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Synthesis, Characterization, and Catalytic Performance of a ZSM-5/Y Composite

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A ZSM-5/Y composite zeolite was prepared from Y synthesis gel by using an in-situ two-step hydrothermal crystallization process. Optimized synthetic parameters made it practical to control the transformation of zeolite Y to P, or other undesired phases during ZSM-5/Y composite formation. It was found that the pH value range of the second phase (ZSM-5 in ZSM-5/Y composite) formation is much narrower than ordinary ZSM-5 synthesis, and a lower crystallization temperature was preferred in the second step. SEM showed that the morphology of the ZSM-5/Y composite was influenced by the different synthetic methods, such as seeds and templates. The NMR data indicated that the distribution of the Si(*n*Al) environments are different in ZSM-5/Y composite samples that made by different methods. The composite synthesized with seeds possessed a higher ratio of Si(0Al) to Si(1Al) than that of the composite synthesized with templates. Compared to a catalyst of a mechanical mixture of ZSM-5 and Y zeolite, the catalytic cracking performance of the ion-exchanged composite zeolite catalyst showed a better yield in gasoline formation when heavy oil was used as a feedstock in a MAT reactor, and the isomerization and aromatization abilities to the gasoline fraction were better than the physical mixture of the ZSM-5 and Y. This suggested that the composite structure of the zeolite may benefit the heavy oil transformation and related reaction steps during the course of the cracking process.

Zeolites Y and ZSM-5 are the major active components in fluid catalytic cracking (FCC) catalysts.[1] They are synthesized and purified individually by two different production lines, whereas they are mixed together when they are used in FCC catalysts.[2] In this way, hundreds of thousands of tons of Y and ZSM-5 zeolites are used every year worldwide. It is thus obviously advantageous to make them together when they are prepared. However, up to now, no study has been reported on the possibility of synthesizing these two zeolites in one autoclave. On the other hand, an excess silica source is required for the conventional method of zeolite Y synthesis. Although there are several methods for the recovery of this silica source,[3–6] it seems more attractive to develop a procedure to use this silica source in-situ.

Moreover, it is scientifically very interesting to synthesize composite molecular sieves,[7–18] and to study their catalytic synergism.[9–11,16,18] Research on composite molecular sieve materials has received much attention, and the synergism effect is always the goal in advanced catalysis. Y and ZSM-5 are two of the most important zeolites, how about their performance? Thus, from a scientific point of view, it is very interesting to make it possible and to see their synergism.

We reported preliminary results in a previous letter.[19] In this paper, we report on this one-pot methodology for preparing the Y and ZSM-5 composite, and its characterization as well as its catalytic performance for a heavy oil cracking reaction.

Experimental

Zeolites. The ZSM-5/Y composite zeolite was prepared in a Teflon-lined autoclave under static hydrothermal conditions by a two-step procedure. The first step was to synthesize NaY by a direct agent method. After the gel was autoclaved, and cooled to room temperature, templates, or seeds were added. Then the pH value was adjusted and it was crystallized to form the ZSM-5/Y composite material. The typical procedure was as follows. A direct agent, with a mole composition of $16Na_2O:Al_2O_3:15SiO_2:345H_2O$, was prepared by adding a solution of 0.37 g of sodium aluminate (C.P., SiO_2 54 wt %, Na_2O 41 wt %, Beijing Guohua New Chemicals Company), 7.54 g of deionized water and 1.63 g of sodium hydroxide (A. R., Beijing Chemicals Company) to 6.78 g of water glass (SiO_2 26 wt %, Na_2O 8.2 wt %, Beijing Hongxing Company), then aging at 40 °C for 4 h. The NaY gel was prepared as follows: 1.31 g of 50 wt % aluminium sulfate (A. R., Nanzhao Huaxin Company) solution; 4.03 g of the direct agent; 5.25 g of 30 wt % sodium aluminate solution and 3.77 g of 25 wt % sulfuric acid were in turn added to 17.30 g of water glass diluted with 13.46 g of deionized water with vigorous stirring for 60 min. The total composition of gel is $2.9Na_2O:Al_2O_3:8.4SiO_2:209H_2O$. After this, the resulting gel was introduced to an autoclave (100 mL), and kept at 100 °C for 24 h. Then, cooling to room temperature, 0.68 g of tetraethylammonium bromide (TEABr) and 1.06 g of tetrabutylammonium bromide (TBABr, A. R. Beijing Daxing) or 0.67 g of ethanol (A. R., Beijing Chemicals Company), or 0.22 g of ZSM-5 zeolite powder (Si/Al = 250) were added. After being stirred for 30 min, 25 wt % sulfuric acid or 30 wt % phosphoric acid (A. R., Beijing Chemicals Company) were added to change the pH value to 10.2, and stirred for 1 h. Finally, the gel was introduced to 100 mL of an autoclave and heated at 160 °C for 24 h. The crystalline product was collected by filtration, washed with deionized water, and dried at 120 °C for 2 h. The variable synthetic conditions were studied for preparing the ZSM-5/Y composite.

