Catalytic property of Pt/AlSBA-15 in selective catalytic reduction of NO

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Pt-supported on mesoporous SBA-15 catalysts (Pt/SBA-15) have been investigated for selective catalytic reduction of NO by propene in excess oxygen. Catalytic activity of Pt/SBA-15 can be improved by the post-synthesis modification of Al into SBA-15. The resulting material (AlSBA-15) retains a hexagonal order and physical properties of the parent SBA-15 and shows new acid sites. Increased acid sites are confirmed by NH_3-TPD. Medium acid site appeared at 200 °C and broad strong acid site appeared above 250 °C. A certain degree of support acidity of Pt/AlSBA-15 increased the NO conversion, but when there are too many acid sites, carbon deposition becomes extensive and lead to decrease the NO conversion. The influence of different Pt loadings on the catalytic properties was also investigated.

KEY WORDS: Pt/AlSBA-15; post-synthesis modification; support acidity; carbon deposition.

1. Introduction

Selective catalytic reduction (SCR) of NO in lean-burn diesel engine exhausts is a world wide challenging research subject that has been extensively studied due to the increasingly severe emission limitation, which culminated in an agreement in Kyoto. Nitrogen oxides, formed during combustion processes in power plants, wastes incinerators and diesel engines, are the major air pollutants leading to the formation of photochemical smog and acid rain. A large number of catalysts for NO reduction have been screened within last three decades, zeolite based catalysts (Cu/ZSM-5, Pt/ZSM-5) were founded as active catalysts in lean burn condition. Particularly, Pt/ZSM-5 catalyst was reported to be very active for SCR at low temperature [1–5] and showed a high stability in the presence of H_2O and SO_2 [6, 7]. In addition, supported Pt catalysts have shown high activity in real exhaust gas treatment application and appear to be promising candidates for the SCR of NO.

It has also been reported that different catalyst supports have remarkable effects on the activity of the resulting catalyst for NO reduction [8]. Chen and Kawi [9] reported that the NO reduction activity of aluminum containing Pt/MCM-41 has shown higher NO removal performance than that of Pt/MCM-41 and hydrothermal stability in the presence of steam and Woo and co-workers [10, 11] also reported that Pt/V/MCM-41 showed high NO reduction activity in the presence of SO_2. These results draw our attention to investigate the use of other material as the catalyst supports for NO reduction. Even though MCM-41 has a high thermal stability, it loses its structure during the exposure to high-temperature steam and boiling water [12]. The collapse of the structure has limited the applications of MCM-41 especially in the catalytic reactions involving high-temperature steam.

SBA-15, ordered honeycomb mesostructured member of the M41S family, has been attracted much interest because of their potential application in catalytic reactions or separations involving bulky molecules [13–16]. It shows different topologies depending on the nature of templating surfactants, the composition, the pH and the temperature of the synthesis mixture. Although MCM-41 has the similar property to SBA-15, SBA-15 is believed to be a very promising structure because of the higher thickness of its walls, which improve its hydrothermal stability [17]. However, this pure siliceous mesoporous material is lack of acid sites and acidity, therefore incorporation of aluminum into SBA-15 was performed in order to create acid sites [18–21]. With the presence of acid sites, the performance of NO removal will be enhanced because the acidic property of support material plays a crucial role in $DeNO_x$ process.

In this work, the Pt-containing catalysts were prepared by using the mesoporous silica SBA-15 and AlSBA-15 as support materials and tested for the SCR of NO with C_3H_6. The influence of different metal loadings as well as acidic property of support on the catalytic behavior of sample catalysts was investigated.

2. Experimental

2.1. Preparation of catalysts

Tetraethyl orthosilicate (TEOS) (Aldrich 99%), Pluronic P123 ($EO_{20}PO_{70}EO_{20}$; average molecular weight (M_{av}) = 5800) were purchased from Aldrich and

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BASF, respectively. The synthesis was carried out in a similar manner as reported in literature [17]. In a typical preparation process, a homogeneous mixture, comprising 10.0 g of Pluronic P123 was dissolved in 1.6 M HCl solution, heated at 35 °C during stirring, and then 22.0 g of TEOS was added to the mixture. This mixture was kept at 35 °C for 24 h and heated to 100 °C for 12 h. After reaction, the resulting gel was dried at 110 °C and added the EtOH + HCl solution (EtOH : HCl = 600 mL : 2 g) to remove the surfactant with vigorous stirring. Synthesized SBA-15 was separated by filtration, washed several times with EtOH and dried at room temperature in a vacuum oven. Then, the sample powder was heated to 500 °C at a rate of 1 °C min^{-1} and calcined at 500 °C for 3 h. The addition of aluminum ions to SBA-15 was carried out by post-synthesis modification method. $Al(NO_3)_3 \cdot 9H_2O$ was used as a precursor of aluminum. This metal precursor dissolved in EtOH + H_2O solution (4 mL + 21 mL) was impregnated to 1.5 g of SBA-15, precalcined at 400 °C, leading to Si/Al ratio = 29. This slurry was heated at 60 °C under stirring followed by using rotary evaporator to remove EtOH + H_2O solution. After removing the solution, the sample powder was dried at 110 °C and calcined in air at 500 °C for 3 h.

