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Preparation of uniform and nano-sized NaA zeolite using silatrane and alumatrane precursors

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Nano-sized Na A zeolite was successfully synthesized via the sol–gel process and microwave techniques. The synthesis parameters, such as hydroxide ion concentration, seed amount, as well as heating time and temperature, were studied to obtain the most uniform and very small sized NaA zeolite using the composition of SiO_2:Al_2O_3 : $x$$Na_2O$:410$H_2O$; $3 \leq x \leq 6$. It was found that hydroxide ion concentration affects the crystal size and heating time, whereas a higher amount of seed provides smaller sized NaA zeolite. The zeolite product can be synthesized using a higher temperature for a shorter time or lower temperature for a longer time. The best conditions for synthesizing the smallest size, 0.1–0.2 µm, and the most homogeneous NaA zeolite is to use the composition of SiO_2:Al_2O_3:3Na_2O:410H_2O and 3 wt% crystal seed at 80 °C microwave heating for 6 h. The synthesized NaA zeolite was characterized using XRD and SEM. Copyright © 2006 John Wiley & Sons, Ltd.

KEYWORDS: NaA zeolite; sol-gel process; silatrane and alumatrane; nano-sized

INTRODUCTION

Zeolite A, an aluminosilicate having a small pore size of 4 Å and containing aluminum in the framework, results in requiremed balancing cations. The common form generally used as a membrane is the sodium form, NaA. NaA zeolite has also been employed in gas separation membranes to enhance the selectivity of the support.[1] In addition, NaA zeolites are used in many applications such as household products, aquaculture, agriculture, water treatment, etc., due to their adsorption, ion exchange and size selectivity properties.[2] Commercial NaA zeolite is synthesized by both conventional and microwave heating techniques. The microwave technique is used in NaA zeolite synthesis due to its homogeneous heating, resulting in tiny hot spots throughout the synthesis mixture, which reduce synthesis time.[3]

The hydrothermal synthesis of aluminosilicate zeolite corresponds to the conversion of a mixture of silicon and aluminum compounds, alkali metal cation, organic molecules and water via an alkaline supersaturated solution into microporous crystalline aluminosilicate.[4] The variables, such as the composition of reaction mixture, temperature and time, have a major influence on zeolite structure crystallized. When the reactants are mixed, these solid phases are dissolved, mediated by the mineralizing agent or alkali. The solution is filled with silicate and aluminate monomer and oligomer which are organized into a more extended structure and finally into the crystalline phase. The crystallization process is thermally activated and allows a high yield of crystal to be achieved in an acceptable period of time. The crystallization period of zeolite can be subdivided into two steps: nucleation of the new population of zeolite crystals and growth of the existing population of crystals.

Seeding is a technique in which the supersaturated system is inoculated with small particles of material to be crystallized. Using this operation, the nucleation stage is by-passed and the induction period is eliminated.[4] Seed crystals are added to a synthesis mixture which may act as pure seeds in that mass is deposited upon them and they grow or give rise to secondary nuclei and hence a new crop of crystals. In the absence of seed crystals, the reaction gives either very low yield or products heavily contaminated with impurity phases.[5]

Recently, Wongkasemjit *et al.*[6–8] have developed a method to produce uniform zeolites using synthesis silatrane and

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alumatrane as precursors via the sol–gel process and microwave techniques. To produce a strong and efficient film for zeolite membrane application, the characteristic of being uniform is crucial. Thus, this work is focused on the production of NaA zeolite from those atrane precursors cooperating with the seeding technique to reduce crystal size. We also investigate the effect of hydroxide concentration, microwave temperature and time as well as seed wt% on small size and uniform NaA zeolite synthesis.