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Characterization. XRD patterns were collected with a Shimadzu Lab XRD-6000 diffractometer using Cu Kα radiation. XRD profiles were recorded in the range of 2θ values between 5 and 40°, with a continuous scan of 2°/min. The Si/Al of the framework of zeolite Y was calculated with the Breck–Flanigen equation.[20]

The weight percent of the zeolite ZSM-5 phase in the ZSM-5/Y composite was obtained by comparing the total areas of the diffraction peaks at the (101, 011) and (200, 020) crystal planes with the standard work curve (obtained by plotting the value of the total area of the diffraction peaks at the (101, 011) and (200, 020) crystal planes against the weight percent of ZSM-5 in Y and ZSM-5 zeolites mixtures), since there are no diffraction peaks in this range for the zeolite Y phase. A mechanical mixture sample was prepared by grinding ZSM-5 and Y zeolites according to this weight percent of ZSM-5 in the composite zeolite.

Elements in the mother liquor were analyzed with a Varian inductively coupled plasma-optical emission (ICP-OES) spectrometer, Vista AXCCD.

SEM pictures were taken with a LEO 435VP instrument equipped with an energy-dispersive spectroscopy (EDS) unit of type Oxford Link Isis. A gold film was sputtered onto samples supported on alumina disks prior to their observation. EDS was used to estimate the Si/Al ratio and sodium content of the composite zeolite.

N_2 adsorption–desorption isotherms were measured with a Micromeritics ASAP 2405N system. The zeolite thermal stability was evaluated following the differential thermal analysis (DTA) technique. Thermograms were obtained in a Universal V2.5H TA instrument under a flowing air atmosphere at a heating rate of 10 °C/min; typically, 200 mg samples were used.

^{29}Si MAS NMR spectroscopy was carried out with a Varian Unity INOVA300 spectrometer at 59.6 MHz, whereas ^{27}Al MAS NMR were carried out at 78.2 MHz. Samples were spun at 3 kHz for ^{27}Al and ^{29}Si MAS experiments. The chemical shifts were referenced to TMS for ^{29}Si and $[Al(H_2O)_6]^{3+}$ for ^{27}Al.

Catalyst and Tests. The composite zeolite and the corresponding mechanical mixture (NaY + ZSM-5) samples were ion-exchanged and post-treated by the same procedure of "two exchanges two calcinations" with $RECl_3$ and $(NH_4)_2SO_4$. The zeolites were then modified with phosphorus and magnesium by incipient wetness impregnation, followed by calcinations (RE_2O_3: 4.5 wt %; P: 1.5 wt %; Mg: 0.8 wt %). This modification made it suitable as an active component of a heavy oil catalytic cracking catalyst.

A typical procedure for model catalyst sample preparation was as follows: an oven-dried slurry of 35% (wt) zeolites, 50% (wt) kaolin, and 15% (wt) alumina hydrogel was calcined at 540 °C for 4 h. It was then mashed to 40–60 mesh particles and further steam-deactivated (800 °C/4 h/100% steam) before use.

