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APPENDIX

APPENDIX A

Physical Properties of $NO-NH_3-O_2$ System

Table A1 Enthalpy of formation, ΔH_f^o , at 298 K, [KJ/mole]

component	ΔH_f^o,298
H_2O (g)	-242
NO (g)	+90
N_2O (g)	+82
NO_2 (g)	+33
NH_3 (g)	-46
N_2 (g)	0
O_2 (g)	0

From : G.H. Aylward & T.J.V. Findly, " SI Chemical Data",
John Wiley & Son Aust Pty. Ltd., 1974

Table A2 Gibbs free energy, ΔG_f^o , at 298 K, [kJ/mole]

component	ΔG_f^o,298
H_2O (g)	-229
NO (g)	+87
N_2O (g)	+103
NO_2 (g)	+51
NH_3 (g)	-16
N_2 (g)	0
O_2 (g)	0

Table A3 Molecular weight and density of gases in this work (density at S.T.P.)

component	M.W.	density, g/cm^3
He (g)	4.00	0.1769×10^{-3}
H_2O (g)	18.02	0.5863×10^{-3}
NO (g)	30.01	1.3400×10^{-3}
NH_3 (g)	17.03	0.7720×10^{-3}
N_2 (g)	28.02	1.2528×10^{-3}
O_2 (g)	32.00	1.4289×10^{-3}

From : G.H. Aylward & T.J.V. Findly, " SI Chemical Data", John Wiley & Son Aust Pty. Ltd., 1974

Table A4 Heat capacity, $C_p^o(A_i)$, of gas-component in reaction of $NO-NH_3-O_2$ system simplified as equation below

$$C_p^o(A_i) = a + bT + cT^2 \qquad [J/mole.K]$$

component	a	$bx10^2$	$cx10^5$
H_2O (g)	33.460	0.6880	0.7604
NO (g)	29.500	0.8188	-0.2925
N_2O (g)	37.660	4.1510	-2.6940
NO_2 (g)	36.070	3.9700	-2.8800
NH_3 (g)	35.150	2.9540	0.4421
N_2 (g)	29.000	0.2199	-2.8710
O_2 (g)	29.100	1.1580	-0.6076

From : G.H. Aylward & T.J.V. Findly, " SI Chemical Data", John Wiley & Son Aust Pty. Ltd., 1974

APPENDIX B

PARAMETERS DESCRIBING THE QUALITY OF CATALYST

B1 Equation for Finding Volume of Gases from Chromatogram

To find linear equations to express relation between volume of gases and peak area from chromatogram of gas chromatography, linear regression method is applied.

By means of linear regression, the relation between volume and peak area of each gas can be shown as equation below

$$V_i = A + BP_i$$

where A = constant term, (interception point)

B = regression coefficient, (slope)

V_i = volume of gas component i

P_i = peak area of gas component i

Table B1 Result of calculation to find relation between volume of each gas and peak area by means of linear regression.

Item	gas component				
	NO	NH_3	O_2	N_2	H_2O
Amt.of data	5	5	5	5	5
constant,A	0	0	0	0	0
slope, B	4.411×10^{-7}	3.035×10^{-7}	3.256×10^{-7}	1.792×10^{-9}	4.174×10^{-9}
error of A	5.991×10^{-5}	1.537×10^{-6}	1.103×10^{-7}	6.599×10^{-9}	6.921×10^{-9}
error of B	6.92×10^{-10}	9.673×10^{-9}	1.42×10^{-13}	1.90×10^{-15}	4.85×10^{-11}
R	0.999999	0.999998	0.999987	0.99999	0.999999

Remark : Amt. of data = amount of data

R = simple correlation coefficient

B2 Space Velocity, SV $[hr^{-1}]$

$$SV = \frac{\text{volumetic flow rate of gas passed through the reactor}}{\text{volume of catalyst}}$$

B3 Conversion of Gas Component i (as %), X_i

$$X_i = \frac{\text{converted reactant component i} \times 100}{\text{reactant component i in feed}}$$

B4 Sample of Calculation for Volume of Catalyst packing in Reactor, V_C

$$V_C = (\pi D_R^2)h/4$$

where D_R = inside diameter of reactor = 1.2 cm

h = height of catalyst in reactor = 0.15 cm

$$V_C = \pi x(1.2)^2 x 0.15/4 = 0.1696 \text{ cm}^3$$

B5 Sample of Calculation for Space Velocity

Data : Volumetric flow rate of feed gas at 1 atm and 32 ^oC is equal to 87 cm^3

Find the volumetric flow rate of feed gas at 1 atm and 100 ^oC (reaction temperature)