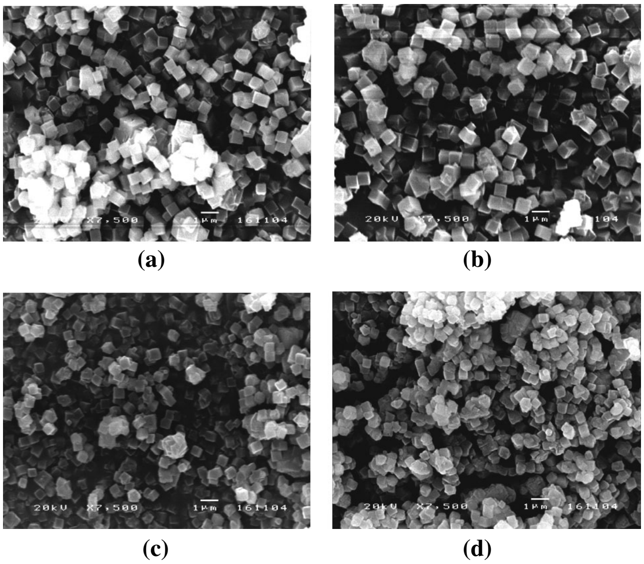

Figure 1. Effect of hydroxide concentration on zeolite formation using $SiO_2 : Al_2O_3 : xNa_2O{:}410H_2O$, 1 wt% seed, 110 °C microwave heating for 2 h 20 min with hydroxide concentration, x, of (a) 3, (b) 4, (c) 5 and (d) 6.

EXPERIMENTAL

Materials

Fumed silicon dioxide (SiO_2), aluminum hydroxide hydrate [$Al(OH)_3xH_2O$], triisopropanolamine [TIS, $N(CHCH_3CH_2OH)_3$] and triethanolamine [TEA, $N(CH_2CH_2OH)_3$] purchased from Sigma Aldrich Chemical and Carlo Erba, respectively, were used as reactants. Ethylene glycol (EG, $HOCH_2CH_2OH$) purchased from J.T. Baker Inc. was used as the reaction solvent. Sodium hydroxide (NaOH) from EKA Chemical was used as the base catalyst. Nitrogen gas with 99.99% purity was purchased from Thai Industrial Gases public company (TIG); and acetonitrile (CH_3CN) from Lab-Scan Co. Ltd. All chemicals were used as received.

Instruments

Fourier transform infrared (FTIR) spectra were obtained on a Nicolet spectrometer with 16 scans, a resolution of 4 cm^{-1} and a frequency range of 4000–400 cm^{-1}. The powder samples were pressed as pellets by mixing with pure and dry potassium bromide (KBr). Thermogravimetric Analysis (TGA) on a Perkin Elmer, TG-DTA, using a platinum pan with a sample weight of 10 μg, was carried out at a temperature range from room temperature to 750 °C with a heating rate of 10 °C/min under nitrogen atmosphere with a nitrogen flow rate of 20 ml/min. Scanning Electron Micrographs (SEM) were obtained on a Joel 5200-2AE (MP15152001) using a voltage of 15–20 kV at 1500–7500 magnification. Wide-angle

