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Study on Pt/Al-MCM-41 for NO selective reduction by hydrogen

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ABSTRACT

Al species can be introduced to the framework of Si-MCM-41 support by isomorphous substitution or wet impregnation and Brønsted acid sites can be created in Al-MCM-41. Pt catalysts supported on Si-MCM-41 and Al-MCM-41 are studied for the selective catalytic reduction of NO by hydrogen in excess oxygen. The introduction of Al species to MCM-41 at suitable Si/Al ratio shows great promotion effect on H_2-SCR over Pt/MCM-41. Pt catalyst supported on Al-MCM-41 prepared by isomorphous substitution with the Si/Al ratio of 10 exhibits the best catalytic performance and *ca.* 80% NO_x conversion as well as *ca.* 85% N_2 selectivity can be achieved at 140 °C at high GHSV of 80,000 h^{-1}. The H_2-SCR reaction over Pt/Al-MCM-41, together with Pt/Si-MCM-41, are investigated by means of in situ DRIFT spectra and the existence of Brønsted acid sites in Al-MCM-41 is found to play a decisive role on the H_2-SCR reaction pathway.

1. Introduction

Catalytic elimination of nitrogen oxides (NO_x) is always a hot topic in the field of environmental catalysis. Selective catalytic reduction of NO_x by ammonia (NH_3-SCR) is a well-known process for NO_x emissions control and has been successfully applied in the post-treatment of NO_x from stationary sources [1]. Recently, hydrogen has been proved to be effective reductant for NO_x reduction even in excess oxygen over a series of catalysts [2–13]. It is desirable to replace ammonia with less processed and less expensive nontoxic hydrogen in the SCR process. Therefore, selective catalytic reduction of nitrogen oxides by hydrogen (H_2-SCR) is studied as possible alternative for NH_3-SCR.

In the research of possible H_2-SCR catalysts, noble metal catalysts, *e.g.* Pt catalysts [2–9] and Pd catalysts [10–12], have received considerable attention due to their high activities at low temperatures ($T < 200$ °C). For Pt catalysts, it is found that the activity and selectivity are strongly influenced by the properties of support materials. On the one side, acidic supports, *e.g.* zeolites with Brønsted acid sites, are helpful to stabilize active metallic Pt and to store ammonia as intermediate for SCR process [7]. As a result, high SCR activity and high N_2 selectivity are achieved on Pt catalysts on acidic supports. On the other side, basic supports with Lewis base sites favor the adsorption of NO_2^--type species as intermediates for SCR process and high SCR activity as well as high N_2 selectivity is also achieved on Pt catalysts on basic supports [8]. Obviously, different adsorption species are observed as intermediates during H_2-SCR reaction on different Pt catalysts, and correspondingly, different H_2-SCR pathways are involved in the reaction. Mesoporous MCM-41 has been successfully applied as high-surface-area supports for HC-SCR catalysts [14,15]. In our previous work, Pt catalysts supported on mesoporous Si-MCM-41 exhibits much better activity in H_2-SCR reaction than Pt/silicate and Pt/SiO_2 [16]. The mesoporous structure of Si-MCM-41 support is found to be essential to achieve the high activity. However, as we know, Si-MCM-41 is a neutral support without Brønsted acid sites or Lewis base sites. Therefore, the introduction of Brønsted acid sites or Lewis base sites to Si-MCM-41 may be possible means to enhance the SCR activity and N_2 selectivity of Pt/Si-MCM-41.

In the present study, Al species are introduced to Si-MCM-41 support by isomorphous substitution or wet impregnation and Al-MCM-41 with Brønsted acid sites are obtained. The catalytic performance of Pt/Al-MCM-41 is investigated for H_2-SCR reaction and the promotion effect of Al species is studied. Moreover, the H_2-SCR reaction pathways on Pt/Al-MCM-41 and Pt/Si-MCM-41 are investigated by means of in situ DRIFT spectra.

2. Experimental

2.1. Catalyst preparation

Mesoporous Si-MCM-41 sample was prepared by a hydrothermal route with cetyltrimethylammonium bromide (CTAB) as template and the details can be found in our previous work [16]. The