A catalytic cracking experiment was performed in a standard micro-activity test (MAT) unit designed in accordance with ASTM D-3907. The reactions were carried out at 500 °C, 60 s time on stream. The ratio of the catalyst to oil was 3.75:1 (wt/wt), and the amount of catalyst was 3.75 g.

Daqing heavy oil was employed as the feedstock for model catalyst testing, this heavy oil was composed of 70% (wt) Daqing VGO (Vacuum Gas Oil, density (20 °C): 0.8748 g cm^{-3}; Percent distilled off at 500 °C: 38 wt %; Element analysis: C, 86.30; H, 13.70%) and 30% (wt) Daqing VR (Vacuum Residue), density (20 °C): 0.9310 g cm^{-3}; Percent distilled off at 500 °C: 7 wt %; Element analysis: C, 87.0; H, 12.67%).

A production distribution analysis, gasoline group analysis, and octane number analysis were carried out off-line using a gas chromatograph. The coke content in the spent catalyst was determined with an HV-4B analyzer.

Results and Discussion

In the present work, ZSM-5/Y composite zeolites were prepared in a Teflon-lined autoclave under static hydrothermal conditions by a two-step procedure. The first step was to synthesize NaY by a direct agent method. We then went to the second step. In the second step, the autoclave was cooled to room temperature, and then templates or seeds were added to the mother liquor with NaY particles, and the alkalinity was adjusted to the proper range to crystallize ZSM-5. It was then crystallized at a higher temperature, such as 160 °C. Alkalinity is one of the most important parameters in the synthesis of molecular sieves. A suitable pH value for the crystallization of ZSM-5 was in the range of 9 to 11; therefore, the proper pH value for synthesizing the ZSM-5/Y composite was investigated in this range. It showed that when templates were used, ZSM-5/Y composite zeolites were formed in a pH range of 10.0–10.5. However, there was no ZSM-5 phase detected at any pH value if no templates were used. Although at a low pH value (such as 9.42) ZSM-5 appeared, zeolite Y was completely transferred to zeolite P. The present work indicated that the suitable range of pH value for ZSM-5 formation in ZSM-5/Y composite synthetic system was narrower than that of pure ZSM-5 phase crystallization. The same results were observed when seeds (ZSM-5 powder) were used.

In an effort to synthesize ZSM-5/Y composite, it was found that one of the major tasks was to control the transformation of zeolite Y to zeolite P in the second step of crystallization. It is known that zeolite Y is a metastable zeolite, and we could transform it to zeolite P during the crystallization of Y by the directing agent method,[21] or to ZSM-4 by the addition of tetramethylammonium ions.[22] In the present work, we found that the transformation is very sensitive to the crystallization temperature. When it was under 180 °C, Y easily transformed to zeolite P, and finally formed a thermal-stable zeolite analcime (Fig. 1). However, when it was under 160 °C, the situation remarkably changed; zeolite Y was stable during the crystallization time range investigated in our experiments (Fig. 2). An even lower temperature, such as 140 °C, was investigated. There were no phases other than zeolite Y and ZSM-5 formed

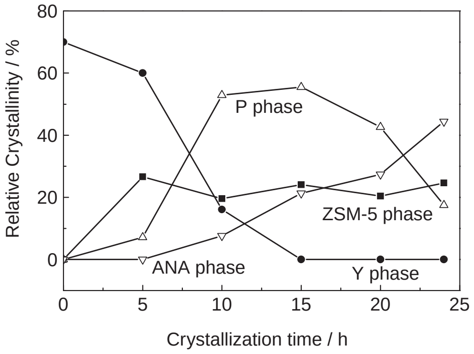

Fig. 1. Crystallization kinetics at 180 °C [n (templates)/n (SiO_2 of mother liquor) = 0.20, pH = 10.0 to 10.5].