About 0.1, 0.6, 1, and 10 wt.% Pt/AlSBA-15 catalysts were prepared by incipient wetness method, using hexa-chloro platinic acid as a Pt precursor and 0.1, 0.6, and 1 wt.% Pt/SBA-15 was also prepared for comparison. The concentration of the Pt and Al was determined by inductively coupled plasma-atomic emission spectroscopy measurement using Perkin-Elmer Optima 3000.

2.2. Characterization of catalysts

2.2.1. Temperature programmed desorption of ammonia (NH_3-TPD)

The acidity of Pt/SBA-15 and Pt/AlSBA-15 was measured by NH_3-TPD analysis. About 100 mg of the sample was placed in a quartz tubular reactor, where the samples were activated at 500 °C in air for 2 h followed by outgassing for 30 min in a flow of helium (50 mL/min). Subsequently, the samples were cooled down to 35 °C and saturated for about 15 min in 10% NH_3/He (50 mL/min), Then, the samples were purged in the He flow until constant baseline level was attained. Desorption was carried out with a linear heating rate (10 °C/min) in a flow of He (50 mL/min) and NH_3-TPD spectra were obtained by thermal conductivity detector (TCD).

2.2.2. Temperature programmed oxidation (TPO)

Temperature programmed oxidation (TPO) was carried out with 100 mg of used 1 wt.% Pt/AlSBA-15 of various Si/Al ratio at a ramp rate of 5 °C/min under a 10% O_2/He flow of 30 mL/min. Formation of CO_2 during the TPO was confirmed by gas chromatograph.

2.2.3. Small angle X-ray diffraction scattering (SAXS)

Small angle XRD patterns were recorded for prepared SBA-15 and AlSBA-15 powder in order to confirm the hexagonal structure of samples. Rigaku miniflex X-ray diffractometer with nickel filtered Cu Kα radiation (λ = 1.54056) was adopted in the 2θ range of 1–2.5°.

2.2.4. BET analysis for surface area, pore volume, and CO chemisorption for metal dispersion

Nitrogen physisorption measurements were performed at 77 K on a Micrometrics ASAP 2010 apparatus. Prior to analysis about 300 mg of sample was dehydrated at 100 °C for 1 h and then degassed at 120 °C for 4 h. The pore size distribution from corresponding isotherms were obtained using Horvath–Kawazoe (HK) method for micropore range (0.5–1.7 nm) and Barrett–Joyner–Halenda (BJH) method for mesopore range (> 1.7 nm). Catalyst dispersion was determined by CO chemisorption at 308 K in Micrometrics ASAP 2020 equipment. Previously, the samples were *in situ* reduced under hydrogen at 623 K for 3 h. Carbon monoxide isotherms were considered until 450 mmHg and the value for platinum dispersion were calculated by assuming a stoichiometry (CO : Pt) of 1 : 1.

2.2.5. TEM analysis

A FEI FE-TEM (Technai G2 F3 S-Twin, The Netherland), operating at 300 kV, was used to obtain the HRTEM images of the catalysts. Samples were mounted on a copper grid-supported carbon film by placing a few droplets of an ultrasonically dispersed suspension of samples in ethanol on the grid, followed by drying at ambient conditions.

2.3. Catalytic activity test

The catalytic activities of all the catalysts were evaluated by using a plug flow quartz reactor (o.d. 1/4 in.). The total gas flow rate was 140 mL/min and 100 mg of catalyst was placed in the reactor. The composition of reactant gas mixture consisted of 2000 ppm of NO, 2700 ppm of C_3H_6, 3 vol.% of O_2 and He balance. The product gas mixture was analyzed by a GC (Donam instrument, Molecular sieve 5A column for N_2 and CO, and Porapak Q column for N_2O, CO_2, and C_3H_6) at desired temperature and the concentration of NO of effluent gas was detected by NO_x analyzer.