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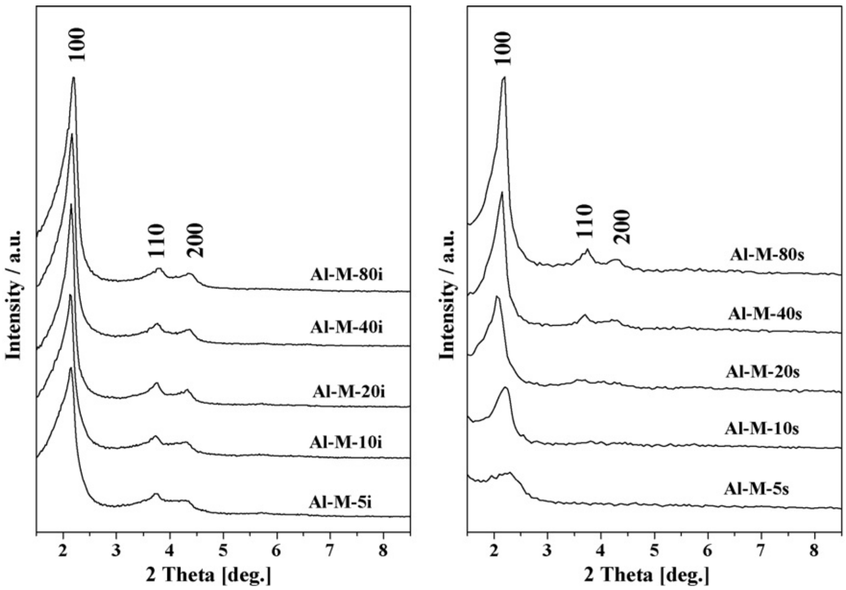

Fig. 1. XRD patterns of Al-MCM-41 prepared by different methods.

Al-MCM-41 samples were prepared by isomorphous substitution and wet impregnation. For isomorphous substitution, a hydrothermal route similar to the synthesis of Si-MCM-41 was employed. Briefly, CTAB, tetraethyl orthosilicate (TEOS) and $Al_2(SO_4)_3$ were slowly added into distilled water in turn under stirring and the pH value of obtained gel was adjusted to *ca.* 10.5. After further stir at 60 °C for 4 h, the gel was transported into an autoclave for crystallization at 110 °C for 12 h. The resulted solid was filtered, washed, dried at 80 °C and calcined at 500 °C. The product was defined as Al-M-*x*s, in which *x* indicating the Si/Al ratio. For wet impregnation, 1 g MCM-41 was impregnated with 5 mL $Al(NO_3)_3$ solution. The slurry was evaporated under stirring at 80 °C and then calcined in air at 500 °C. The final product was defined as Al-M-*x*i, in which *x* indicating the Si/Al ratio.

Pt catalysts on different supports with Pt loading of *ca.* 1% were prepared by incipient wetness impregnation method, using potassium chloroplatinate as platinum precursor. The catalysts were dried at 80 °C for 12 h and then calcined in air at 400 °C for 3 h.

2.2. Catalyst characterization

Powder X-ray diffraction (XRD) measurements were performed on the samples using a Rigaku D/max 2500 diffractometer, equipped with a graphite monochromator and using Cu Kα radiation.

The textural properties of samples were analyzed by low temperature N_2 adsorption/desorption using a Quantachrome NOVA-1200 gas absorption analyzer and the specific surface areas were calculated using the BET equation.

Transmission electron microscopy (TEM) images of samples were acquired on a Tecnai G^2 2010 S-TWIN transmission electron microscope at an accelerate voltage of 200 kV.

The dispersion of platinum in different catalysts was determined by H_2 pulse adsorption on a chemisorption analyzer (Chemisorb 2720, Micromeritics). In a typical experiment, *ca.* 100 mg sample in the quartz reactor was pre-oxidized in O_2/He at 400 °C for 1 h to ensure removal of hydrocarbons on the surface and then cooled to room temperature in flowing He. Subsequently, the sample was heated in 5%H_2/Ar to 350 °C, reduced in 5%H_2/Ar at 350 °C for 1 h and then treated in He at 350 °C until no hydrogen desorption can be observed. The possible hydrogen spillover is believed to be eliminated. After cooling down to room temperature in flowing He, pulses of H_2 were injected to the reactor every 1 min until no further changes in intensity of outlet H_2 (*ca.* 20 min). The dispersion of platinum was calculated assuming the equimolar adsorption of H on platinum.

The ^{27}Al MAS-NMR spectra were recorded on a Bruker DRX400 spectrometer (9.4 T). The samples were spun in the rotors. The r.f. pulse with the length of 0.3 μs was applied and the MAS frequency was fixed at 20 kHz. The spectrum was acquired after 2000 accumulations with the acquisition time of 0.1 s. A time interval of 1 s was selected between two successive accumulations to avoid saturation effects. Saturated $Al(NO_3)_3$ solution was used as the ^{27}Al chemical shift reference.