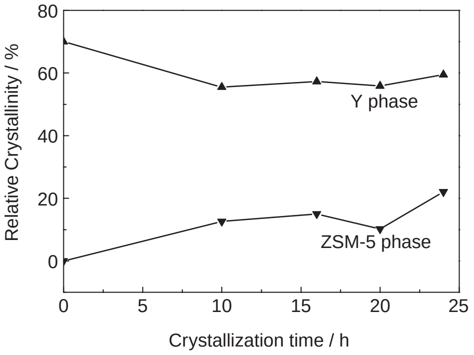

Fig. 2. Crystallization kinetics at 160 °C [n (templates)/n (SiO_2) = 0.20, pH = 10.0 to 10.5].

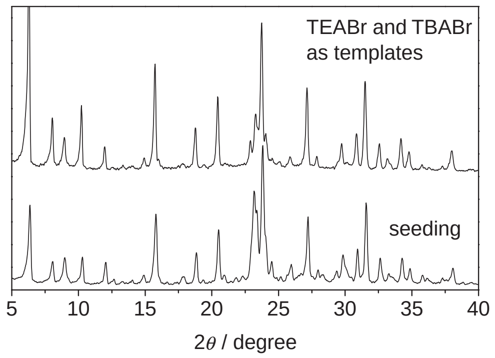

Fig. 3. Powder XRD patterns of ZSM-5/Y composite zeolites.

Table 1. Results of Si/Al Ratio and Mother Liquid Analysis

Sample	Total Si/Al[c)]	Y phase		Crystallinity of ZSM-5/%	Mother liquor[e)]		Usage percentage of silica source/mass %
		Crystallinity/%	Si/Al[d)]		SiO_2/g L^{-1}	Al_2O_3/g L^{-1}	
NaY	2.7	93	2.6	—	53.2	0.0668	55
S12[a)]	4.3	47	2.7	11	2.3	<0.0001	88
S13[b)]	4.1	58	2.7	15	7.2	<0.0001	86

a) Synthesized with seeded method. b) Synthesized with TEABr and TBABr. c) Analyzed by XRF. d) Calculated from XRD data. e) Analyzed by ICP.

during the crystallization process, but a prolonged crystallization time was needed. Thus, a lower temperature was preferred to synthesize ZSM-5/Y composites.

Experiments conducted under 160 °C indicated that there was no indication of a simultaneous crystallization of zeolite P and ZSM-5, i.e. an XRD analysis showed that no zeolite P phase was formed until completion of the crystallization of ZSM-5, which was in accordance with the results of Dwyer.[22] The crystallinity of zeolite Y phase was almost the same after the initial stage. When crystallization of ZSM-5 completed, zeolite Y began to transform to P, then to analcime. During the transformation, Y, P, analcime, and ZSM-5 could coexist. At last, only the most stable zeolite analcime and ZSM-5 remained. Mertens[23] reported that the use of an effective amount of crown ether could reduce the formation of unwanted zeolite P. In the present synthetic system, the zeolite Y phase avoided a transformation completely by controlling the crystallization temperature and time in the second step. Compared to the composite synthesized with seeds, the composite synthesized with quaternary ammonium salt templates had a higher relative crystallinity of the zeolite Y phase (Table 1). At the same time, using the templates method made it much easier to avoid a transformation to zeolite P.

To investigate the reliability and to obtain the usage percentage of silica, the experiments were conducted 80 times from 100 to 1000 mL scale. Table 1 gives the typical results of the Si and Al analyse. The usage percentage of silica in the mother liquid was increased by about 30% by the present method compared to that of ordinary zeolite Y synthesis. That means the silica was incorporate into the ZSM-5/Y structures by this in-situ treatment to NaY synthesis gel. This is obviously advantageous compared with the known silica recovery process of the NaY mother liquor, which was usually treated by adding some acidic chemical to precipitate the silica species.