From ideal gas law and at the same pressure;

$$V_f = 87x(273+100)/(273+32)$$

$$= 106.4 \text{ cm}^3/\text{min} = 6384 \text{ cm}^3/\text{hr}.$$

From definition B2

$$SV = 6384 \text{ cm}^3/\text{hr} / 0.1696 \text{ cm}^3$$

$$= 37640 \text{ hr}^{-1}$$

B6 Sample of Calaulation for Concentration of NO and NH_3 in the Feed

At the same volumetric flow rate of the feed as condition in B5, the feed is composed of reactants as following data;

NO/He (5000 ppm NO)	=	20 cm^3/min
NH_3/He (5000 ppm NH_3)	=	20 cm^3/min
O_2	=	3 cm^3/min
He (carrier gas)	=	44 cm^3/min
Total volumetric flow rate	=	87 cm^3/min

Actual volumetric flow rate of NO in the feed

$= 5000 \times 10^{-6}\ cm^3_{NO}/cm^3_{NO/He} \times 20\ cm^3_{NO/He}/min$

$= 0.1\ cm^3_{NO}/min$

Actual volumetric flow rate of NH_3 in the feed

$= 5000 \times 10^{-6}\ cm^3_{NH3}/cm^3_{NH3/He} \times 20\ cm^3_{NH3/He}/min$

$= 0.1\ cm^3_{NH3}/min$

Concentration of NO in the feed

$= 0.1\ cm^3_{NO}/min\ /\ 87\ cm^3_{feed}/min$

$= 1.149 \times 10^{-3}\ cm^3_{NO}/cm^3_{feed} = 1149$ ppm

Concentration of NH_3 in the feed

$= 0.1\ cm^3_{NH3}/min\ /\ 87\ cm^3_{feed}/min$

$= 1.149 \times 10^{-3}\ cm^3_{NH3}/cm^3_{feed} = 1149$ ppm

Volume % of NO in the feed = (0.1x100)/87 = 0.115 %

Volume % of NH_3 in the feed = (0.1x100)/87 = 0.115 %

Volume % of O_2 in the feed = (3.0x100)/87 = 3.450 %

B7 Sample of Calculation for NO conversion

Experimental condition :

Reaction Temperature = 100 ^{o}C

Space velocity at 100 ^{o}C = 37640 hr^{-1}

The presence of O_2 in the feed = 3.45 volume %

Feed section : (at 32 ^{o}C and 1 atm)

Volumetric flow rate of the feed = 87 cm^3/min

Actual volumetric flow rate of NO in the feed = 0.1 cm^3/min

Effluent section : (Measuring at 1 atm and the temperature of the exist gas is equal to 36 ^{o}C)

Volumetric flow rate of effluent gas = 90 cm^3/min

Convert to volumetric flow rate at 32 ^{o}C and 1 atm

$$= 90x(273+32)/(273+36) = 88.35\ cm^3/min$$

Peak area of NO from 1 cm^3 of sampling gas = 994 unit

From calibration equation

$$V_{NO} = 4.4110x10^{-7} \times \text{Peak area of NO}$$

$$= 4.411ox10^{-7} \times 994$$

$$= 4.384x10^{-4}\ cm^3_{NO}/cm^3_{effluent\ gas}$$

Actual volumetric flow rate of NO in volumetric flow rate of effluent gas

$$= 4.384x10^{-4}\ cm^3_{NO}/cm^3_{effluent\ gas} \times 88.35\ cm^3_{effluent\ gas}/min$$

$$= 3.873x10^{-2}\ cm^3_{NO}/min$$

From definition B3, the conversion of NO = X_{NO}

$$X_{NO} = (0.1 - 3.873x10^{-2})x100 / 0.1$$

$$= 61.27\ \%$$

B8 Sample of Calculation for Percentage of Apparent N_2 in comparison with Theory

From the condition in B7

Peak area of N_2 in 1 cm^3 of sampling gas = 413075 unit

From calibration equation of N_2

$$V_{N2} = 1.7917 \times 10^{-9} \times \text{Peak area of } N_2$$

$$= 1.7919 \times 10^{-9} \times 412075$$

$$= 7.40106 \times 10^{-4} \ cm^3_{N2}/cm^3_{effluent\ gas}$$

Actual volumetric flow rate of N_2 in volumetric flow rate of effluent gas

$$= 7.40106 \times 10^{-4} \ cm^3_{N2}/cm^3_{effluent\ gas} \times 88.35 \ cm^3_{effluent\ gas}/min$$

$$= 6.538 \times 10^{-2} \ cm^3_{N2}/min$$

Reaction of $NO-NH_3-O_2$

$$4NO + 4NH_3 + O_2 \text{ -------> } 4N_2 + 6H_2O$$

From condition of B7, conversion of NO = 61.27 %

Assume the conversion of NO is followed to the above reaction.