The acidity of Si-MCM-41 and Al-MCM-41 was determined by FT-IR spectra of pyridine adsorption. Prior to pyridine adsorption, the sample of *ca.* 0.1 g was out gassed at 400 °C for 2 h and cooled down to room temperature under vacuum. Pyridine was then introduced to the sample at saturated vapor pressure. Physisorbed pyridine was removed through heating the samples at 150 °C for 2 h and then the IR spectra were recorded. The bands in the IR spectra within the range of 1512–1567 cm^{-1} are corresponding to the Brønsted acid sites [17].

2.3. Catalytic testing

The selective catalytic reduction of NO by hydrogen at atmospheric pressure was carried out in a fixed-bed flow microreactor. The typical reactant gas composition was NO (1000 ppm), H_2 (5000 ppm), O_2 (6.7%), and the balance He. The total flow of the inlet gas was set at 200 mL min^{-1}. A sample weight of 100 mg was employed, corresponding to the gas hourly space velocity (GHSV) of 80,000 h^{-1}. Prior to testing, the samples were pretreated in O_2/He (6.7% O_2, 100 mL min^{-1}) for 1 h at 400 °C. The products were analyzed on-line using a gas chromatograph (HP 6820 series, for N_2 and N_2O analysis) and a chemiluminescence NO_x analyzer (Ecotech EC 9841, for NO and NO_2 analysis). The nitrogen balance is calculated for each step using the following equation, where NO_x is the sum of NO and NO_2: $[NO_x]_{inlet} = [NO_x]_{outlet} + 2[N_2O]_{outlet} + 2[N_2]_{outlet}$. This N-balance was found to be >95% for all experiments. The NO_x conversion is calculated as $XNO_x = ([NO_x]_{inlet} - [NO_x]_{outlet})/[NO_x]_{inlet}$. The N_2 selectivity is calculated as $S_{N_2} = 2 \times [N_2]/([NO_x]_{inlet} -$

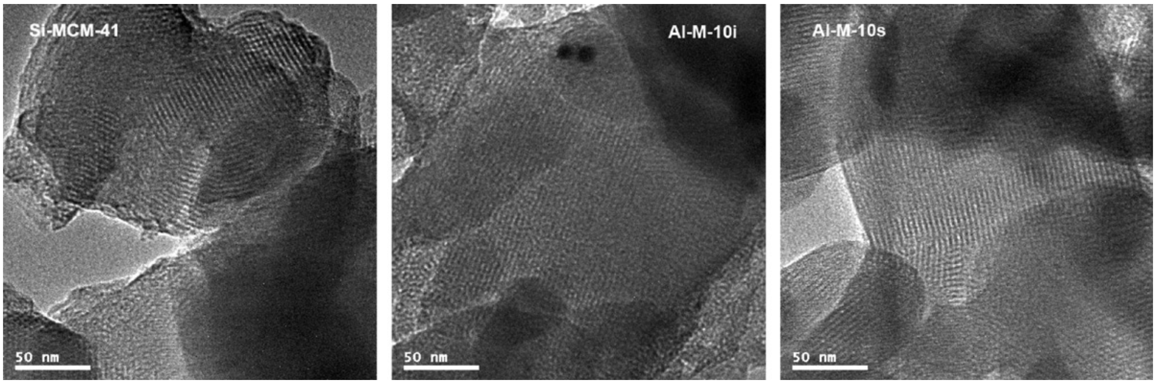

Fig. 2. TEM images of Si-MCM-41, Al-M-10i and Al-M-10s.

Table 1
Pore structural parameters and Brønsted acidity of Al-MCM-41 samples.

Sample	S_{BET} (m^2/g)	Pore diameter (nm)	Pore volume (cm^3/g)	Brønsted acid (mmol/g)
Si-MCM-41	1097	3.21	0.91	–
Al-M-80i	896	2.97	0.66	0.06
Al-M-40i	861	2.96	0.64	0.11
Al-M-20i	827	2.94	0.61	0.16
Al-M-10i	755	2.92	0.56	0.26
Al-M-5i	700	2.85	0.51	0.29
Al-M-80s	952	3.62	0.83	0.09
Al-M-40s	728	3.86	0.73	0.14
Al-M-20s	600	3.96	0.70	0.20
Al-M-10s	542	4.58	0.64	0.33
Al-M-5s	426	4.55	0.48	0.38

$[NO_x]_{outlet}$), and the N_2O selectivity is calculated as $S_{N_2O} = 2 \times [N_2O]/([NO_x]_{inlet} - [NO_x]_{outlet})$.