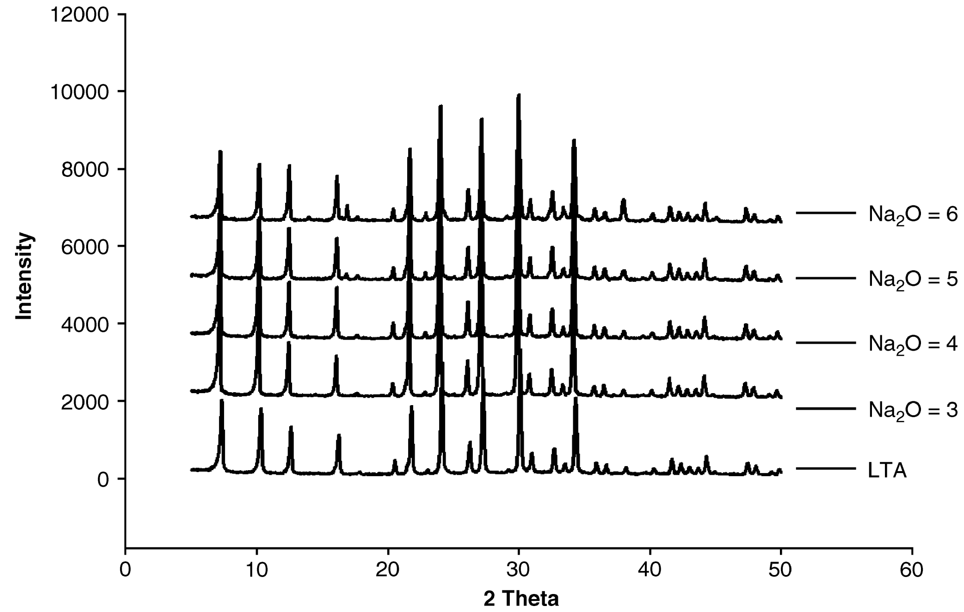

Figure 2. XRD spectra of synthesized NaA zeolite using $SiO_2 : Al_2O_3 : xNa_2O{:}410H_2O$, 110 °C microwave heating for 2 h 20 min with hydroxide concentration, x, of (a) 3, (b) 4, (c) 5 and (d) 6.

X-ray diffractograms (WXRD) on the D/MAX 2000 series of Rigaku X-ray diffractometer system were analyzed to identify the zeolite NaA pattern using CuK_α as a source with 30 mA, 40 kV, a range of 5–50°/min and 0.02° scan step. Microwave thermal treatment was performed on the MSP 1000, CEM Corporation Spec 1000 W and 2450 MHz, using a teflon vessel, organic digestion mode and time to temperature program. Surface areas of the samples were determined by physisorption of nitrogen at liquid temperature (77 K) using an Autosorb I instrument.

Methodology

The synthesis mixture for NaA zeolite was prepared according to the procedure reported previously[6] with the molar ratio of $SiO_2 : Al_2O_3 : x Na_2O : 410 H_2O$, $3 \le x \le 6$. Silatrane and alumatrane synthesized via the OOPS (one pot oxide synthesis) process and characterized by the method of Wongkasemjit's group[9,10] were used to prepare NaA incorporation with the seeding technique. NaA from the same process was also used as the crystal seed. The medium was aged for half a day. The effects of parameters (namely, amount of NaOH, microwave heating time and temperature, as well as number of seed crystals) on the small size and uniformity of NaA zeolite were studied. The synthesized products were further characterized for their structure and morphology using XRD and SEM, and chemical analysis using SEM-EDX.

RESULTS AND DISCUSSION

Precursor synthesis

Silatrane [tris (silatranyloxyethyl)amine or Si-TEA] and alumatrane [tris-(alumatranyloxy-1-propyl)amine or ALTIS] were characterized using FTIR and TGA. FTIR confirmed the characteristic peaks of silatrane and alumatrane precursors. Silatrane: 3000–3700 cm^{-1} (w,νO–H), 2860–2986 cm^{-1} (s,νC–H), 1244–1275 cm^{-1} (m,νC–N), 1170–1117 cm^{-1} (bs,νSi–O), 1093 cm^{-1} (s,νSi–O–C), 1073 cm^{-1} (s,νC–O), 1049 cm^{-1} (s,νSi–O), 1021 cm^{-1} (s,νC–O), 915–940 cm^{-1} (m,δSi–O–C), 785 and 729 cm^{-1} (s,δSi–O–C) and 379 cm^{-1} (w,Si ← N). Alumatrane: 3404 cm^{-1} (b,νO–H), 2937–2881 cm^{-1} (s,νC–H), 2750, 2670, 3700 cm^{-1} (w, NR^3 salt Si ← N) 1356 (w,νC–N), 1259 (m,νC–O) 1040 cm^{-1} (m,νSi–O), 885 cm^{-1} (s,νAl–O–C) and 659–400 cm^{-1} (s,δ Al–O). Thermal properties and stability were analyzed by thermogravimetric analysis. The TGA results of alumatrane give two mass loss transitions at 141 and 383 °C corresponding to the decomposition of the EG solvent trapped in their structure and organic ligand, respectively. The ceramic yield, 31.6, corresponds to the structure of $Al(OCHCH_3CH_2)_3N$, which is larger than the theoretical (23.72%), presumably due to the incomplete combustion. Silatrane exhibits only one weight loss transition at around 400 °C corresponding to the decomposition of organic ligand. The char yield is around 18.5%, which is close to the theoretical yield of $Si(OCH_2CH_2)_3N_2)H_2$ (18.4%).

