47.35	0.98	51.67	0.0245	37.19	0.77	78.54
	CO2	0.7056	0.0088							
	CO	0.0134	0.0002							
	CH4	0.4202	0.0053							
45	H2	1.0259	0.0128	47.78	1.42	50.80	0.0368	38.31	1.14	80.19
	CO2	1.0909	0.0137							
	CO	0.0305	0.0004							
	CH4	0.5820	0.0073							
60	H2	1.0169	0.0127	48.26	1.29	50.45	0.0490	36.65	0.98	75.95
	CO2	1.0629	0.0133							
	CO	0.0272	0.0003							
	CH4	0.9422	0.0118							
75	H2	1.1161	0.0140	49.64	1.28	49.08	0.0613	36.03	0.93	72.59
	CO2	1.1036	0.0138							
	CO	0.0288	0.0004							
	CH4	1.3420	0.0168							
90	H2	0.9520	0.0119	53.00	1.16	45.85	0.0736	38.57	0.84	72.77
	CO2	0.8235	0.0103							
	CO	0.0208	0.0003							
	CH4	1.5999	0.0200							
105	H2	0.8054	0.0101	61.66	0.76	37.59	0.0858	43.01	0.53	69.76
	CO2	0.4910	0.0061							
	CO	0.0099	0.0001							
	CH4	2.0724	0.0260							
120	H2	0.7461	0.0093	76.47	0.18	23.35	0.0981	51.75	0.12	67.67
	CO2	0.2279	0.0029							
	CO	0.0017	0.0000							
	CH4	2.5324	0.0317							
135	H2	0.7295	0.0091	89.40	0.00	10.60	0.1104	60.84	0.00	68.06
	CO2	0.0865	0.0011							
	CO	0.0000	0.0000							
	CH4	2.8150	0.0353							
150	H2	0.7874	0.0099	94.93	0.00	5.07	0.1226	64.19	0.00	67.62
	CO2	0.0421	0.0005							
	CO	0.0000	0.0000							
	CH4	3.1706	0.0397							
165	H2	0.0000	0.0000	0.00	0.00	100.00	0.1349	0.00	0.00	68.22
	CO2	0.0205	0.0003							
	CO	0.0000	0.0000							
	CH4	3.4235	0.0429							

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
180	H2	0.0000	0.0000	0.00	0.00	100.00	0.1471	0.00	0.00	66.94
	CO2	0.0232	0.0003							
	CO	0.0000	0.0000							
	CH4	3.8842	0.0486							
195	H2	0.0000	0.0000	0.00	0.00	100.00	0.1594	0.00	0.00	66.77
	CO2	0.0254	0.0003							
	CO	0.0000	0.0000							
	CH4	4.2299	0.0530							
210	H2	0.0000	0.0000	0.00	0.00	100.00	0.1717	0.00	0.00	66.72
	CO2	0.0274	0.0003							
	CO	0.0000	0.0000							
	CH4	4.5620	0.0571							
225	H2	0.0000	0.0000	0.00	0.00	100.00	0.1839	0.00	0.00	66.90
	CO2	0.0293	0.0004							
	CO	0.0000	0.0000							
	CH4	4.8623	0.0609							
240	H2	0.0000	0.0000	0.00	0.00	100.00	0.1962	0.00	0.00	66.92
	CO2	0.0313	0.0004							
	CO	0.0000	0.0000							
	CH4	5.1820	0.0649							
265	H2	0.0000	0.0000	0.00	0.00	100.00	0.2166	0.00	0.00	67.99
	CO2	0.0336	0.0004							
	CO	0.0000	0.0000							
	CH4	5.5372	0.0693							
280	H2	0.0000	0.0000	0.00	0.00	100.00	0.2289	0.00	0.00	66.55
	CO2	0.0372	0.0005							
	CO	0.0000	0.0000							
	CH4	6.1140	0.0766							
295	H2	0.0000	0.0000	0.00	0.00	100.00	0.2411	0.00	0.00	67.12
	CO2	0.0385	0.0005							
	CO	0.0000	0.0000							
	CH4	6.3324	0.0793							