2.4. *In situ DRIFT study for H_2-SCR*

In situ DRIFT spectra were recorded on the Bruker Tensor 27 spectrometer, equipped with a liquid N_2 cooled high sensitivity MCT detector. The catalyst samples of *ca.* 25 mg were finely ground and placed in the chamber. Prior to each experiment, the samples were pretreated in O_2/He at 400 °C for 1 h, and cooled to the desired temperature for taking a reference spectrum. Then, the reaction gas mixture (1000 ppm NO, 5000 ppm H_2, 6.7% O_2, He balance, 25 mL/min) was fed and the steady-state spectra were recorded with a resolution of 4 cm^{-1} and an accumulation of 128 scans.

3. Results and discussion

3.1. *Characterization of Al-MCM-41*

Fig. 1 shows the XRD patterns of Al-MCM-41 samples prepared by different methods. For Al-MCM-41 prepared by wet impregnation, three diffraction peaks at $2\theta = 2.1°$, 3.8° and 4.4° are clearly observed on all samples, indexed as (1 0 0), (1 1 0) and (2 0 0) diffraction peak of characteristic MCM-41 structure respectively [18]. With increasing Al loadings, *i.e.* decreasing Si/Al ratios from 80 to 5, only slight decreases in peak intensities are observed, indicating that the hexagonally order structure are well preserved with the introduction of Al species by wet impregnation. For Al-MCM-41 prepared by isomorphous substitution, three diffraction peaks at $2\theta = 2.1°$, 3.8° and 4.4° are observed at low Al loadings, *i.e.* Si/Al = 80 and 40. With increasing Al loadings, the intensity of (1 0 0) diffraction peak decreases while the (1 1 0) and (2 0 0) diffraction peaks gradually disappear. Obviously, substitution of framework Si^{4+} by Al^{3+} in MCM-41 may result in the partially destruction of hexagonally order structure.

The textural properties of Al-MCM-41 samples are characterized and the results are summarized in Table 1. It is seen that the introduction of Al species to Si-MCM-41 results in general decreases in the specific surface areas. For Al-MCM-41 prepared by wet impregnation, both the pore diameters and pore volumes gradually decrease as Al loadings increase. For Al-MCM-41 prepared by isomorphous substitution, the pore diameters gradually increase as Al loadings increase (Si/Al ratios decrease from 80 to 10) while the pore volumes decrease probably due to the partially destruction of mesoporous structure as revealed in XRD patterns. The wormlike morphology consisting of mesopores are clearly observed on Si-MCM-41 and Al-MCM-41 at Si/Al = 10, as indicated by the TEM images in Fig. 2.

Introduction of Al species to MCM-41 by wet impregnation or isomorphous substitution results in the appearance of Brønsted acid sites and higher amount of Brønsted acid sites are observed with higher Al loadings, as indicated by the data in Table 1. It is also seen that more Brønsted acid sites are created in Al-MCM-41 sample prepared by isomorphous substi-

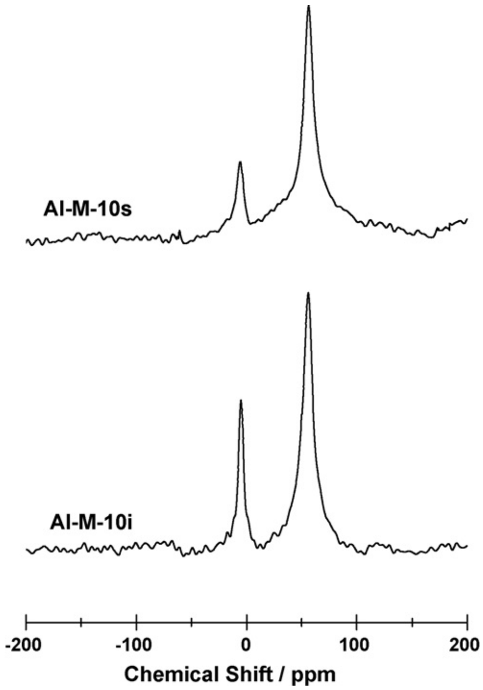

Fig. 3. ^{27}Al MAS-NMR spectra of Al-M-10i and Al-M-10s.