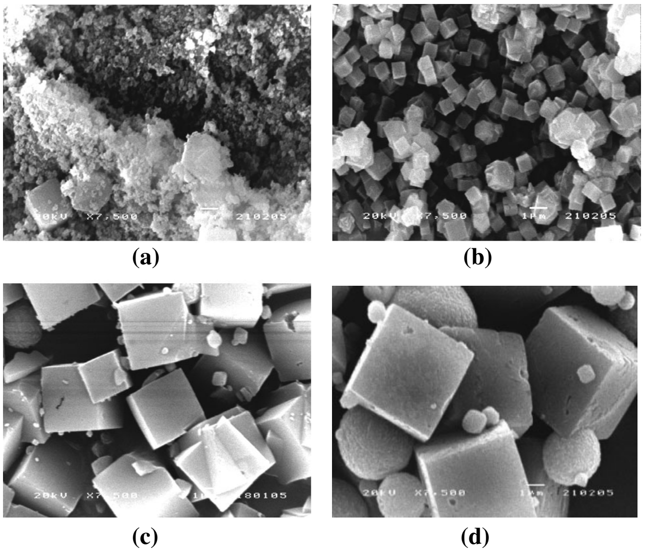
(a) (b) (c) (d)

Figure 3. Effect of temperature on zeolite formation without seeding using $SiO_2 : Al_2O_3 : x Na_2O : 410 H_2O$ at microwave heating temperature of (a) 90°, (b) 110°, (c) 130° and (d) 150 °C for 2 h 20 min.

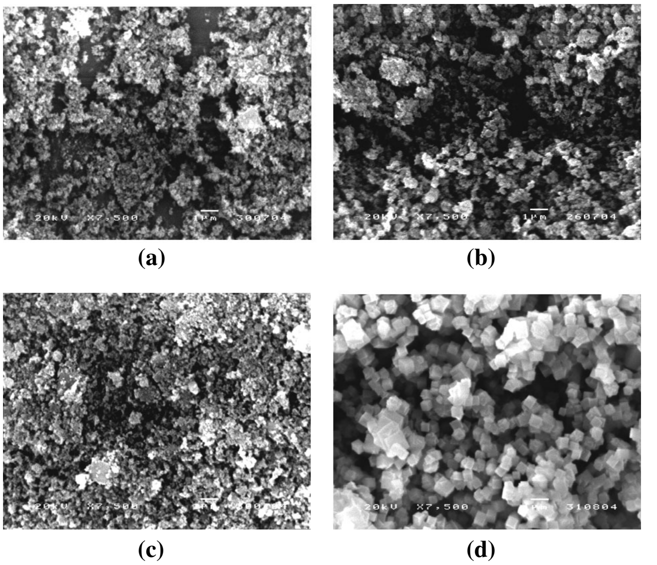
(a) (b) (c) (d)

Figure 4. SEM micrographs of NaA zeolite synthesized using $SiO_2 : Al_2O_3 : x Na_2O : 410 H_2O$ with 3wt% crystal seed at microwave heating temperatures of (a) 80°, (b) 90°, (c) 100° and (d) 110 °C for 2 h 20 min.

Effect of hydroxide concentration

From previous study,[6] increasing NaOH concentration also increased the hydroxide concentration, which in turn enhanced the dissolution of the amorphous gel and nucleation

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rate, leading to the growth of more crystals. In this study, the effect of hydroxide concentration was investigated parallel to the seeding effect by fixing the microwave heating temperature at 110 °C for 2 h 20 min and starting material composition at $SiO_2 : Al_2O_3 : xNa_2O{:}410H_2O$, $3 \leq x \leq 6$. Figure 1 shows that 1 wt% seeding NaA zeolite crystals were observed when $x = 3$. The crystal sizes slightly decreased with increasing OH^-:Si ratio. At a hydroxide concentration equal to 3, the average size was 0.7–0.8 μm. When hydroxide concentration is increased to 4, the particle size distribution increased to 0.9–1 μm. However with hydroxide concentrations changed to 5 and 6, the crystal size was as small as 0.4–0.6 μm. The phase of NaA crystals was confirmed by XRD in Fig. 2 compared with the commercial one. XRD data confirmed the reference pattern matching the one when using hydroxide concentrations equal to 3 and 4. We know that the hydroxide concentration is decreased with the crystallization time reduction. When hydroxide concentration is increased to 5 and 6 at the microwave heating time and temperature of 2 h 20 min and 110 °C, respectively, the over-run zeolite product occurs. The XRD pattern (Fig. 2) also shows some impurity at $2\theta = 18$ of NaP zeolite.[11] Referring to the previous study,[6] the same conditions and without the seeds, NaA zeolite crystals were synthesized by microwave heating at 110 °C for 2 h 40 min. By the seeding operation, the nucleation stage is shorter due to the formation of new nuclei on the seed crystal surface and the subsequent growth of the new crystallite of the same phase.[12] Thus, crystallization was clearly enhanced because NaA zeolite crystals were observed in a shorter time.