3. Results and discussion

Small-angle XRD patterns of the prepared SBA-15 and AlSBA-15 (Si/Al = 29) showed typical mesoporous structures in accordance with those reported in literature [17] as shown in figure 1. These materials exhibits one strong reflection (100) at $2\theta \asymp 1°$ and two weaker peaks
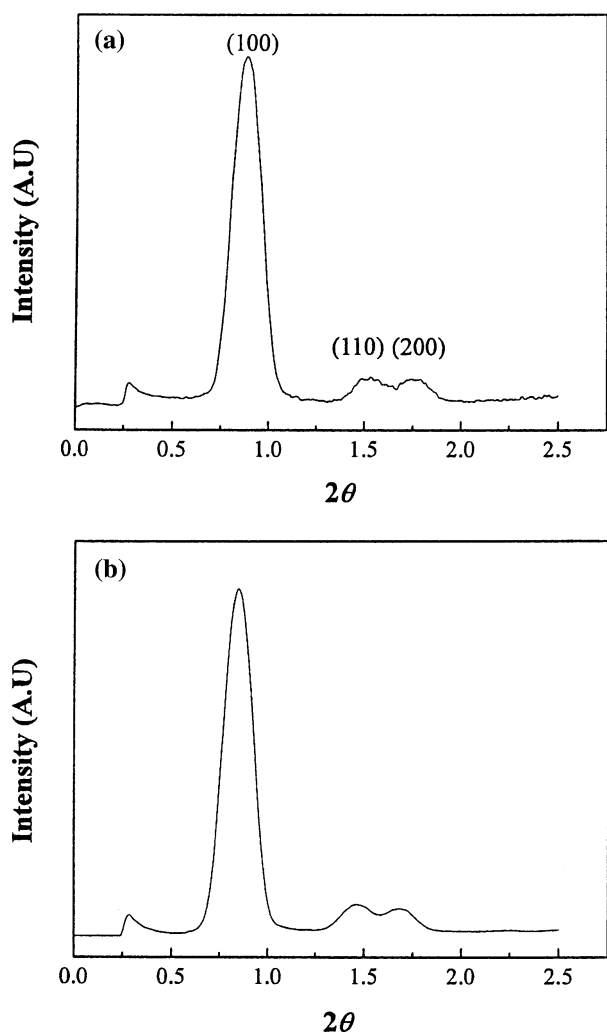
Figure 1. SAXS patterns of SBA-15 (a) and AlSBA-15 (b).

(110), (200) at higher 2θ, associated with the hexagonal symmetry and likewise characteristic of the hexagonal ordered structure.

Table 1 reports the physical characteristics of the support materials and the supported metal catalysts. It is apparent that the loading of active component has a notable impact on the surface area of mesoporous SBA-15 and AlSBA-15. The specific surface area and pore volume of SBA-15 and AlSBA-15 dropped appreciably with metal loading, interestingly, average pore size increased. The large pores of SBA-15 and AlSBA-15 can provide more space for access of more active components, producing a decrease in surface area and pore volume. On the other hand, some micropores were preferably blocked by metal particle, and the apparent average pore size of SBA-15 and AlSBA-15 increased with metal loading. Surface area and pore volume of AlSBA-15 is lower than that of the corresponding SBA-15, signifying that the Pt dispersion of supported AlSBA-15 catalysts is slightly lower than that of supported SBA-15 catalysts. Surface area, pore volume, and pore diameter of 10 wt.% Pt/AlSBA-15 are very low, which means excess amount of Pt particle blocked the micro and mesopores of AlSBA-15.

We used the TEM technique to investigate the Pt particle size and topology of catalysts. As shown in figure 2, the Pt particles are spherical and well dispersed on each catalyst. TEM images show that dispersion level of 1 wt.% Pt/SBA-15 and Pt/AlSBA-15 catalysts is similar in spite of different aluminum contents, resulting 29% and 23% of Pt dispersion, respectively. The mean particle size was approximately 6 nm for Pt metal particles for each catalyst.

The catalytic performance of 0.1–1 wt.% Pt/SBA-15 for the reduction of NO with C_3H_6 and turnover frequencies (TOF) are shown in table 2. The TOF for the conversion of NO, defined as the number of NO molecules converted per surface Pt atom per second. At lower temperature (below 200 °C), NO conversion and C_3H_6 conversion were small. With increasing temperature, NO reduction was found to increase at first, passing through maximum, then decreased at high temperatures. The maximum NO conversion appeared at the temperature at which propylene conversion reached above 92%. In all cases the reduction of NO begins with the oxidation of propene. These two conversions tend to rise together until the NO reduction reaches a maximum close to the temperature at which the propene oxidation is complete. Similar to other platinum based catalysts reported in the many literatures, the formation of nitrous oxides was more than that of nitrogen because platinum is a poor N_2O decomposition catalyst [22, 23].