Table 1 shows that the retaining crystallinity of the zeolite Y phase in a composite synthesized with templates was higher than that of the zeolite Y phase in a composite synthesized with seeds. It has been reported that organic cations, such as TPA^+ ions in silicalite-1 and ZSM-5, prevent the caustic from entering the zeolite channels, thereby limiting any hydroxide ion attack to the external surfaces of the crystals.[24–26] The difference in retaining the crystallinity of zeolite Y phase in a composite synthesized by a different method can also be explained with the same reason.

The powder XRD patterns of the ZSM-5/Y composite zeolites synthesized by templates and seeded methods are shown in Fig. 3. According to Fig. 3, all of the characteristic peaks of Y and ZSM-5 were observed, and no other peaks, such as P, analcime zeolite existed.

Concerning the morphology of the ZSM-5/Y composite, SEM revealed the formation of fairly complex aggregates of the MFI-type material and FAU. SEM images of composites synthesized with TEABr and TBABr are presented in Fig. 4. There is no typical morphology of ZSM-5 synthesized with the two templates used in the experiments. It was shown that the composite included two different shapes and sizes of particles: small irregular-shaped particles and big spheroid particles. For the ZSM-5/Y composite was synthesized with seeds, SEM images are shown in Fig. 5. There were also two different particles: laminar cuboids spotted by some parallel rods and small irregular particles.

To determine the structure of two different particles, EDS was used to determine the composition of two particles of the ZSM-5/Y composites. The results of EDS are given in Table 2. Firstly, the Si/Al of small particles of the composite was similar to that of pure zeolite Y. Also, the morphology of small particles was also similar to that of pure zeolite Y. It was thus concluded that the small particles in the composite were

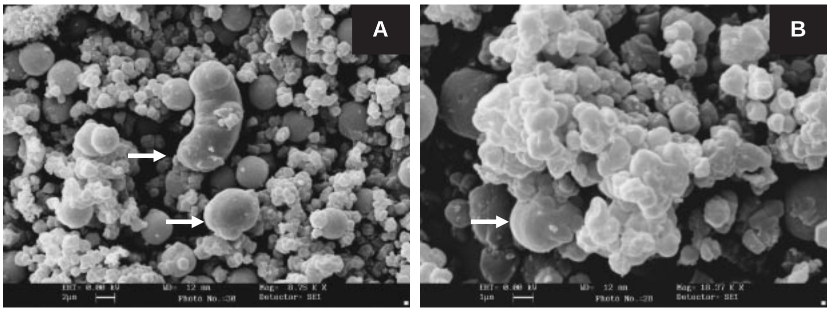

Fig. 4. SEM images of ZSM-5/Y composite (S14) synthesized with TEABr and TBABr: (A) ×8.75 K; (B) ×18.37 K at 160 °C for 24 h (*n* (templates)/*n* (SiO_2 in mother liquor) = 0.20; the gel was adjusted to pH = 10.2 with sulfuric acid; the relative crystallinity of Y and ZSM-5 phase are 57 and 15%, respectively; bulk Si/Al = 8.9, Si/Al (framework of Y) = 2.6).

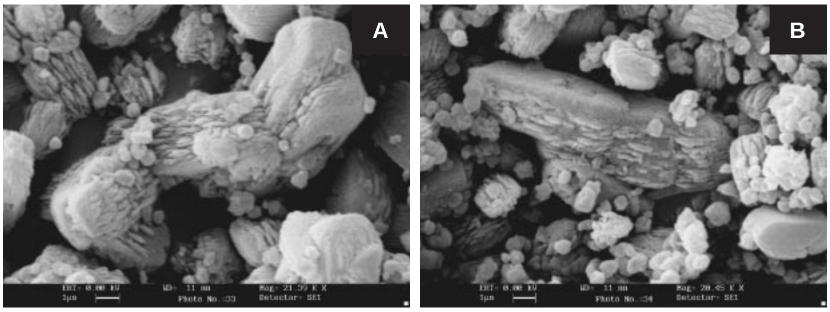

Fig. 5. SEM images of ZSM-5/Y composite (S15) synthesized with seeds: (A) ×21.39 K; (B) ×20.45 K (Si/Al (seeds) = 250 (adding amount of seeds/SiO_2 content in gel = 3 wt %; and the gel was adjusted to pH = 10.4 with phosphoric acid; crystallization at 160 °C for 24 h; the relative crystallinity of Y and ZSM-5 phase are 55 and 22%, respectively; bulk Si/Al = 8.8, Si/Al (framework of Y) = 2.6).