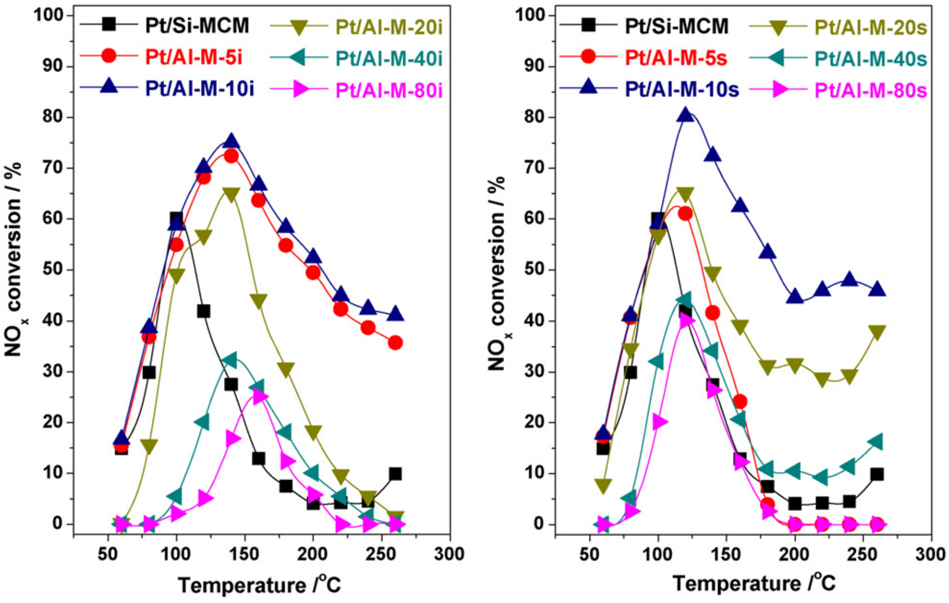

Fig. 4. Catalytic activities for H_2-SCR on Pt/Si-MCM-41 and Pt/Al-MCM-41. Reaction conditions: NO = 1000 ppm, H_2 = 5000 ppm, O_2 = 6.7%, He balance, GHSV = 80,000 h^{-1}.

tution than that prepared by wet impregnation with similar Al loading.

The ^{27}Al MAS-NMR spectra provide useful information regarding the coordination of aluminum in Al-MCM-41. As seen in Fig. 3, two types of aluminum are observed in Al-MCM-41 at Si/Al = 10: tetrahedrally coordinated Al in the framework at *ca.* 53 ppm and framework-connected octahedral Al species at *ca.* 0 ppm [19]. It is clearly indicated that not all the aluminum species are in the framework of MCM-41 and some aluminum may exist in the form of bulk Al_2O_3 connecting with the framework. As is known, Al species in the framework are the origin of Brønsted acid sites in Al-MCM-41. More framework Al species are observed in Al-MCM-41 sample prepared by isomorphous substitution than that prepared by wet impregnation at Si/Al = 10. And correspondingly, more Brønsted acid sites should be present in Al-M-10s than that in Al-M-10i, as proved by the data in Table 1.

Based on all above characterization results, it can be concluded that Al species can be introduced to the framework of MCM-41 by isomorphous substitution or wet impregnation and the mesoporous structure can be well preserved at certain Si/Al ratios. Brønsted acid sites are created due to the introduction of framework Al species and more Brønsted acid sites are observed on Al-MCM-41 prepared by isomorphous substitution than that prepared by wet impregnation with similar Al loading.

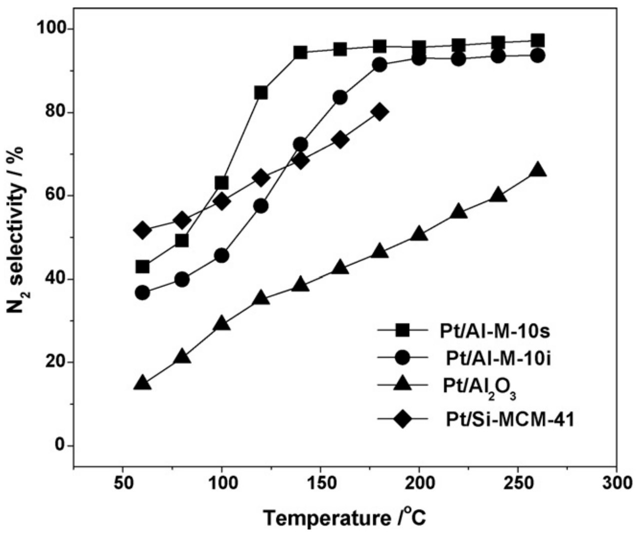

Fig. 5. N_2 selectivity during H_2-SCR on different Pt catalysts. Reaction conditions: NO = 1000 ppm, H_2 = 5000 ppm, O_2 = 6.7%, He balance, GHSV = 80,000 h^{-1}.

Table 2
Pt loadings and Pt dispersion in different Pt catalysts.