Table F9 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni loading at R = 2 (repeat)

rxn. time (min)	Gas	V (L)	n _P (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H ₂	0.8052	0.0101	47.46	1.51	51.03	0.0245	39.60	1.26	83.43
	CO ₂	0.8658	0.0108							
	CO	0.0255	0.0003							
	CH ₄	0.3244	0.0041							
45	H ₂	0.9508	0.0119	47.76	1.60	50.64	0.0368	38.46	1.29	80.52
	CO ₂	1.0079	0.0126							
	CO	0.0318	0.0004							
	CH ₄	0.5723	0.0072							
60	H ₂	1.0053	0.0126	48.57	1.52	49.91	0.0490	37.65	1.17	77.51
	CO ₂	1.0330	0.0129							
	CO	0.0214	0.0004							
	CH ₄	0.8807	0.0110							
75	H ₂	0.8927	0.0112	50.83	1.29	47.88	0.0613	37.70	0.95	74.16
	CO ₂	0.8409	0.0105							
	CO	0.0226	0.0003							
	CH ₄	1.2652	0.0158							
90	H ₂	0.8519	0.0107	55.23	1.14	43.62	0.0736	39.88	0.82	72.20
	CO ₂	0.6728	0.0084							
	CO	0.0176	0.0002							
	CH ₄	1.6932	0.0205							
105	H ₂	0.6886	0.0086	70.00	0.42	29.58	0.0858	48.61	0.29	69.44
	CO ₂	0.2910	0.0036							
	CO	0.0041	0.0001							
	CH ₄	2.0947	0.0262							
120	H ₂	0.6328	0.0079	89.26	0.00	10.74	0.0981	61.89	0.00	69.34
	CO ₂	0.0762	0.0010							
	CO	0.0000	0.0000							
	CH ₄	2.4015	0.0301							
135	H ₂	0.7056	0.0088	94.33	0.00	5.67	0.1104	64.02	0.00	67.87
	CO ₂	0.0424	0.0005							
	CO	0.0000	0.0000							
	CH ₄	2.8317	0.0355							
150	H ₂	0.0000	0.0000	0.00	0.00	100.00	0.1226	0.00	0.00	67.17
	CO ₂	0.0465	0.0006							
	CO	0.0000	0.0000							
	CH ₄	3.2149	0.0403							
165	H ₂	0.0000	0.0000	0.00	0.00	100.00	0.1349	0.00	0.00	69.14
	CO ₂	0.0203	0.0003							
	CO	0.0000	0.0000							
	CH ₄	3.3244	0.0416							

rxn. time (min)	Gas	V (L)	n _{r,p} (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
180	H2	0.0000	0.0000	0.00	0.00	100.00	0.1471	0.00	0.00	67.80
	CO2	0.0229	0.0003							
	CO	0.0000	0.0000							
	CH4	3.7839	0.0474							
195	H2	0.0000	0.0000	0.00	0.00	100.00	0.1594	0.00	0.00	68.41
	CO2	0.0243	0.0003							
	CO	0.0000	0.0000							
	CH4	4.0213	0.0504							
210	H2	0.0000	0.0000	0.00	0.00	100.00	0.1717	0.00	0.00	71.38
	CO2	0.0236	0.0003							
	CO	0.0000	0.0000							
	CH4	3.9230	0.0491							
225	H2	0.0000	0.0000	0.00	0.00	100.00	0.1839	0.00	0.00	65.41
	CO2	0.0284	0.0004							
	CO	0.0000	0.0000							
	CH4	5.0803	0.0636							
240	H2	0.0000	0.0000	0.00	0.00	100.00	0.1962	0.00	0.00	67.91
	CO2	0.0297	0.0004							
	CO	0.0000	0.0000							
	CH4	5.0282	0.0630							
265	H2	0.0000	0.0000	0.00	0.00	100.00	0.2166	0.00	0.00	68.12
	CO2	0.0332	0.0004							
	CO	0.0000	0.0000							
	CH4	5.5155	0.0691							
280	H2	0.0000	0.0000	0.00	0.00	100.00	0.2289	0.00	0.00	68.34
	CO2	0.0347	0.0004							
	CO	0.0000	0.0000							
	CH4	5.7870	0.0725							
295	H2	0.0000	0.0000	0.00	0.00	100.00	0.2411	0.00	0.00	68.04
	CO2	0.0367	0.0005							
	CO	0.0000	0.0000							
	CH4	6.1539	0.0771							