Table 2. Compositions of ZSM-5/Y Composites Analyzed by EDS

Sample	SiO_2 (compound %)	Al_2O_3 (compound %)	Na_2O (compound %)	Si/Al (mol)	Al/Na (mol)
Spheroid particle of S14	90.15	5.76	4.09	13.3	0.9
Small particle of S14	74.14	14.64	11.22	4.3	0.8
Cuboids of S15	93.76	3.37	2.87	23.6	0.7
Small particle of S15	77.41	13.37	9.22	4.9	0.9
NaY from the first synthetic step	70.36	18.43	11.22	3.2	1.0

zeolite Y particles.

How about the big particles? The Si/Al of big spheroid particles (Table 2) was in the range of the ZSM-5 zeolite. The big spheroid particles, indicated by the arrows in Fig. 4, were obviously not single particles, while they were agglomerates composed of several particles. From the synthetic process, it can be imagined that the zeolite Y particles agglomerated with the aid of the amorphous gel when the alkaline was adjusted with acid. The zeolite Y particles could be embedded in the big spheroid particles. Overgrowth of mesoporous MCM-41 on faujasite was stimulated by a surface cation exchange of CTA-cations and Na^+.[15] In our situation, the exchange of TEA^+ and TBA^+ with surface cations would cause the growth of ZSM-5 on the NaY particle surface. Another non-cationic template, ethanol, which was not a cationic template, was used as templates. A SEM image of ZSM-5/Y composite synthesized with ethanol is shown in Fig. 6, which displays a similar morphology to the composite synthesized with seeds. The distinction between quaternary ammonium salts and ethanol confirmed the above suggestion.

The adsorption isotherms of ZSM-5/Y composite zeolites were typical microporous isotherms of type I. The results of the N_2 physisorption of the ZSM-5/Y composite are listed in Table 3. The BET surface area was in accord with the XRD data. However, the external surface area of S14 was about half that of S15, which indicated that the agglomerates in S14 were more compact than that in S15.

The DTA profiles of S14, S15, and NaY are shown in Fig. 7. The thermal stability of the ZSM-5/Y composite was analyzed by DTA and compared with pure NaY (taken from the first step). The structural collapse temperatures of S14, S15, mechanical mixture (NaY + ZSM-5), NaY, and NaZSM-5 were 992, 926, 914, 946, and 866 °C, respectively. The DTA analysis indicated that the thermal stability of the ZSM-5/Y composite (S14) synthesized with TEABr and TBABr was higher than that synthesized with seeds (S15) and NaY. This result seems to contradict the facts that S14 have a lower Si/Al ratio compared with S15 (see Table 2 and Fig. 8), since it is well

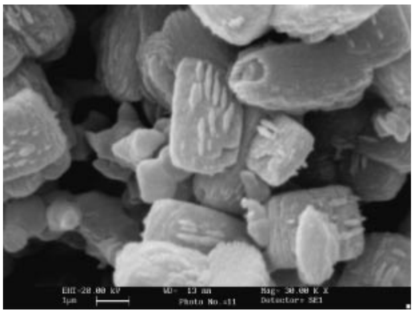

Fig. 6. SEM image of ZSM-5/Y composite synthesized with ethanol (at 160 °C for 24 h and the gel was adjusted to pH = 10.3 with sulfuric acid, *n* (ethanol)/*n* (SiO_2 in mother liquor) = 0.19).

