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APPENDICES

Appendix A Calculation of NO conversion and N_2/N_2O selectivity.

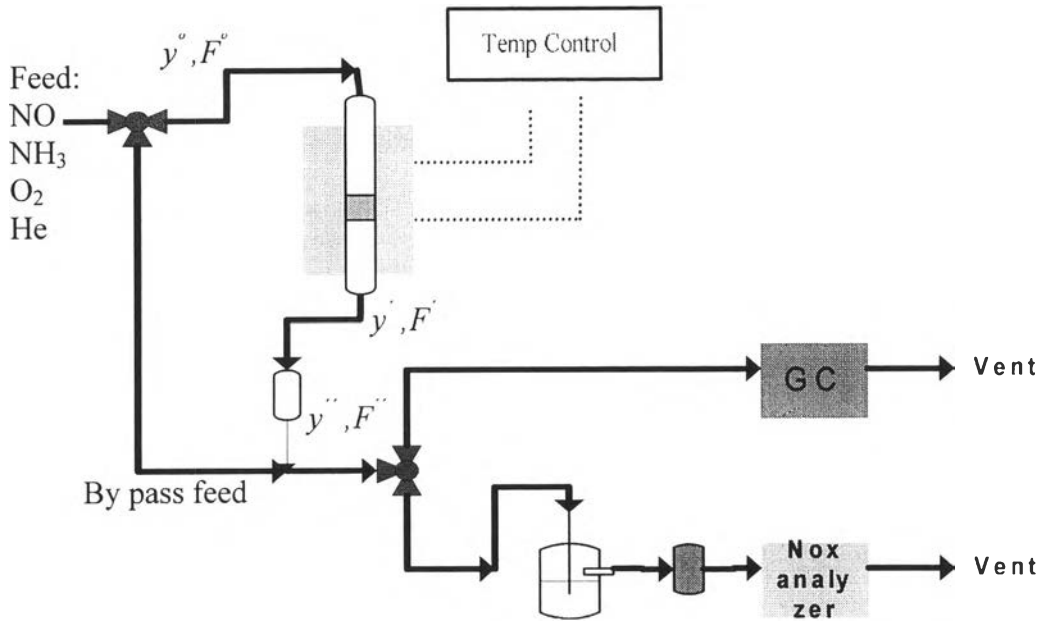

Figure A1 SCR process flow diagram (for calculation).

F^o	=	Flow rate in to reactor (ml/min)
y^o	=	Volume fraction of NO in F^o
F'	=	Flow rate out from reactor (ml/min)
y'	=	Volume fraction of NO in F'
F''	=	Flow rate after water trapping (ml/min)
y''	=	Volume fraction of NO in F''

1. <u>NO Conversion</u>

$$\text{NO Conversion} = \frac{y^{\circ}_{NO}F^{\circ} - y'_{NO}F'}{y^{\circ}_{NO}F^{\circ}}$$

2. Volume fraction of N_2O

Volume fraction of N_2O in product can be determined by O_2 balance

O_2 balance

In reactor = Out reactor

$$2y^{\circ}_{O_2}F^{\circ} + y^{\circ}_{NO}F^{\circ} = 2y'_{O_2}F' + y'_{NO}F' + 2y'_{NO_2}F' + y'_{N_2O}F' + y'_{H_2O}F'$$

$$y'_{N_2O} = 2y^{\circ}_{O_2}\frac{F^{\circ}}{F'} + y^{\circ}_{NO}\frac{F^{\circ}}{F'} - 2y'_{O_2} + y'_{NO} + 2y'_{NO_2} + y'_{H_2O}$$

3. Volume fraction of H_2O

If water amount produced per run = A g

Testing temperature = T K

Running time = t min

Volume of water produced = $\frac{\text{A g* 0.0821 L/mol K* T K}}{\text{18 g/mol* 1 atm}}$

= B liter

Volume of water produced per min = $\frac{\text{B g* 1000 ml}}{\text{t min}}$

= V ml/s

y H_2O = V / F′

4. N_2/NO, N_2O/NO, and H_2O/NO selectivity

N_2/NO Selectivity = $\dfrac{y'_{N_2}}{y^{\circ}_{NO}\dfrac{F^{\circ}}{F'} - y'_{NO}}$

N_2O/NO Selectivity = $\dfrac{y'_{N_2O}}{y^{\circ}_{NO}\dfrac{F^{\circ}}{F'} - y'_{NO}}$

H_2O/NO Selectivity = $\dfrac{y'_{H_2O}}{y^{\circ}_{NO}\dfrac{F^{\circ}}{F'} - y'_{NO}}$

5. N_2/N_2O Selectivity

N_2/N_2O Selectivity = $\dfrac{y'_{N_2}}{y'_{N_2} - y'_{N_2O}}$

Appendix B Raw data.