Table 1
Specific surface area, pore size, volume and metal dispersion of Pt catalysts supported on SBA-15 and AlSBA-15 (Si/Al = 29)

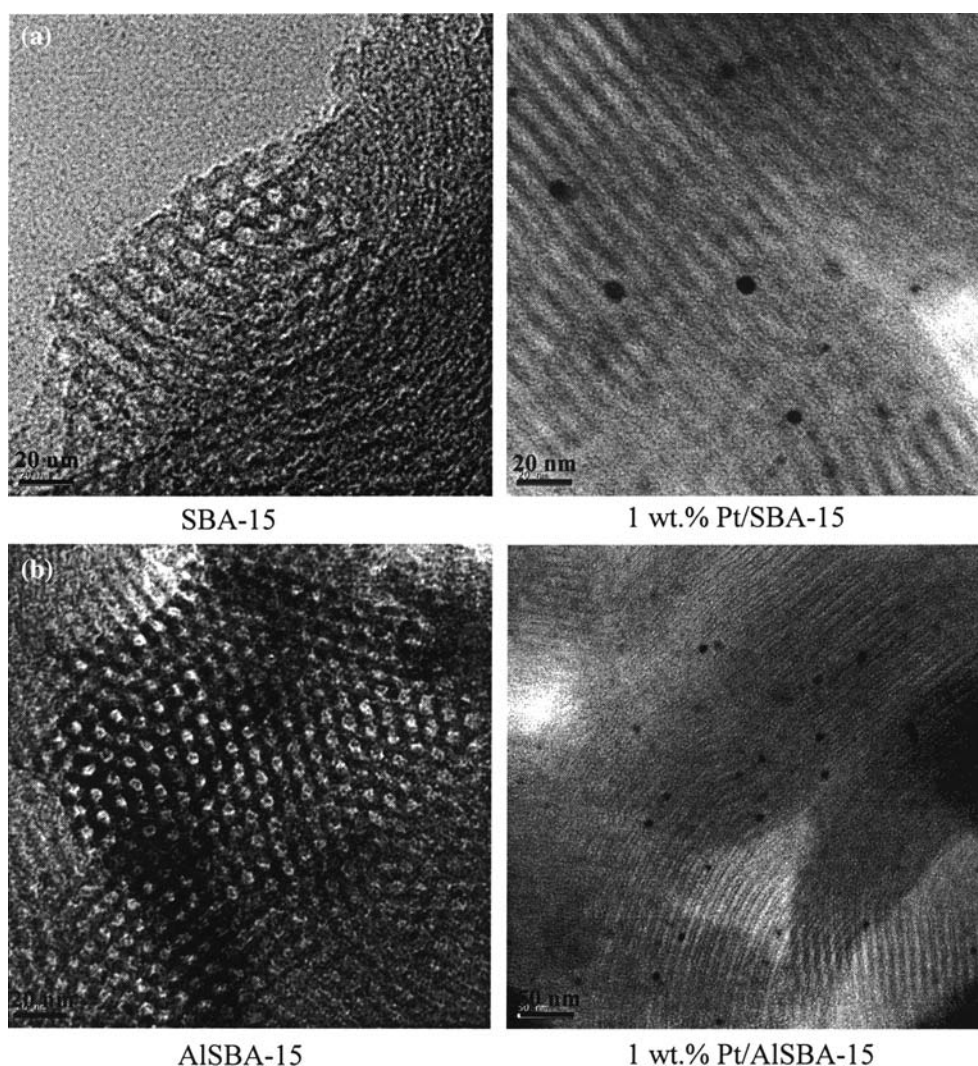| Catalyst	S_{BET} (m^2/g)	Pore volume (cm^3/g)	Pore diameter (Å)	Dispersion (%)
SBA-15	782	1.04	51.5	–
0.1 wt.% Pt/SBA-15	683	0.96	53.9	100
0.6 wt.% Pt/SBA-15	657	0.93	54.0	43.5
1 wt.% Pt/SBA-15	644	0.92	54.2	29.4
AlSBA-15	558	0.79	54.6	–
0.1 wt.% Pt/AlSBA-15	473	0.69	56.3	100
0.6 wt.% Pt/AlSBA-15	445	0.66	56.7	35.3
1 wt.% Pt/AlSBA-15	418	0.65	56.9	22.5
10 wt.% Pt/AlSBA-15	265	0.51	52.5	5.5

Figure 2. TEM image of catalysts: (a) SBA-15 and Pt/SBA-15; (b) AlSBA-15 and Pt/AlSBA-15 (29).