From the stoichiometric coefficient of above reaction

Volume of disapparent NO from Rx^n = Volume of apparent N_2 from Rx^n

Volume of N_2 from Rx^n (theory) = 0.1×0.6127

$$= 6.127 \times 10^{-2} \ cm^3/min$$

Percentage of apparent N_2 in comparison with theory

$$= \frac{6.538 \times 10^{-2}}{6.127 \times 10^{-2}} \times 100 = 106.70 \ \%$$

Remark : Rx^n = reaction

APPENDIX C

EXPERIMENTAL DATA

Table C1 Activities of Pt-HY catalyst for NO-NH_3 reaction;

Composition of the feed: NO(0.119%)-NH_3(0.119%) (by volume) He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	27.64	29.38	1:1.063
	37640	25.67	28.34	1:1.104
	56460	23.67	27.32	1:1.154
	75280	20.20	25.48	1:1.173
200	18820	37.46	34.46	1:0.921
	37640	35.49	33.50	1:0.944
	56460	33.53	32.45	1:0.968
	75280	29.60	30.31	1:1.027
300	18820	41.38	36.54	1:0.883
	37640	39.90	35.75	1:0.896
	56460	37.45	34.46	1:0.921
	75280	34.54	32.94	1:0.955
400	18820	40.42	36.01	1:0.891
	37640	38.46	34.98	1:0.911
	56460	36.01	33.85	1:0.973
	75280	30.60	30.91	1:1.010

Remark : Pt-HY (Pt = 5 wt%)

Table C2 Activities of Cu-Pt-HY catalyst for $NO-NH_3$ reaction;

Composition of the feed: NO(0.119%)-NH_3(0.119%)(by volume) He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion of NO:NH_3 from reaction
		NO	NH_3	
100	18820	36.49	34.01	1:0.932
	37640	34.53	32.98	1:0.955
	56460	30.60	30.91	1:1.060
	75280	29.12	30.14	1:1.035
200	18820	47.23	39.58	1:0.838
	37640	43.35	37.54	1:0.866
	56460	40.42	36.05	1:0.892
	75280	38.94	35.24	1:0.906
300	18820	53.09	42.64	1:0.803
	37640	46.76	39.24	1:0.841
	56460	44.79	38.34	1:0.855
	75280	43.35	37.54	1:0.866
400	18820	50.16	41.08	1:0.819
	37640	44.31	37.66	1:0.859
	56460	43.34	37.53	1:0.866
	75280	40.31	35.96	1:0.892

Remark : Cu-Pt-HY (Cu = 4.9 wt%,Pt = 5 wt%)

Table C3 Activities of V_2O_5-TiO_2 No.1 catalyst for NO-NH_3 reaction;
Composition of the feed: NO(0.119%)-NH_3(0.119%)(by volume) He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	40.34	35.78	1:0.886
	37640	38.46	34.96	1:0.909
	56460	35.01	33.22	1:0.949
	75280	30.60	30.91	1:1.010
200	18820	48.72	40.34	1:0.828
	37640	45.49	38.83	1:0.848
	56460	43.16	35.95	1:0.834
	75280	38.23	35.29	1:0.923
300	18820	51.12	31.19	1:0.814
	37640	48.24	40.04	1:0.830
	56460	46.23	39.16	1:0.847
	75280	42.45	37.18	1:0.875
400	18820	48.61	40.25	1:0.828
	37640	46.76	39.28	1:0.841
	56460	45.27	38.48	1:0.852
	75280	41.38	36.78	1:0.887

Remark : V_2O_5-TiO_2 No.1 (V_2O_5 8.54 wt%)

Table C4 Activities of V_2O_5-TiO_2 No.2 catalyst for NO-NH_3 reaction;
Composition of the feed: NO(0.119%)-NH_3(0.119%)(by volume) He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	39.90	35.75	1:0.896
	37640	38.46	34.96	1:0.909
	56460	36.49	33.94	1:0.931
	75280	33.08	31.16	1:0.942
200	18820	48.79	40.30	1:0.826
	37640	44.79	38.26	1:0.972
	56460	43.34	37.53	1:0.866
	75280	39.41	35.46	1:0.901
300	18820	52.09	42.08	1:0.808
	37640	48.31	40.05	1:0.829
	56460	46.27	39.24	1:0.848
	75280	43.83	37.83	1:0.863
400	18820	51.12	41.71	1:0.814
	37640	47.42	41.68	1:0.721
	56460	44.31	38.06	1:0.859
	75280	42.46	37.14	1:0.874