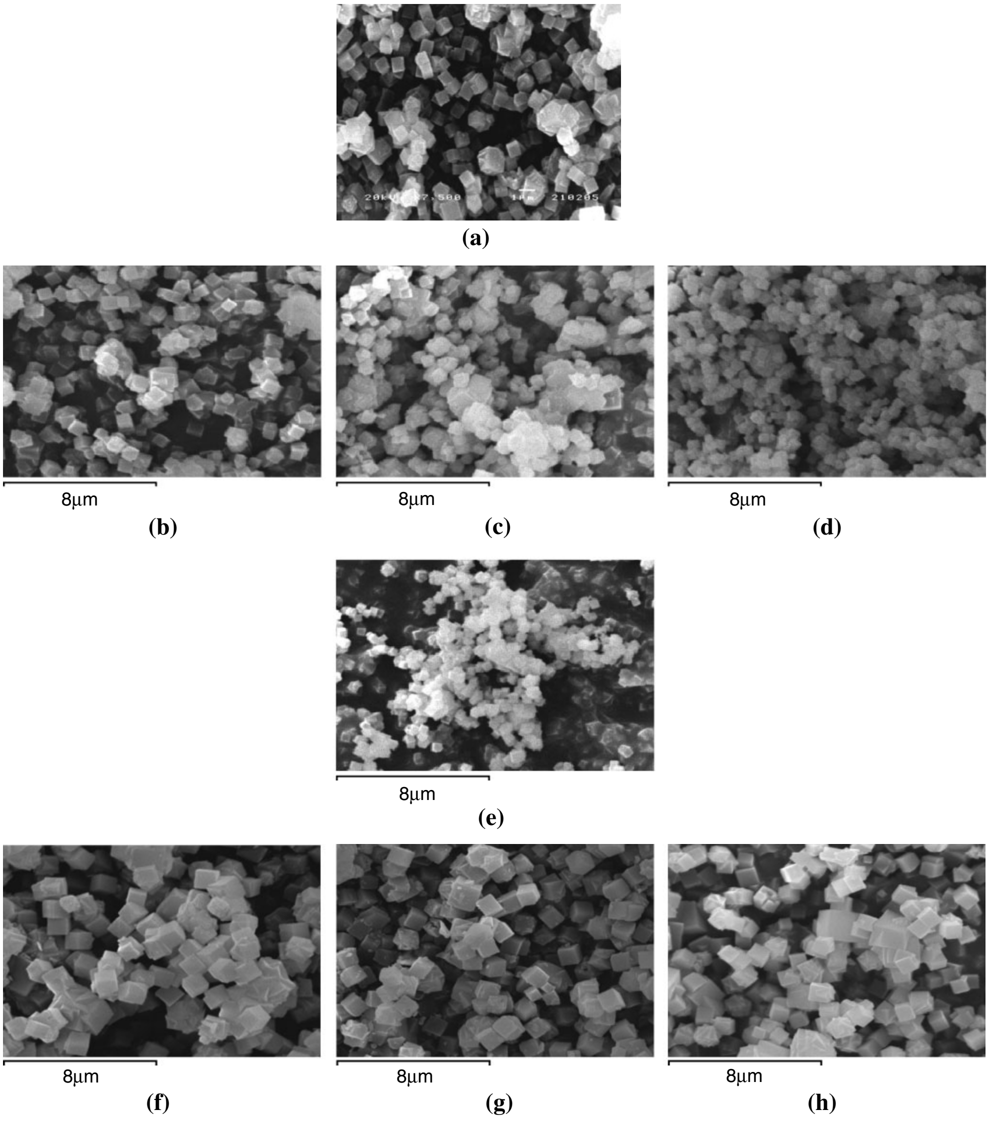

Figure 5. SEM micrographs of synthesized NaA zeolite using $SiO_2 : Al_2O_3 : 3Na_2O{:}410H_2O$ at 100 °C heating temperature for 2 h 20 min with aging temperature 25 °C and seed amount (a) 0, (b) 1, (c) 2 and (d) 3 wt%, and with aging temperature 40 °C and seed amount (e) 0, (f) 1, (g) 2 and (h) 3 wt%.

Effect of microwave temperature

In general, as crystallization is an activated process, within certain limits temperature has a positive influence on the zeolitization process.[13] Increase of the temperature results in shorter crystallization times, as illustrated in Fig. 3. With the same formula of $SiO_2 : Al_2O_3 : 3Na_2O{:}410H_2O$ for a half-day aging time and varying the heating temperatures at 90, 110, 130 and 150 °C for 2 h 20 min, the results showed that the temperature of 90 °C provided mostly amorphous materials with few NaA crystals. Upon increasing the temperature to 110 °C, uniform and homogeneous NaA crystals occurred. However, large crystals with small crystals of NaA from secondary growth emerging on their surface were produced at the high temperatures of 130 and 150 °C.

Temperature strongly influences the formation of NaA zeolite. The optimal temperature range depends on the Si : Al ratio.[14] It can alter the zeolite phase obtained as well as changing the induction period before the start of crystallization.[15] We fixed the composition at $SiO_2 : Al_2O_3 : 3Na_2O{:}410H_2O$, aging for half a day, and heating with microwave at 80, 90, 100 and 110 °C for 2 h 20 min. The results are shown in Fig. 4, which indicates that heating from 80 to 100 °C provided a mostly amorphous material with few NaA crystals. At 110 °C, the uniform and homogeneous growth of NaA crystals is observed.

Effect of aging temperature