Table B1 The d-spacing of all pillared-clays

Types of pillared clay	2θ (°)	d-spacing (Å)
1. DA-clay		
- Dried 110°C	2.65	32.90
- Calcined 250°C	3.14	28.11
- Calcined 300°C	3.78	23.36
- Calcined 350°C	11.26*	7.86*
- Calcined 400°C	11.28*	7.86*
- Calcined 500°C	42.62*	2.11*
2. PW_{12}-clay		
- Dried	6.02	14.67
- Calcined 250°C	6.02	14.67
- Calcined 350°C	11.86*	7.46*
- Calcined 500°C	42.92*	2.11*
3. SiW_{12}-clay		
- Dried	6.02	14.67
- Calcined 250°C	6.02	14.67
- Calcined 350°C	8.92*	9.91*
- Calcined 500°C	42.74*	2.11*

*data taken from the first peak observed

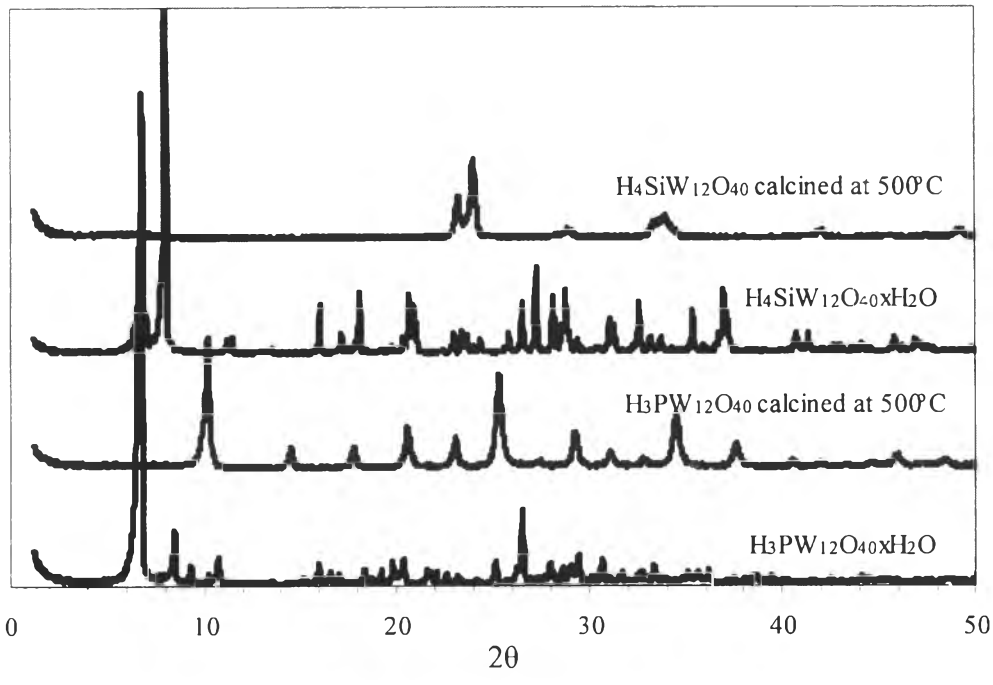

Figure B1 XRD patterns of fresh $H_3PW_{12}O_{40}$, $H_3PW_{12}O_{40}$ calcined at 500°C, fresh $H_4SiW_{12}O_{40}$, and $H_4SiW_{12}O_{40}$ calcined at 500°C.

Table B2 BET characterization

Catalyst	Surface area (m^2/g)	Pore volume (cc/g)	Average pore diameter (Å)
1. DA-clay			
- Calcined 350°C	48.71	0.14	114.60
- Calcined 500°C	125.60	0.26	82.68
2. PW_{12}-clay			
- Calcined 350°C	78.99	0.25	127.40
- Calcined 500°C	107.90	0.24	90.53
3. SiW_{12}-clay			
- Calcined 350°C	77.87	0.17	86.52
- Calcined 500°C	116.30	0.18	109.2