Remark : V_2O_5-TiO_2 No.2 (V_2O_5 15.0 wt%)

Table C5 Activities of V_2O_5-TiO_2 No.3 catalyst for NO-NH_3 reaction;
Composition of the feed: NO(0.119%)-NH_3(0.119%) (by volume) He balance

T (^oC)	SV.(hr^{-1})	conversion; %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	40.53	35.95	1:0.887
	37640	38.53	35.06	1:0.909
	56460	35.34	33.43	1:0.946
	75280	33.16	32.16	1:0.971
200	18820	49.20	40.19	1:0.824
	37640	47.23	39.57	1:0.838
	56460	43.49	37.69	1:0.866
	75280	39.35	35.53	1:0.903
300	18820	54.06	43.14	1:0.798
	37640	50.16	41.11	1:0.819
	56460	46.31	39.13	1:0.845
	75280	41.38	36.41	1:0.883
400	18820	52.09	42.08	1:0.808
	37640	49.20	40.59	1:0.825
	56460	46.34	39.57	1:0.845
	75280	40.34	35.98	1:0.891

Remark : V_2O_5-TiO_2 No.3 (V_2O_5 25.61 wt%)

Table C6 Activities of pure V_2O_5 catalyst for NO-NH_3 reaction;
Composition of the feed: NO(0.119%)-NH_3(0.119%) (by volume) He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion of NO:NH_3 from reaction
		NO	NH_3	
100	18820	38.41	34.99	1:0.911
	37640	35.21	30.21	1:0.858
	56460	32.56	31.91	1:0.981
	75280	30.08	30.65	1:1.019
200	18820	41.86	36.79	1:0.879
	37640	39.83	35.76	1:0.897
	56460	36.56	33.96	1:0.929
	75280	32.89	32.13	1:0.977
300	18820	42.16	37.82	1:0.897
	37640	40.49	35.46	1:0.888
	56460	37.49	34.53	1:0.963
	75280	34.01	32.76	1:0.883
400	18820	41.38	36.53	1:0.951
	37640	35.86	34.07	1:0.954
	56460	34.53	32.80	1:0.962
	75280	30.08	30.62	1:1.018

Table C7 Activities of Pt-HY catalyst for $NO-NH_3-O_2$ reaction;

Composition of the feed: $NO(0.115\%)-NH_3(0.115\%)-O_2(3.45\%)$ (by volume)

He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of $NO:NH_3$ from reaction
100	18820	42.39	47.05	1:1.110
	37640	40.59	45.46	1:1.125
	56460	39.15	44.39	1:1.135
	75280	35.73	41.80	1:1.173
200	18820	53.67	55.76	1:1.039
	37640	52.35	53.20	1:1.056
	56460	47.49	51.01	1:1.074
	75280	44.73	44.53	1:0.996
300	18820	58.55	59.52	1:1.016
	37640	55.25	57.02	1:1.031
	56460	54.32	56.22	1:1.036
	75280	51.95	54.49	1:1.049
400	18820	57.08	59.13	1:1.023
	37640	54.76	56.62	1:1.034
	56460	52.85	55.16	1:1.044
	75280	50.48	53.41	1:1.058

Remark : Pt-HY (Pt = 5 wt%)

Table C8 Activities of Cu-Pt-HY catalyst for $NO-NH_3-O_2$ reaction;

Composition of the feed: NO(0.115%)-NH_3(0.115%)-O_2(3.45%)(by volume)

He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	54.15	56.13	1:1.036
	37640	49.48	52.48	1:1.062
	56460	46.51	50.23	1:1.080
	75280	44.71	48.96	1:1.095
200	18820	68.20	66.84	1:0.982
	37640	63.47	63.34	1:0.998
	56460	61.65	62.08	1:1.007
	75280	57.68	58.94	1:1.023
300	18820	73.00	70.66	1:0.968
	37640	67.80	66.72	1:0.984
	56460	66.39	65.73	1:0.990
	75280	62.47	62.67	1:1.003
400	18820	69.63	68.13	1:0.978
	37640	66.38	64.12	1:0.988
	56460	64.49	64.26	1:0.996
	75280	60.58	62.64	1:1.011

Remark : Cu-Pt-HY (Cu = 4.9 wt%,Pt = 5 wt%)