Catalyst	S_{BET} (m^2/g)	Pt loading (%)[a]	Pt dispersion (%)[b]
Pt/Si-MCM-41	1029	0.93	39.7
Pt/Al-M-80i	877	0.97	42.4
Pt/Al-M-40i	858	0.94	44.5
Pt/Al-M-20i	801	0.96	48.7
Pt/Al-M-10i	739	0.92	53.1
Pt/Al-M-5i	682	0.97	58.6
Pt/Al-M-80s	943	0.94	40.4
Pt/Al-M-40s	706	0.95	41.9
Pt/Al-M-20s	587	0.91	45.7
Pt/Al-M-10s	538	0.94	49.2
Pt/Al-M-5s	419	0.92	51.9

[a] Determined by ICP.
[b] Determined by H_2 chemisorption.

After the impregnation of Pt, the surface areas of mesoporous support materials decrease slightly (within 5%), as seen in Table 2. The exact Pt loadings in Pt catalysts are determined to be in the range of 0.91–0.97%, very much close to the theoretic value of 1%. Moreover, after the introduction of Al species to MCM-41 supports, the Pt dispersion increases to some extent. Typically, the Pt dispersion increases from 39.7% in Pt/Si-MCM-41 to 58.6% in Pt/Al-M-5i, while it increases to 51.9% in Pt/Al-M-5s.

3.2. *Catalytic performance of Pt/Al-MCM-41*

The catalytic activities for H_2-SCR over Pt/Si-MCM-41 and Pt/Al-MCM-41 are shown in Fig. 4. It is seen that the introduction of Al species to MCM-41 support shows obvious effects on NO_x conversion, which are dependent on the preparation methods of Al-MCM-41 and Si/Al ratios. Generally, promotion effect is observed at relative high Al loadings, *i.e.* Si/Al ratios <20. For Pt/Al-M-*x*i, the catalytic activity increases as Si/Al ratios decrease from 80 to 10 and the catalytic activity decreases slightly as Si/Al ratio further decreases to 5. The highest NO_x conversion of *ca.* 75% is obtained on Pt/Al-M-10i at 140 °C. For Pt/Al-M-*x*s, the catalytic activity also

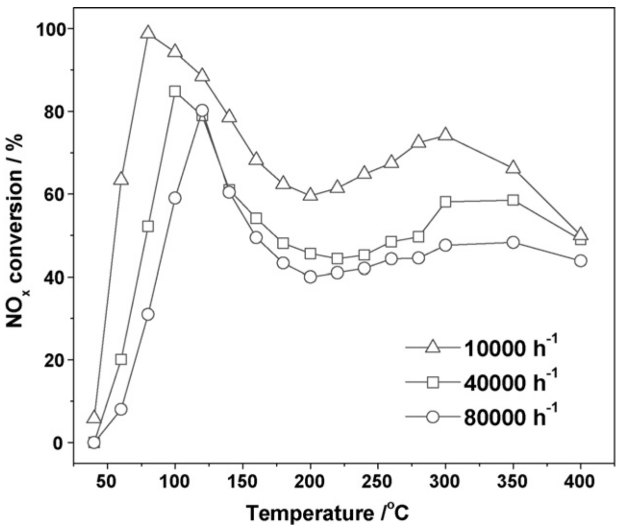

Fig. 6. Effect of GHSV on NO$_x$ conversion over Pt/Al-M-10s. Reaction conditions: NO = 1000 ppm, H_2 = 5000 ppm, O_2 = 6.7%, He balance.

increases as Si/Al ratios decrease from 80 to 10, while a dramatic decrease is observed as Si/Al ratio further decreases to 5. The highest NO$_x$ conversion of *ca.* 80% is obtained on Pt/Al-M-10s at 120 °C. The low activity of Pt/Al-M-5s is presumably ascribed to the serious destruction of mesoporous structure, as indicated by XRD and low temperature N_2 adsorption/desorption results.

The N_2 selectivity during H_2-SCR over several Pt catalysts is displayed in Fig. 5. The N_2 selectivity over Pt/Al_2O_3 is rather low at low temperatures, but it gradually increases with increasing temperatures. In contrast, N_2 selectivity of 50–80% can be obtained on Pt/Si-MCM-41 in the temperature range of 60–160 °C. The introduction of Al species to MCM-41 results in the enhancement of N_2 selectivity at high temperatures. For Pt/Al-MCM-41 at Si/Al = 10, N_2 selectivity of *ca.* 40% is observed at 60 °C and it gradually increases to >90% with increasing temperatures. It is also seen that Pt/Al-M-10s shows higher N_2 selectivity than Pt/Al-M-10i in the whole temperature range studied. As indicated by NMR results, more bulk Al_2O_3 are present in Pt/Al-M-10i, and hence more Pt species may locate on the Al_2O_3 to form Pt/Al_2O_3 instead of locating on the mesoporous structure of Al-MCM-41. Since the N_2 selectivity on Pt/Al_2O_3 is rather low, the N_2 selectivity on Pt/Al-M-10i is thus lower than that on Pt/Al-M-10s.