When the platinum amount increased from 0.1 to 1 wt.%, the peak of NO conversion temperature slightly decreased from 300 to 250 °C. At high temperature, the decrease in NO conversion was due to the oxidation of C_3H_6 by oxygen. With the increase of platinum amount, the maximum TOF was found to decrease. The steady state NO reduction rate (mmol/g h) was calculated as [24].

$$\text{NO reduction rate} = -d[\text{NO}]/dt = F_0X/\omega$$

where F_0 is inlet molar flow rate of NO, X is NO conversion, and ω is weight of the catalyst.

Table 3 gives the catalytic performance of 0.1, 0.6, 1 and 10 wt.% Pt/AlSBA-15 for the reduction of NO with C_3H_6 and TOF. Compared with table 1, one can see that Pt catalysts supported on AlSBA-15 prepared in this study are more active than that of SBA-15 catalysts. The maximum NO conversion of 0.1, 0.6, 1 wt.% Pt/SBA-15 is 27%, 49%, and 59%, but is increased to 35%, 54%, and 65% on corresponding Pt supported on

AlSBA-15. The calculated NO reduction rate on 0.1–1 wt.% Pt/AlSBA-15 were 1.8–3.4 mmol/g h at 250 or 300 °C, which was higher than 1.4–3.2 mmol/g h obtained from corresponding Pt/SBA-15 catalysts. The relationship between the NO reduction and the propene oxidation is largely responsible for the trend in NO conversion with Pt metal loading. As the metal loading is increased, the activity per gramme of catalyst increases and the oxidation of propene sets in progressively lower temperatures. Thus for 10 wt.% Pt/AlSBA-15 the reaction starts below 200 °C and peaks at around 250 °C, whereas with 0.1 wt.% Pt/AlSBA-15 catalyst there is no detectable conversion below 250 °C and peak is at about 300 °C. It is notable that N_2O formation of Pt/AlSBA-15 catalysts is lower than that of Pt/SBA-15. The N_2 selectivity at the maximum NO reduction increased from 36% to 45% at 1 wt.% Pt/AlSBA-15 and over 50% with increasing reaction temperature. Similar trend was found at 0.1 and 0.6 wt.% of Pt/AlSBA-15, while the N_2 selectivity of 10 wt.%

Table 2
Catalytic performance of Pt/SBA-15 for selective catalytic reduction of NO with C_3H_6 at different temperatures

Catalyst	Temp. (°C)	NO conv. to N_2 (%)	NO conv. to N_2O (%)	C_3H_6 conv. (%)	NO conv. (%)	NO reduction rate (mmol/g h)	TOF/10^{-3} (s^{-1})
0.1 wt.% Pt/SBA-15	200	0	0	0	0	0	0
	250	0	0	9.8	0	0	0
	300	10	17.5	92.4	27.5	1.4	75.9
	350	7.1	12.4	100	19.5	1.0	54.3
	400	6.5	8.8	100	15.3	0.8	43.1
	450	0	0	100	0	0	0
	500	0	0	100	0	0	0
0.6 wt.% Pt/SBA-15	200	6.2	0	0	6.2	0.3	7.3
	250	7.1	11.5	14.5	18.6	1.0	24.3
	300	18.4	30.4	100	48.8	2.6	63.3
	350	12.3	18.2	100	30.5	1.6	38.9
	400	6.5	10.3	100	16.9	0.9	21.9
	450	0	7.2	100	7.2	0.4	9.7
	500	0	0	100	0	0	0
1.0 wt.% Pt/SBA-15	200	11.2	0	11.6	11.2	0.6	11.1
	250	21.4	37.9	100	59.3	3.2	59.2
	300	18.7	22.8	100	41.5	2.2	40.4
	350	13.5	16.3	100	29.8	1.6	29.4
	400	6.2	9.5	100	15.7	0.8	14.7
	450	0	7.5	100	7.5	0.4	7.3
	500	0	0	100	0	0	0

Pt/AlSBA-15 was maintained below 45% within whole temperature range. This suggests that a certain amount of Pt loading is favorable for N_2 formation. The higher catalytic activity of Pt/AlSBA-15 and N_2 selectivity is suggested to be contributed by surface acidity originated from aluminum insertion. Similar results for

Table 3
Catalytic performance of Pt/AlSBA-15 for selective catalytic reduction of NO with C_3H_6 at different temperatures