Table F10 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni/3%Ce/ZSM-5

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H2	1.9902	0.0249	53.85	6.52	39.63	0.0245	53.85	6.52	100.00
	CO2	1.4646	0.0183							
	CO	0.2409	0.0030							
	CH4	0.0000	0.0000							
45	H2	2.9983	0.0375	54.92	7.35	37.73	0.0368	54.92	7.35	100.00
	CO2	2.0598	0.0258							
	CO	0.4010	0.0050							
	CH4	0.0000	0.0000							
60	H2	3.9535	0.0495	54.15	7.17	38.68	0.0490	54.15	7.17	100.00
	CO2	2.8242	0.0354							
	CO	0.5233	0.0066							
	CH4	0.0000	0.0000							
75	H2	4.9440	0.0619	54.35	7.02	38.63	0.0613	54.35	7.02	100.00
	CO2	3.5133	0.0440							
	CO	0.6385	0.0080							
	CH4	0.0000	0.0000							
90	H2	5.8203	0.0729	54.23	7.18	38.58	0.0736	54.23	7.18	100.00
	CO2	4.1404	0.0518							
	CO	0.7710	0.0097							
	CH4	0.0000	0.0000							
105	H2	7.0084	0.0878	53.97	6.98	39.04	0.0858	53.97	6.98	100.00
	CO2	5.0697	0.0635							
	CO	0.9066	0.0114							
	CH4	0.0000	0.0000							
120	H2	7.9314	0.0993	54.26	7.20	38.55	0.0981	54.26	7.20	100.00
	CO2	5.6347	0.0706							
	CO	1.0524	0.0132							
	CH4	0.0000	0.0000							
135	H2	8.8436	0.1107	54.27	7.11	38.62	0.1104	54.22	7.10	99.90
	CO2	6.2936	0.0788							
	CO	1.1580	0.0145							
	CH4	0.0088	0.0001							
150	H2	9.6608	0.1210	53.87	6.91	39.22	0.1226	53.74	6.89	99.76
	CO2	7.0326	0.0881							
	CO	1.2393	0.0155							
	CH4	0.0232	0.0003							
165	H2	10.5529	0.1321	53.85	7.05	39.10	0.1349	53.60	7.02	99.55
	CO2	7.6640	0.0960							
	CO	1.3817	0.0173							
	CH4	0.0489	0.0006							