As indicated by the catalytic results, the introduction of Al species to Si-MCM-41 at certain Si/Al ratios shows significant promotion effect on H_2-SCR over Pt/MCM-41. The promotion effect is observed not only in the catalytic activity but also in the N_2 selectivity. Among all Pt/Al-MCM-41 catalysts studied, Pt/Al-M-10s shows the best catalytic performance and a maximal NO$_x$ conversion of *ca.* 80% can be obtained at 120 °C when N_2 selectivity is as high as *ca.* 85%. It has been reported that surface acidity produced by Al species can greatly improve the activity of Pt/MCM-41 for HC-SCR reaction [20]. In this study, similar results are obtained on Pt/MCM-41 for H_2-SCR reaction and the functions of acid sites will be discussed integrated with in situ DRIFT results in the following section.

The effects of GHSV on NO$_x$ conversion over the best catalyst Pt/Al-M-10s are shown in Fig. 6. It is seen that NO$_x$ conversion increases with decreasing GHSV from 80,000 to 10,000 h^{-1}. Meanwhile, the temperature when maximal NO$_x$ conversion is achieved shifts to lower temperatures. At low GHSV of 10,000 h^{-1}, an overall NO$_x$ conversion can be obtained at 80 °C.

3.3. *In situ DRIFT study on H_2-SCR*

In situ DRIFT spectra obtained under NO–H_2–O_2 reaction condition over Pt/Si-MCM-41 catalyst at various temperatures are shown in Fig. 7. Bands at 1840, 1755, 1625, 1585, 1420 and 1370 cm^{-1} are observed and their intensities change with reaction temperatures. The band at 1420 cm^{-1} is ascribed to adsorbed nitrites on support

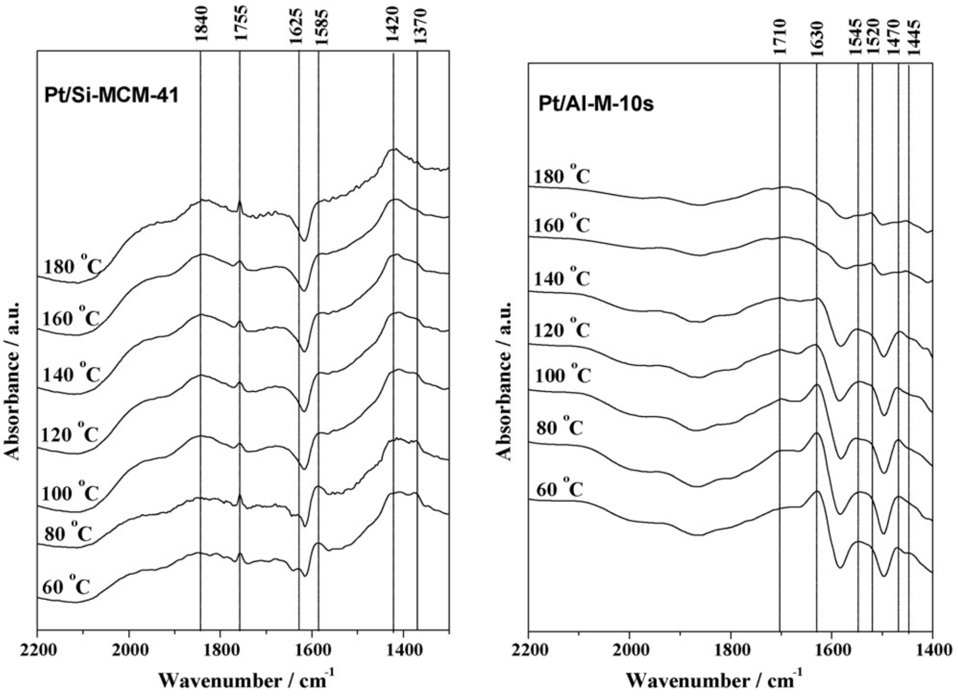

Fig. 7. In situ DRIFTS study of adsorbed species formed on Pt/Si-MCM-41 and Pt/Al-M-10s under NO–H_2–O_2 reaction condition. Reaction conditions: NO = 1000 ppm, H_2 = 5000 ppm, O_2 = 6.7%, He balance.