Catalyst	Temp. (°C)	NO conv. to N_2 (%)	NO conv. to N_2O (%)	C_3H_6 conv. (%)	NO conv. (%)	NO reduction rate (mmol/g h)	TOF/10^{-3} (s^{-1})
0.1 wt.% Pt/AlSBA-15	200	0	0	0	0	0	0
	250	0	0	7.5	0	0	0
	300	15.4	19.8	89.4	35.2	1.8	97.5
	350	14.5	12.4	100	26.9	1.4	75.9
	400	11.4	7.5	100	18.9	1.0	54.2
	450	0	0	100	0	0	0
	500	0	0	100	0	0	0
0.6 wt.% Pt/AlSBA-15	200	6.5	0	11.6	6.5	0.3	9.2
	250	10.3	13.5	25.4	23.8	1.2	36.8
	300	22.4	31.7	100	54.1	2.8	85.9
	350	20.5	20.4	100	40.9	2.1	64.4
	400	16.5	9.5	100	26.0	1.3	39.9
	450	10.2	7.2	100	17.4	0.9	27.6
	500	0	0	100	0	0	0
1.0 wt.% Pt/ AlSBA-15	200	10.1	0	12.7	10.1	0.5	12.0
	250	21.6	27.1	100	48.7	2.6	62.8
	300	29.5	35.7	100	65.2	3.4	82.1
	350	23.5	16.6	100	40.1	2.1	50.7
	400	12.5	9.2	100	21.6	1.1	26.5
	450	8.5	5.4	100	13.9	0.7	16.9
	500	0	0	100	0	0	0
10 wt.% Pt/AlSBA-15	200	14.5	13.5	25.5	28.0	1.5	14.7
	250	33.3	42.3	100	75.6	4.0	39.4
	300	20.4	40.3	100	60.7	3.2	31.5
	350	17.1	34.4	100	51.5	2.7	26.5
	400	8.9	28.2	100	37.1	19	18.7
	450	6.8	24.5	100	31.3	1.6	15.7
	500	0	18.5	100	18.5	0.9	8.8

Pt/SiO$_2$–Al$_2$O$_3$ and Pt/SiO$_2$ were reported by Primet and co-workers [25].

In order to confirm the surface acidity of the sample catalysts, we used the temperature-programmed desorption of ammonia (NH$_3$-TPD). Figure 3 shows desorption patterns recorded for the Pt catalysts supported on SBA-15 and AlSBA-15. Desorption of ammonia of Pt/SBA-15 catalysts (figure 3(a)) proceeded in one stage with similar position and intensity in spite of different metal loading. The lower temperature peak around 120 °C can be attributed to physisorbed and weakly held ammonia molecules. Figure 3(b) presents the ammonia desorption patterns recorded for the Pt/AlSBA-15 catalysts. In general, the Pt/AlSBA-15 catalysts characterized by higher concentration of surface acid sites compared to the Pt/SBA-15 catalysts. All the Pt/AlSBA-15 samples with different metal contents consist of two low-temperature peaks with a maximum at 120 and 200 °C. The peak shown at 200 °C is related to mild acid sites, while the desorption peak of ammonia is too broad to be detected at a temperature higher than

300 °C corresponding to strong acid sites. These results clearly illustrate that the acidity of support material plays an important role in determining the activity of Pt/AlSBA-15.

In order to find the effect of different aluminum loading into SBA-15 on SCR activity of NO, we studied the catalytic performance on the 1 wt.% Pt/AlSBA-15 with Si/Al ratio of ∞, 40, 29, 10, and 5 as shown in table 4. It can be seen that the maximum NO conversion over 1 wt.% Pt/SBA-15 is increased from 250 to 300 °C after Al is incorporated to SBA-15. A maximum NO conversion of 65% can be obtained over 1 wt.% Pt/AlSBA-15 having a Si/Al ratio of 29. Pt/SBA-15 is more active than Pt/AlSBA-15 below 250 °C, because Pt-dispersion of Pt/SBA-15 is higher than Pt/AlSBA-15 and the presence of acid site in Pt/AlSBA-15 will make Pt particles to electron-deficient. However, the acid sites generated by Al incorporation produced mild and strong acid sites which will depress the complete oxidation of propene, resulting in the higher NO conversion at higher reaction temperatures. Pt/AlSBA-15 (5) containing the largest amount of acid site shows the maximum NO conversion temperature at 350 °C. However, maximum NO conversion decreased from 63.7 to 61.2% when Si/Al ratio decreased from 10 to 5 because of coke formation.