rxn. time (min)	Gas	V (L)	n _{i,F} (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
180	H ₂	11.5149	0.1442	53.58	6.94	39.48	0.1471	53.32	6.91	99.52
	CO ₂	8.4857	0.1063							
	CO	1.4922	0.0187							
	CH ₄	0.0559	0.0007							
195	H ₂	12.3656	0.1548	53.55	6.81	39.64	0.1594	53.20	6.77	99.35
	CO ₂	9.1553	0.1146							
	CO	1.5729	0.0197							
	CH ₄	0.0828	0.0010							
210	H ₂	12.9959	0.1627	52.67	6.60	40.73	0.1717	52.25	6.54	99.19
	CO ₂	10.0485	0.1258							
	CO	1.6278	0.0204							
	CH ₄	0.1114	0.0014							
225	H ₂	14.1365	0.1770	53.40	6.70	39.90	0.1839	52.84	6.63	98.97
	CO ₂	10.5645	0.1323							
	CO	1.7741	0.0222							
	CH ₄	0.1518	0.0019							
240	H ₂	15.1237	0.1894	53.15	6.78	40.08	0.1962	52.50	6.69	98.79
	CO ₂	11.4039	0.1428							
	CO	1.9281	0.0241							
	CH ₄	0.1898	0.0024							
265	H ₂	16.8270	0.2107	53.64	6.51	39.85	0.2166	52.88	6.42	98.58
	CO ₂	12.4985	0.1565							
	CO	2.0422	0.0256							
	CH ₄	0.2459	0.0031							
280	H ₂	17.7465	0.2222	53.96	6.71	39.33	0.2289	53.09	6.60	98.38
	CO ₂	12.9343	0.1620							
	CO	2.2058	0.0276							
	CH ₄	0.2954	0.0037							
295	H ₂	18.7468	0.2348	53.53	6.76	39.71	0.2411	52.60	6.64	98.26
	CO ₂	13.9069	0.1741							
	CO	2.3658	0.0296							
	CH ₄	0.3351	0.0042							

Table F11 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni/5%Ce/ZSM-5

rxn. time (min)	Gas	V (L)	n _{i,P} (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H ₂	2.0334	0.0255	52.48	5.61	41.91	0.0245	51.92	5.55	98.93
	CO ₂	1.6239	0.0203							
	CO	0.2174	0.0027							
	CH ₄	0.0209	0.0003							
45	H ₂	2.9547	0.0370	54.66	7.24	38.10	0.0368	54.19	7.18	99.15
	CO ₂	2.0596	0.0258							
	CO	0.3916	0.0049							
	CH ₄	0.0251	0.0003							
60	H ₂	3.9524	0.0495	54.20	7.08	38.72	0.0490	53.65	7.01	99.00
	CO ₂	2.8241	0.0354							
	CO	0.5163	0.0065							
	CH ₄	0.0393	0.0005							
75	H ₂	4.9110	0.0615	54.65	7.25	38.10	0.0613	54.10	7.18	99.01
	CO ₂	3.4240	0.0429							
	CO	0.6519	0.0082							
	CH ₄	0.0486	0.0006							
90	H ₂	5.9609	0.0746	54.46	7.15	38.39	0.0736	53.84	7.07	98.86
	CO ₂	4.2028	0.0526							
	CO	0.7827	0.0098							
	CH ₄	0.0667	0.0008							
105	H ₂	6.8563	0.0859	54.45	7.16	38.39	0.0858	53.84	7.08	98.88
	CO ₂	4.8339	0.0605							
	CO	0.9021	0.0113							
	CH ₄	0.0769	0.0010							
120	H ₂	7.8253	0.0980	54.25	7.13	38.62	0.0981	53.58	7.04	98.77
	CO ₂	5.5715	0.0698							
	CO	1.0289	0.0129							
	CH ₄	0.0962	0.0012							
135	H ₂	8.6799	0.1087	54.40	7.26	38.33	0.1104	53.69	7.17	98.68
	CO ₂	6.1158	0.0766							
	CO	1.1586	0.0145							
	CH ₄	0.1165	0.0015							
150	H ₂	9.7067	0.1216	54.49	7.31	38.20	0.1226	53.77	7.21	98.69
	CO ₂	6.8061	0.0852							
	CO	1.3020	0.0163							
	CH ₄	0.1280	0.0016							
165	H ₂	10.7044	0.1340	54.06	7.25	38.69	0.1349	53.28	7.14	98.57
	CO ₂	7.6625	0.0960							
	CO	1.4354	0.0180							
	CH ₄	0.1545	0.0019							