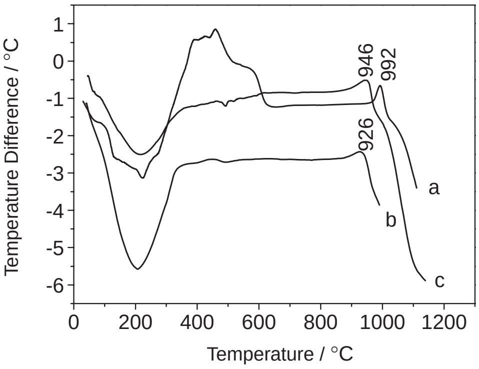

Fig. 7. DTA profiles of (a) S14, (b) S15, and (c) NaY taken from the first synthetic step.

Table 3. Surface Area, Micropore Volume, and Pore Volume of Calcined Composite Zeolites and Mechanical Mixture Samples

Sample	BET surface area/$m^2\,g^{-1}$	External surface area/$m^2\,g^{-1}$	Micropore volume/$cm^3\,g^{-1}$	Total pore volume/$cm^3\,g^{-1}$	Meso- and macro-pore volume/$cm^3\,g^{-1}$
S14	594	6.2	0.28	0.29	0.01
S15	539	12.9	0.25	0.29	0.04

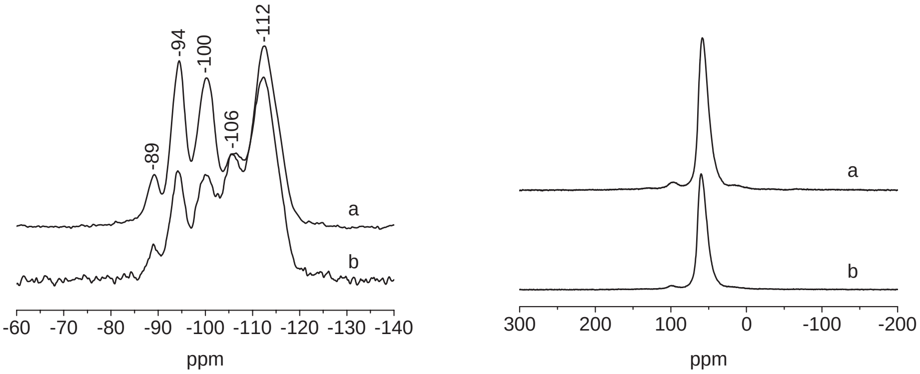

Fig. 8. ^{29}Si and ^{27}Al MAS NMR spectra of ZSM-5/Y composites: (a) S14; (b) S15.

known that the thermal stability of zeolite is dependent upon the framework Si/Al ratio. However, when we compared the collapse temperatures of NaZSM-5, NaY, and the mechanical mixture (NaY + ZSM-5), we found that NaY contributed to a higher apparent thermal stability of the mechanical mixture sample compared to NaZSM-5. This meant that we should also consider the contribution of NaY to the thermal stability in the composite zeolite samples. According to the data gives in Table 1 and our experience, the crystallinity of the Y phase in the composite synthesized with template method was always higher than that by the seeds method. This may indicate that S14 was composed of more effective NaY than S15, and this fact contributed to the higher thermal stability of S14 than S15.

The ^{29}Si MAS NMR spectra of the ZSM-5/Y composite synthesized with different methods are shown in Fig. 8. In the ^{29}Si spectrum of S14, four peaks with the maxima positioned at chemical shift values of δ -106.48, assigned to Si(0Al); at δ -100.15, assigned to Si(1Al); at δ -94.41, assigned to Si(2Al); at δ -89.15, assigned to Si(3Al) were attributed to zeolite Y,[27] and the peak at δ -112.32 was assigned to Si(0Al), to ZSM-5[28] of the composite. The NMR data indicated that the distribution of the Si(*n*Al) environments are different in the ZSM-5/Y composite samples that made by different methods. The composite synthesized with seeds (S15) possessed a higher ratio of Si(0Al) to Si(1Al) than that of the composite synthesized with templates. This suggested that when the composite was prepared with seeds, zeolite Y dissolved during the course of crystallization, but when the composite (S14) was prepared with TEABr and TBABr, the NaY did not undergo much change. From the ^{27}Al MAS NMR spectrum, only one peak, assigned to the framework aluminum, existed. This means that there was no detectable amorphous alumina species in the product.