Table B3 SCR activity test of 0.1 g DA-clay calcined at 250°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	1.6	99.8	0.2	0	0
200	1.5	99.8	0.2	0	0
250	2.2	99.8	0.2	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B4 SCR activity test of 0.1 g PW_{12}-clay calcined at 250°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	5.6	99.4	0.6	0	0
200	5.1	99.6	0.4	0	0
250	6.2	99.9	0.1	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B5 SCR activity test of 0.1 g SiW_{12}-clay calcined at 250°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	5.9	99.9	0.1	0	0
200	6.3	99.7	0.3	0	0
250	6.3	99.8	0.2	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B6 SCR activity test of 0.1 g DA-clay calcined at 350°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	6.2	99.8	0.2	0	0
200	6.2	99.5	0.5	0	0
250	6.2	99.7	0.3	0	0
300	6.6	99.7	0.3	0	0
350	6.6	99.5	0.5	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B7 SCR activity test of 0.1 g PW_{12}-clay calcined at 350°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	6.1	99.9	0.1	0	0
200	6.2	99.9	0.1	0	0
250	6.3	99.7	0.3	0	0
300	6.0	99.9	0.1	0	0
350	6.2	99.9	0.1	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B8 SCR activity test of 0.1 g SiW_{12}-clay calcined at 350°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	6.2	99.6	0.4	0	0
200	6.3	99.4	0.6	0	0
250	6.2	99.8	0.2	0	0
300	6.3	99.7	0.3	0	0
350	6.7	99.9	0.1	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B9 SCR activity test of 0.1 g DA-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	10.3	99.7	0.3	0	0
200	11.8	99.8	0.2	0	0
250	11.8	99.8	0.2	0	0
300	14.7	99.6	0.4	0	0
350	16.2	99.8	0.2	0	0
400	19.1	99.4	0.6	0	0
450	20.6	99.6	0.4	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B10 SCR activity test of 0.1g PW_{12}-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	11.8	99.7	0.3	0	0
200	13.2	99.7	0.3	0	0
250	13.4	99.8	0.2	0	0
300	15.0	99.7	0.3	0	0
350	19.0	99.6	0.4	0	0
400	26.5	99.4	0.6	0	0
450	31.2	99.4	0.6	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B11 SCR activity test of 0.1 g SiW_{12}-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	10.3	99.5	0.5	0	0
200	11.8	99.4	0.6	0	0
250	11.8	99.4	0.6	0	0
300	17.9	99.8	0.2	0	0
350	18.5	99.3	0.7	0	0
400	22.1	99.4	0.6	0	0
450	26.5	99.3	0.7	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B12 SCR activity test of 0.1 g DA-clay calcined at 900°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	5.7	99.9	0.1	0	0
200	7.6	99.6	0.4	0	0
250	7.6	99.7	0.3	0	0
300	8.7	99.5	0.5	0	0
350	9.4	99.8	0.2	0	0
400	11.8	99.6	0.4	0	0
450	15.1	99.9	0.1	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B13 SCR activity test of 0.1 g PW_{12}-clay calcined at 900°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	13.2	99.4	0.6	0	0
200	13.2	99.4	0.6	0	0
250	14.7	99.5	0.5	0	0
300	16.3	99.8	0.2	0	0
350	17.6	99.6	0.4	0	0
400	20.7	99.2	0.8	0	0
450	26.0	99.4	0.6	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B14 SCR activity test of 0.1 g SiW_{12}-clay calcined at 900°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	7.1	99.4	0.6	0	0
200	7.1	99.9	0.1	0	0
250	8.3	99.7	0.3	0	0
300	9.9	99.8	0.2	0	0
350	10.6	99.2	0.8	0	0
400	15.9	99.1	0.9	0	0
450	17.3	99.1	0.9	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B15 SCR activity test of 0.1 g Fe-DA-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	11.8	99.7	0.3	0	0
200	12.4	99.4	0.6	0	0
250	12.4	99.7	0.3	0	0
300	14.7	99.7	0.3	0	0
350	19.1	99.6	0.4	0	0
400	26.5	99.5	0.5	0	0
450	30.9	99.2	0.8	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B16 SCR activity test of 0.1g Fe-PW_{12}-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	11.8	99.8	0.2	0	0
200	13.2	99.8	0.2	0	0
250	13.7	99.8	0.2	0	0
300	15.7	99.5	0.5	0	0
350	20.6	99.6	0.4	0	0
400	30.9	99.7	0.3	0	0
450	39.9	99.4	0.6	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B17 SCR activity test of 0.1 g Fe-SiW_{12}-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	11.8	99.4	0.6	0	0
200	11.8	99.9	0.1	0	0
250	11.8	99.5	0.5	0	0
300	18.2	99.7	0.3	0	0
350	19.1	99.7	0.3	0	0
400	25.1	99.6	0.4	0	0
450	29.4	99.4	0.6	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B18 SCR activity test of 0.2 g Fe-DA-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	15.6	99.6	0.4	0	0
200	16.5	99.6	0.4	0	0
250	17.1	99.7	0.3	0	0
300	19.3	99.6	0.4	0	0
350	24.1	99.0	1.0	0	0
375	26.5	99.0	1.0	0	0
400	31.2	99.1	0.9	0	0
450	35.6	99.0	1.0	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B19 SCR activity test of 0.2g Fe-PW_{12}-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	16.0	99.8	0.2	0	0
200	17.9	99.8	0.2	0	0
250	18.4	99.1	0.9	0	0
300	22.4	99.1	0.9	0	0
350	26.6	99.3	0.7	0	0
375	29.1	99.2	0.8	0	0
400	36.9	99.0	1.0	0	0
450	46.3	99.2	0.8	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

Table B20 SCR activity test of 0.2 g Fe-SiW_{12}-clay calcined at 500°C for 12 hours

Temperature (°C)	NO Conversion (%)	Selectivity (%)			
		N_2[a]	N_2O[b]	NO_2	H_2O
150	15.6	99.7	0.3	0	0
200	16.0	99.7	0.3	0	0
250	16.0	99.2	0.8	0	0
300	23.1	99.4	0.6	0	0
350	24.6	99.3	0.7	0	0
375	26.9	99.3	0.7	0	0
400	30.4	99.3	0.7	0	0
450	34.3	99.0	1.0	0	0

[a]N_2/N_2O selectivity [b]N_2O/N_2 selectivity

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