rxn. time (min)	Gas	V (L)	n _P (mol)	S _{H2} (%)	S _{CO} (%)	S _{CO2} (%)	n _{CH4,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
195	H2	12.5481	0.1571	54.11	7.24	38.66	0.1594	53.24	7.12	98.40
	CO2	8.9651	0.1123							
	CO	1.6779	0.0210							
	CH4	0.2035	0.0025							
210	H2	13.4906	0.1689	53.79	7.14	39.06	0.1717	52.92	7.03	98.38
	CO2	9.7961	0.1227							
	CO	1.7918	0.0224							
	CH4	0.2217	0.0028							
225	H2	14.0460	0.1759	53.51	7.32	39.17	0.1839	52.64	7.20	98.37
	CO2	10.2819	0.1288							
	CO	1.9210	0.0241							
	CH4	0.2389	0.0030							
240	H2	15.2983	0.1916	53.78	7.13	39.09	0.1962	52.85	7.00	98.27
	CO2	11.1192	0.1392							
	CO	2.0274	0.0254							
	CH4	0.2705	0.0034							
265	H2	17.1792	0.2151	54.00	6.81	39.19	0.2166	53.05	6.70	98.25
	CO2	12.4677	0.1561							
	CO	2.1679	0.0271							
	CH4	0.3021	0.0038							
280	H2	18.1681	0.2275	54.02	6.79	39.19	0.2289	53.00	6.66	98.11
	CO2	13.1793	0.1650							
	CO	2.2840	0.0286							
	CH4	0.3448	0.0043							
295	H2	18.9237	0.2370	54.06	6.81	39.12	0.2411	53.02	6.68	98.07
	CO2	13.6947	0.1715							
	CO	2.3846	0.0299							
	CH4	0.3709	0.0046							

Table F12 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni/5%Ce/ZSM-5 (repeat)