[21] and the band at 1370 cm^{-1} is ascribed to free nitrates [22]. The bands at 1625 and 1585 cm^{-1} are ascribed to bridging nitrates and chelating nitrates, respectively [23]. The band at 1755 cm^{-1} is ascribed to linear NO on defects of Pt sites [24] while the band at 1840 cm^{-1} is ascribed to mononitrosyl group on Pt^{n+} [25]. Based on above observations, the H_2-SCR pathway on Pt/Si-MCM-41 is proposed as following. In the initial step, gaseous NO adsorbs on the surface of Pt species as $NO(g) + {}^* \rightarrow -NO$ (*: active sites). Adsorbed NO may dissociate ($-NO + {}^* \rightarrow -N + -O$) and recombine to produce N_2 ($-N + -N \rightarrow N_2 + 2^*$) and by-product N_2O ($-N + -NO \rightarrow N_2O + 2^*$), as proposed by Burch et al. [26]. Meanwhile, the adsorbed NO species are oxidized to adsorbed nitrites or nitrates and then store on the support, as observed in the DRIFT spectra. These nitrites and nitrates may be reduced by hydrogen to produce N_2 or N_2O with different reactivity.

With the introduction of Al species to Si-MCM-41 by isomorphous substitution, *i.e.* the introduction of Brønsted acid sites, the adsorbed species on catalysts change greatly and bands at 1710, 1630, 1545, 1520, 1470 and 1445 cm^{-1} are observed on Pt/Al-M-10s. The band at 1710 cm^{-1} is ascribed to linear NO species on the terrace sites of Pt particles [24]. The band at 1630 cm^{-1} is ascribed to bridging nitrates while the bands at 1545 and 1520 cm^{-1} are ascribed to monodentate nitrates [25]. Remarkably, bands at 1470 and 1445 cm^{-1} corresponding to NH_4^+ on Brønsted acid sites [7,11] are observed. The NH_4^+ is key reaction intermediate for NH_3-SCR with high reactivity, as suggested by Richter et al. [27]. Based on the in situ DRIFT spectra and catalytic results, a different H_2-SCR reaction pathway is proposed for Pt/Al-MCM-41 with Brønsted acid sites. In the initial step, gaseous NO adsorbs as linear NO species on the terrace sites of Pt particles: $NO + {}^* \rightarrow -NO$. The adsorbed NO species easily dissociate to produced adsorbed N and adsorbed O ($-NO + {}^* \rightarrow -N + -O$), motivated by the low reaction barrier [28]. The dissociation of NO is also promoted by atomic hydrogen on Pt sites [29,30]. The adsorbed N may reacts with another adsorbed N to produce N_2 ($-N + -N \rightarrow N_2 + 2^*$) and reacts with adsorbed NO to produce N_2O ($-N + -NO \rightarrow N_2O + 2^*$). This may be possible pathway for the formation of N_2 and N_2O at low temperature, similar to that on Pt/Si-MCM-41. Alternatively, the adsorbed N may react with active hydrogen to produce $-NH_3$ on Pt sites ($-N + 3\ -H \rightarrow -NH_3 + 3^*$), which then migrate and store on the Brønsted acid sites as observed in the DRIFT spectra. The NH_4^+ species react with gaseous NO and O_2 to produce N_2 and H_2O with high reactivity and selectivity ($4-NH_4^+ + 4NO + O_2 \rightarrow 4N_2 + 6H_2O + -H^+$). This is another possible pathway for the formation of N_2. At relative lower temperatures, N_2 and N_2O are observed as H_2-SCR products and both of the above-mentioned two reaction pathways may be involved. At relative higher temperatures, N_2 is observed as the main H_2-SCR product and consequently the adsorbed NH_4^+ should be the main reaction intermediates.

4. Conclusion

By isomorphous substitution or wet impregnation, Al species can be introduced to MCM-41 at certain Si/Al ratio with the mesoporous structure well preserved. Some Al species exist in the form of framework Al species and others in the form of bulk Al_2O_3. Brønsted acid sites are created due to the introduction of framework Al species and more Brønsted acid sites are observed on Al-MCM-41 prepared by isomorphous substitution than that prepared by wet impregnation at similar Si/Al ratio. Al-MCM-41 with appropriate Brønsted acid sites appears to be better support for Pt than Si-MCM-41 for H_2-SCR reaction. A maximal NO_x conversion of *ca.* 80% with N_2 selectivity of *ca.* 85% can be obtained at 120 °C at high GHSV of 80,000 h^{-1} over Pt/Al-MCM-41 prepared by isomorphous substitution at Si/Al ratio of 10. The introduction of Brønsted acid sites changes the H_2-SCR reaction pathway and the NH_4^+ species on Brønsted acid sites are observed as key intermediates for N_2 production.

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