Table C9 Activities of V_2O_5-TiO_2 No.1 catalyst for NO-NH_3-O_2 reaction;
Composition of the feed: NO(0.115%)-NH_3(0.115%)-O_2(3.45%) (by volume)
He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	61.09	61.84	1:1.012
	37640	57.13	58.27	1:1.026
	56460	54.97	56.89	1:1.035
	75280	50.03	53.03	1:1.061
200	18820	67.61	66.79	1:0.989
	37640	65.65	65.19	1:0.993
	56460	63.54	63.47	1:0.999
	75280	60.09	60.69	1:1.012
300	18820	70.38	68.97	1:0.980
	37640	67.53	67.12	1:0.987
	56460	65.44	65.02	1:0.993
	75280	62.96	63.02	1:1.001
400	18820	68.08	67.79	1:0.987
	37640	63.77	63.70	1:0.999
	56460	62.59	62.79	1:1.003
	75280	60.58	61.24	1:1.011

Remark : V_2O_5-TiO_2 No.1 (V_2O_5 8.54 wt%)

Table C10 Activities of V_2O_5-TiO_2 No.2 catalyst for NO-NH_3-O_2 reaction;
Composition of the feed: NO(0.115%)-NH_3(0.115%)-O_2(3.45%) (by volume)
He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	63.91	63.97	1:1.001
	37640	59.97	60.81	1:1.014
	56460	56.41	58.02	1:1.028
	75280	51.95	54.49	1:1.049
200	18820	74.51	70.71	1:0.949
	37640	72.71	70.52	1:0.972
	56460	68.78	67.40	1:0.982
	75280	64.58	64.20	1:0.995
300	18820	78.21	75.00	1:0.959
	37640	75.91	73.10	1:0.963
	56460	72.08	70.13	1:0.973
	75280	66.28	65.62	1:0.990
400	18820	76.81	73.96	1:0.963
	37640	74.09	71.72	1:0.968
	56460	69.24	67.85	1:0.981
	75280	63.94	63.68	1:0.996

Remark : V_2O_5-TiO_2 No.2 (V_2O_5 15.0 wt%)

Table C11 Activities of V_2O_5-TiO_2 No.3 catalyst for NO-NH_3-O_2 reaction;
Composition of the feed: NO(0.115%)-NH_3(0.115%)-O_2(3.45%) (by volume)
He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of NO:NH_3 from reaction
100	18820	64.35	64.35	1:1.000
	37640	61.85	62.28	1:1.007
	56460	57.81	59.34	1:1.022
	75280	54.81	56.73	1:1.035
200	18820	75.42	72.86	1:0.966
	37640	71.29	69.51	1:0.976
	56460	69.23	67.84	1:0.981
	75280	64.39	64.13	1:0.996
300	18820	80.03	76.42	1:0.955
	37640	75.97	72.93	1:0.963
	56460	73.48	71.20	1:0.969
	75280	69.15	67.76	1:0.981
400	18820	78.21	75.06	1:0.959
	37640	74.56	72.17	1:0.968
	56460	72.08	70.13	1:0.973
	75280	67.22	66.35	1:0.987

Remark : V_2O_5-TiO_2 No.3 (V_2O_5 25.61 wt%)

Table C12 Activities of pure V_2O_5 catalyst for $NO-NH_3-O_2$ reaction;
Composition of the feed: $NO(0.115\%)-NH_3(0.115\%)-O_2(3.45\%)$ (by volume)
He balance

T (^oC)	SV.(hr^{-1})	conversion, %		mole ratio of conversion
		NO	NH_3	of $NO:NH_3$ from reaction
100	18820	63.45	63.63	1:1.003
	37640	59.01	60.07	1:1.018
	56460	54.48	56.49	1:1.038
	75280	52.40	54.92	1:1.048
200	18820	74.98	72.51	1:0.967
	37640	72.22	68.10	1:0.943
	56460	68.78	67.61	1:0.982
	75280	59.61	60.50	1:1.014
300	18820	77.29	74.35	1:0.962
	37640	72.22	70.27	1:0.973
	56460	68.78	67.54	1:0.983
	75280	63.45	63.44	1:0.999
400	18820	74.51	72.20	1:0.969
	37640	67.53	66.65	1:0.987
	56460	64.49	64.26	1:0.996
	75280	58.93	59.93	1:1.017

AUTOBIOGRAPHY

Mr. Chatchai Kunyawut was born on November 19, 1965, in Nakornrachasi-ma, Thailand. He received his Bachelor Degree in Industrial Chemistry from Chiangmai University in 1987.