The heavy oil MAT results of the ZSM-5/Y composite zeolite catalyst and the catalyst of a mechanical mixture of ZSM-5 and Y zeolite are listed in Table 4. This indicates that compared to a catalyst of a mechanical mixture of ZSM-5 and Y zeolite, the composite zeolite (seeds method) catalyst afforded a 5.20 wt % increase in the gasoline yield, a 1.86 wt % decrease in the light cycle oil (LCO), similar to the yield in LPG, and a 1.09 wt % decrease in coke, i.e., the yield of light liquid (LPG + gasoline + LCO) increased by 2.94 wt %. As for the

Table 4. Products Distribution of ZSM-5/Y Composite Zeolite and Mechanical Mixture in the Catalytic Cracking of Daqing Heavy Oil in a MAT Reactor

Products Distribution /wt%	ZSM-5/Y Composite Zeolite[a)] Catalyst	ZSM-5+Y Mechanical Mixture Catalyst
Dry gas	5.43	8.18
LPG	16.20	16.60
Gasoline	57.97	52.77
LCO	12.82	14.68
Heavy oil	4.50	3.60
Coke	3.08	4.17
LPG + Gasoline + LCO	86.99	84.05

a) The composite zeolite was synthesized with seeds method.

Table 5. PIONA (wt%) and Octane Number Analysis of Gasoline Fraction

Catalyst	ZSM-5/Y Composite Zeolite[a)] Catalyst	ZSM-5+Y Mechanical Mixture Catalyst
Total normal paraffins	5.59	6.20
Total isoparaffins	18.38	17.05
Total olefin	50.24	52.72
Total naphthenes	9.25	8.71
Total aromatics	16.69	15.31
MON	81.00	80.55
RON	89.68	90.02

a) The composite zeolite was synthesized with seeds method.

case of a catalyst made with a templates composite zeolite, it showed similar results: the yield of light liquid increased by 1.99 wt%.

The gasoline fraction PIONA analysis given in Table 5 indicates that the composite zeolite catalyst showed a better isomerization effect, while the olefin content decreased by 2.48 wt%; meanwhile, the aromatics increased by 1.38 wt%. It is worth noting that when the olefin content in gasoline decreased, no loss of the Research Octane Number (RON) and Motor Octane Number (MON) occurred.

The above results indicate that the composite zeolite showed an advantage in the heavy oil catalytic cracking reaction; the agglomerate or layered structure of the composite zeolite may benefit heavy oil transformation during the course of the cracking reaction. This suggests that the ZSM-5 and Y zeolite in the composite exhibited a synergy effect.

Conclusion

A novel one-pot two-step in-situ crystallization procedure for preparing ZSM-5/Y composite materials from NaY synthesis gel has been successfully developed. Synthesis parameters were investigated, indicating that the optimized conditions for the step of zeolitization are: pH 10 to 10.5 and reaction temperature of 160 °C. The transformation of zeolite Y to zeolite P and other unwanted material can be totally avoided; the ratio of ZSM-5 to Y can be controlled. The results of SEM and EDS analyses indicated that the irregular spheroid particles are one of the characteristic morphology of a composite prepared by the templates method. Laminar cuboids spotted by some parallel rods comprise one of the characteristic morphology of composites prepared by the seeded method. The NMR data indicated that the distribution of Si(*n*Al) environments are different in the ZSM-5/Y composite samples that made by different methods. The composite synthesized with seeds possessed a higher ratio of Si(0Al) to Si(1Al) than that of the composite synthesized with templates. Compared to a catalyst of a mechanical mixture of ZSM-5 and Y zeolite, the catalytic cracking performance of the ion-exchanged composite zeolite catalyst showed a better yield in gasoline formation. Also, the isomerization and aromatization ability to the gasoline fraction were better than that of the physical mixture. This suggested that the ZSM-5 and Y zeolite in the composite exhibited a synergy effect.

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