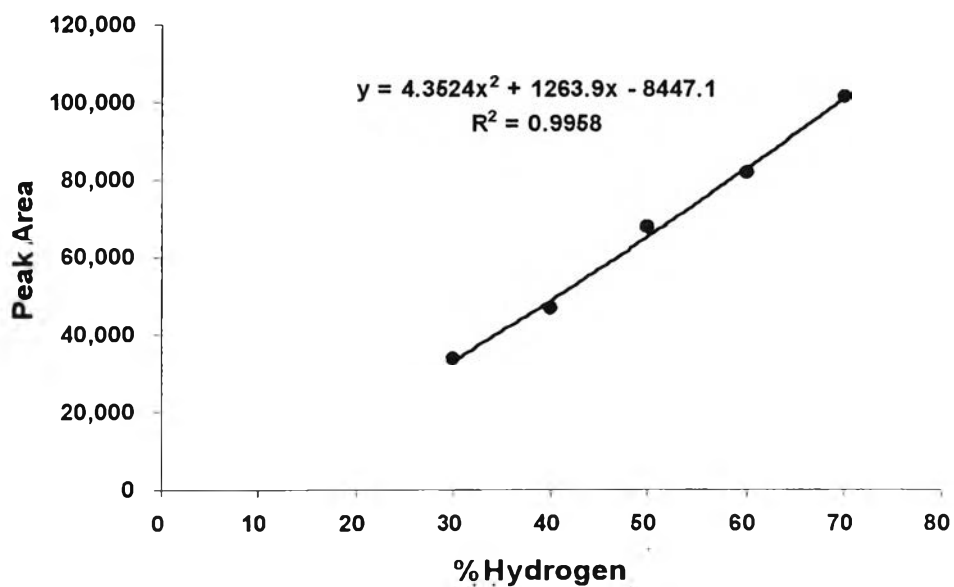
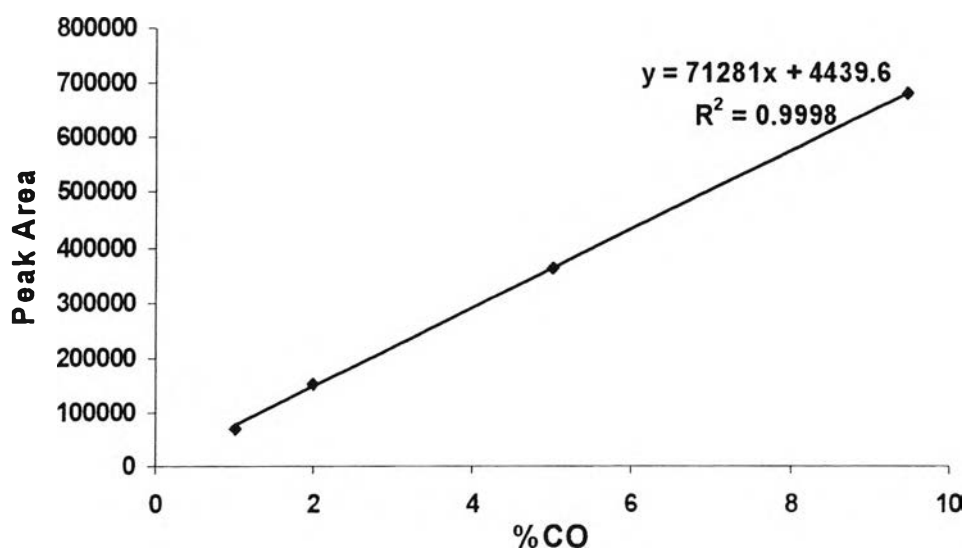
rxn. time (min)	Gas	V (L)	n _P (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
30	H2	2.2345	0.0280	53.37	10.37	36.26	0.0245	57.05	22.16	100.00
	CO2	1.5182	0.0190							
	CO	0.4341	0.0054							
	CH4	0.0000	0.0000							
45	H2	3.3782	0.0423	51.45	8.80	39.75	0.0368	57.50	19.66	100.00
	CO2	2.6103	0.0327							
	CO	0.5775	0.0072							
	CH4	0.0000	0.0000							
60	H2	4.1781	0.0523	52.95	10.21	36.84	0.0490	53.34	20.57	100.00
	CO2	2.9064	0.0364							
	CO	0.8055	0.0101							
	CH4	0.0000	0.0000							
75	H2	5.4656	0.0684	51.11	8.85	40.04	0.0613	55.82	19.33	100.00
	CO2	4.2816	0.0536							
	CO	0.9466	0.0119							
	CH4	0.0000	0.0000							
90	H2	6.5526	0.0821	50.64	8.48	40.88	0.0736	55.77	18.67	100.00
	CO2	5.2890	0.0662							
	CO	1.0968	0.0137							
	CH4	0.0000	0.0000							
105	H2	7.7306	0.0968	51.13	8.60	40.27	0.0858	56.39	18.97	100.00
	CO2	6.0896	0.0763							
	CO	1.3006	0.0163							
	CH4	0.0000	0.0000							
120	H2	8.7085	0.1091	51.15	9.00	39.85	0.0981	55.59	19.55	100.00
	CO2	6.7840	0.0850							
	CO	1.5318	0.0192							
	CH4	0.0000	0.0000							
135	H2	10.0604	0.1260	51.28	8.68	40.04	0.1104	57.08	19.33	100.00
	CO2	7.8564	0.0984							
	CO	1.7037	0.0213							
	CH4	0.0000	0.0000							
150	H2	10.7010	0.1340	51.04	8.80	40.17	0.1226	54.64	18.83	100.00
	CO2	8.4224	0.1055							
	CO	1.8442	0.0231							
	CH4	0.0000	0.0000							
165	H2	12.3417	0.1545	51.16	8.52	40.31	0.1349	57.29	19.09	100.00
	CO2	9.7237	0.1218							
	CO	2.0563	0.0258							
	CH4	0.0000	0.0000							