TPO spectra with coked catalysts are shown in figure 4. Prior to TPO, 1 wt.% Pt/AlSBA-15 with various Si/Al ratio was treated under the same condition with reaction test. The CO$_2$ evolved below 200 °C is negligible because the TCD detector of GC can not detect the low concentration of CO$_2$. All the samples were characterized by having two relatively high CO$_2$ peaks centered around 300 and 450 °C. The low-temperature peak can be assigned to carbonaceous materials present on Pt and the high-temperature peak was contributed by carbonaceous material or more possibly cokes present on AlSBA-15. The CO$_2$ peak of Pt/AlSBA-15 (Si/Al = 5) is larger than that of sample with Si/Al ratio of 10, 29, and 40, indicating a lot of carbonaceous materials were deposited on catalyst in order of surface acidity. Integration of CO$_2$ peak area accounted for 0.014 g 'C'/g catalyst for Si/Al = 5 and 0.011 g 'C'/g catalyst for Si/Al = 10. On the other hand, carbon formation of Si/Al = 29 and 40 was similar even they have different aluminum contents. About 0.007 g 'C'/g catalyst was formed over Pt/AlSBA-15 (Si/Al = 29 and 40). Better NO conversion was obtained by sample of Si/Al = 29, which suggests that a certain degree of acidity—but not to much—is favorable for NO conversion.

As water vapor and sulfur are present in lean-burn engine exhaust, we studied the water vapor and sulfur effect on the activity of 1 wt.% Pt/AlSBA-15 (29). After being on stream for 12 h at 300 °C, NO conversion remained at 65–67%. When 8 vol.% of water vapor was added to the reactant gas, the reduction of NO is found to be slightly suppressed. However, NO conversion
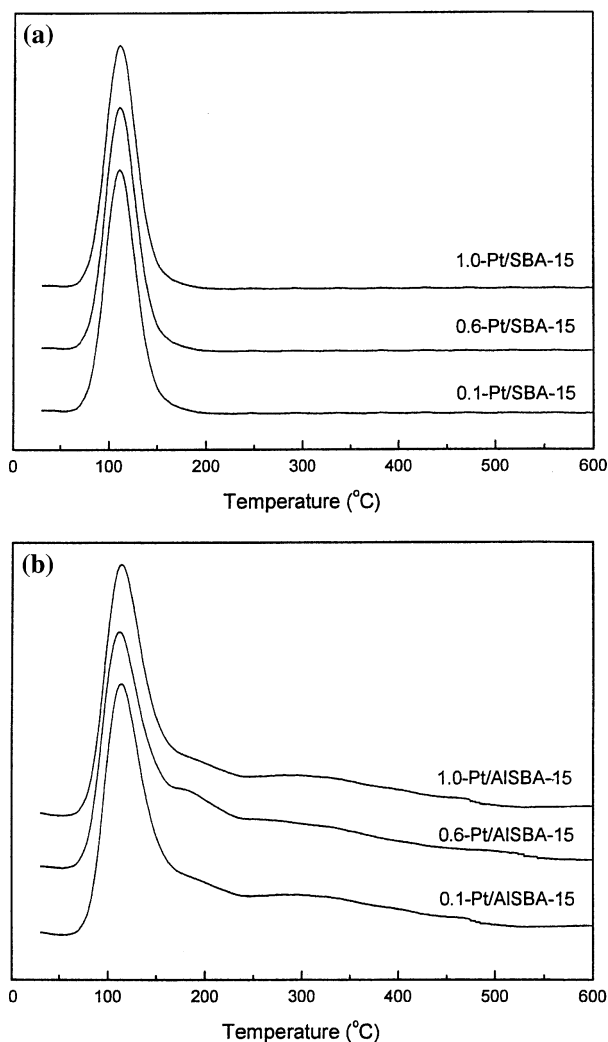

Figure 3. NH$_3$-TPD spectra of Pt/SBA-15 (a) and Pt/AlSBA-15 (b).