rxn. time (min)	Gas	V (L)	n _p (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
195	H ₂	14.0711	0.1762	50.63	8.45	40.93	0.1594	55.27	18.44	100.00
	CO ₂	11.3759	0.1425							
	CO	2.3476	0.0294							
	CH ₄	0.0000	0.0000							
210	H ₂	15.5454	0.1947	51.21	8.58	40.21	0.1717	56.70	19.01	99.99
	CO ₂	12.2080	0.1529							
	CO	2.6054	0.0326							
	CH ₄	0.0009	0.0000							
225	H ₂	16.7736	0.2100	51.41	8.73	39.86	0.1839	57.10	19.38	99.94
	CO ₂	13.0057	0.1629							
	CO	2.8467	0.0356							
	CH ₄	0.0089	0.0001							
240	H ₂	17.2262	0.2157	51.02	8.81	40.18	0.1962	54.98	18.98	99.88
	CO ₂	13.5651	0.1699							
	CO	2.9731	0.0372							
	CH ₄	0.0191	0.0002							
265	H ₂	19.3262	0.2420	51.13	8.63	40.23	0.2166	55.86	18.87	99.74
	CO ₂	15.2048	0.1904							
	CO	3.2635	0.0409							
	CH ₄	0.0456	0.0006							
280	H ₂	19.3837	0.2427	51.58	9.30	39.12	0.2289	53.02	19.13	99.73
	CO ₂	14.7036	0.1841							
	CO	3.4960	0.0438							
	CH ₄	0.0491	0.0006							
295	H ₂	21.5382	0.2697	51.21	8.62	40.16	0.2411	55.92	18.83	99.59
	CO ₂	16.8907	0.2115							
	CO	3.6265	0.0454							
	CH ₄	0.0780	0.0010							

Table F13 Effect of Ni loading: H₂ selectivity, CO selectivity, CO₂ selectivity, H₂ yield, CO yield, CH₄ conversion

11%Ni/7%Ce/ZSM-5

rxn. time (min)	Gas	V (L)	n _{i,P} (mol)	S _{H₂} (%)	S _{CO} (%)	S _{CO₂} (%)	n _{CH₄,F} (mol)	H ₂ Yield (%)	CO Yield (%)	CH ₄ Conversion
45	H ₂	0.2006	0.0025	85.83	9.99	4.18	0.0368	84.07	9.79	97.95
	CO ₂	0.0098	0.0001							
	CO	0.0234	0.0003							
	CH ₄	0.0603	0.0008							
60	H ₂	0.3541	0.0044	89.60	6.73	3.67	0.0490	87.89	6.60	98.09
	CO ₂	0.0145	0.0002							
	CO	0.0266	0.0003							
	CH ₄	0.0750	0.0009							
90	H ₂	0.5890	0.0074	96.54	2.11	1.35	0.0736	94.05	2.06	97.42
	CO ₂	0.0082	0.0001							
	CO	0.0129	0.0002							
	CH ₄	0.1514	0.0019							
105	H ₂	0.6469	0.0081	97.64	1.37	0.99	0.0858	95.22	1.34	97.52
	CO ₂	0.0065	0.0001							
	CO	0.0091	0.0001							
	CH ₄	0.1700	0.0021							
120	H ₂	0.7086	0.0089	98.24	0.96	0.79	0.0981	95.87	0.94	97.59
	CO ₂	0.0057	0.0001							
	CO	0.0069	0.0001							
	CH ₄	0.1888	0.0024							

Appendix G Calibration curves for product gases**Figure G1** H₂ calibration curve.**Figure G2** CO calibration curve.