Table 4
NO conversion of 1 wt.% Pt supported on AlSBA-15 various Si/Al ratio

Catalysts	T (°C)						
	200	250	300	350	400	450	500
1 wt.% Pt/SBA-15 (∞)	11.2	59.3	41.5	29.8	15.7	7.5	0
1 wt.% Pt/AlSBA-15 (40)	9.5	34.5	60.7	33.5	18.1	12.5	0
1 wt.% Pt/AlSBA-15 (29)	10.1	48.7	65.2	40.1	21.6	13.9	0
1 wt.% Pt/AlSBA-15 (10)	10.1	12.7	63.7	34.1	18.1	10.1	0
1 wt.% Pt/AlSBA-15 (5)	11	12.3	31.5	61.2	15.6	7.8	0
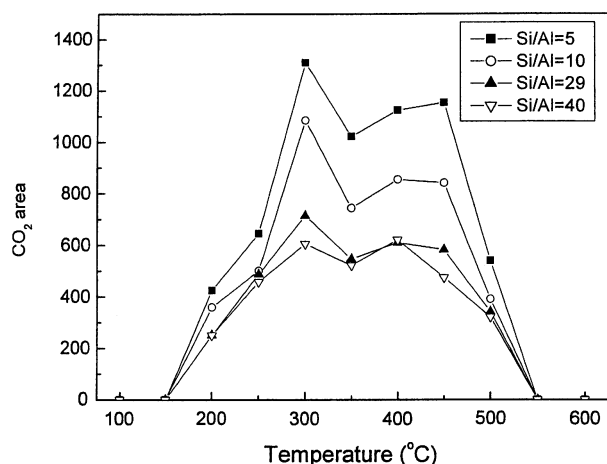
Figure 4. TPO spectra of coked 1 wt.% Pt/AlSBA-15 of various Si/Al ratio.

Figure 5. Effect of 8 vol.% of H_2O and 200 ppm SO_2 on NO conversion on 1.0 wt.% Pt/AlSBA-15 (29) at 300 °C. H_2O: (a) without H_2O, (b) 8 vol.% H_2O, (c) H_2O removed; SO_2: (a) 0 ppm, (b) 200 ppm, (c) 0 ppm; reaction conditions: NO = 2000 ppm, C_3H_6 = 2700 ppm, O_2 = 3 vol.%, GHSV = 35,000 h^{-1}.

restores to its initial value immediately after the water vapor has been removed from the feed stream. No irreversible deactivation is observed after operation in a stream containing 8 vol.% of water vapor for 30 h. The slight decrease of NO conversion over 1 wt.% Pt/AlSBA-15 (29) in the presence of this concentration of water vapor might be attributed to the suppression of the adsorption of reactants on the catalyst surface by water molecules. Figure 5 also shows the effect of SO_2 on catalytic performance of 1 wt.% Pt/AlSBA-15. When 200 ppm of SO_2 was added to the reactant gas after being on stream for 12 h, the catalytic performance remained unchanged in the following 18 h at 300 °C. These results indicate that both H_2O and SO_2 do not cause irreversible deactivation on Pt/AlSBA-15 (29) catalyst for SCR of NO in the presence of excess oxygen.

4. Conclusion

The Pt/AlSBA-15 catalysts prepared by a post-modification method with aluminum substitution into SBA-15 framework shows better catalytic activity than Pt/SBA-15 for the SCR of NO by C_3H_6 in the presence of excess oxygen. NH_3-TPD results show that the addition of aluminum give rise to form medium and strong acid site, particularly medium acid site appeared around
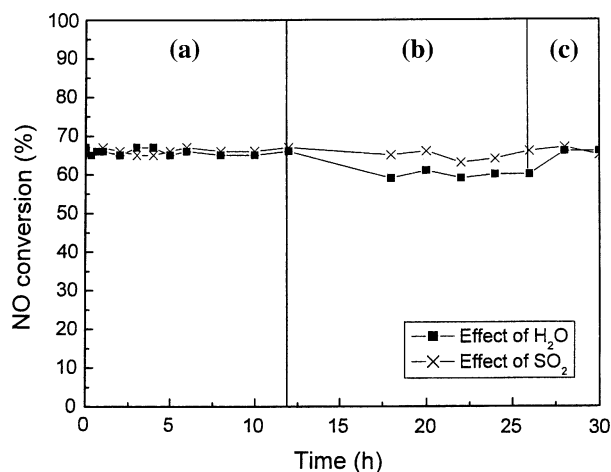
200 °C. For the Pt based catalysts used in our work, a certain degree of support acidity seems to be favorable for NO conversion. More specifically, the maximum NO conversion reaches 65% at a reaction temperature of 300 °C for Pt/AlSBA-15 (Pt = 1 wt.%) with Si/Al ratio of 29 among the Pt/AlSBA-15 with various Si/Al ratio of 5, 10, 29, and 40. The Pt/AlSBA-l5 (29) showed good stability. Addition of 8 vol.% of water vapor give rise to decrease the NO conversion slightly, however, it restored initial value after water vapor was removed from reactant stream. Meanwhile, 200 ppm of SO_2 added to reactant stream did not cause deactivation.

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