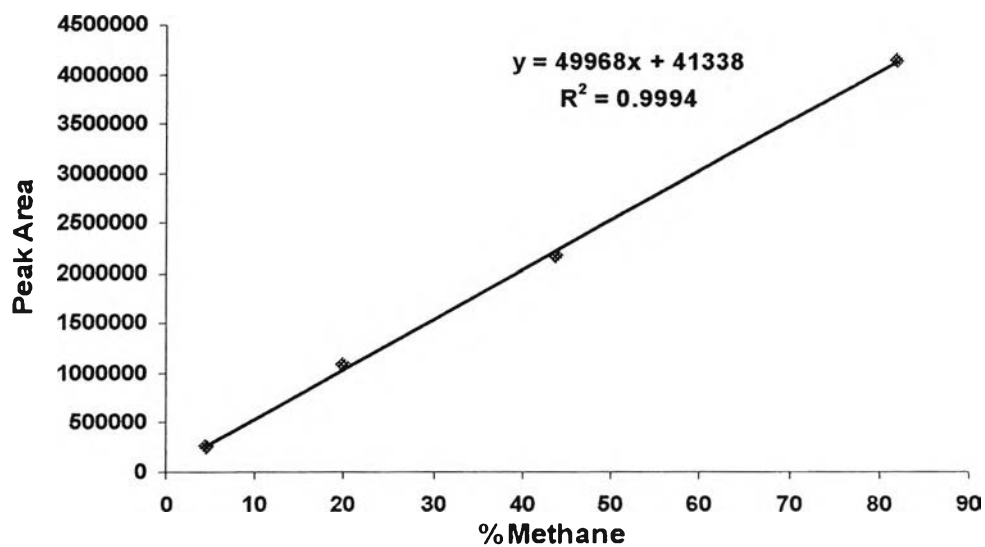


Figure G3 CH₄ calibration curve.

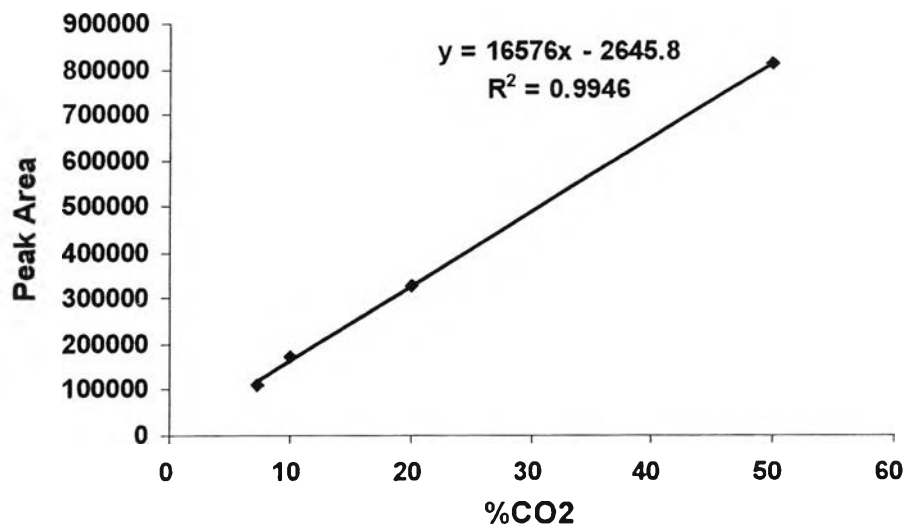


Figure G4 CO₂ calibration curve.

CURRICULUM VITAE

Name: Ms. Wanwanat Noisra

Date of Birth: March 6, 1983

Nationality: Thai

University Education:

2002-2006 Bachelor Degree of Chemical Engineering, Faculty of Engineering, Mahidol University, Nakornpratom, Thailand

Working Experience:

2005-2005 Position: Quality control intern
Company name: PTT Co.Ltd.

Presentations:

1. Noisra, W., Luengnaruemitchai, A., and Jitkarnka, S. (2007, October 29-30) Hydrogen Production from the Steam Reforming of Methane over Ni Supported on ZSM-5 Zeolite Catalysts at Thailand Chemical Engineering & Applied Chemistry Conference 2007, Chiang Mai, Thailand.