For homogeneous crystal size distribution, it would be favorable to have a short nucleation period. A large number of nuclei would result in small crystals. During the aging period, mixing on a molecular scale is envisaged to allow the formation of nuclei necessary for the crystallization of NaA. During microwave synthesis, small crystals which are homogeneous in crystal size distribution are received when the synthesis mixture has been aged sufficiently.[16] Aging of the reaction mixture is frequently carried out at room temperature. However, aging has sometimes been carried out at higher elevated temperatures. In this study, to obtain fully grown crystals, the aging time was fixed (half a day), whereas the aging temperature was varied (25 and 40 °C) concurrently with the effect of seed crystal. Without seeding, when aging temperature was increased to 40 °C, the crystal size was decreased. From the results shown in Fig. 5, crystal size decreases from 1 µm at 25 °C to 0.3–0.4 µm at 40 °C. The results differ from those of other research[17,18] and the mechanism is not clearly understood.

The XRD pattern (Fig. 6) shows only one phase of NaA zeolite compared with the commercial one. Aging at 25 °C with 3 wt% seed, the crystals formed are small and homogeneous at 0.3–0.4 µm. When the seed wt% was decreased to 1–2, the particle size was changed to ~1 µm with non-uniform characteristics. The reason is that the seed crystal provides a surface area on which the required product can grow. As the quantity of added seed material is reduced, the natural supersaturation and consequent self-nucleation will no longer be suppressed and the product crystal size will increase. The XRD results (Fig. 7) show only pure NaA zeolite phase without impurities. However, when aging at 40 °C with 1–3 wt% seed, the product is rather large (~0.8–1 µm) and shows non-uniform crystals. The XRD pattern (Fig. 8) also shows the same results. These phenomena occur due to the growth rate of crystallites, which was strongly influenced by aging temperature due to the fact that the particles formed from the aged solution were agglomerates. Polycrystalline particles have a higher surface area per particle and can therefore assimilate nutrients better than single crystals with regular planar faces.[5] As a result, large crystals grow.

Effect of microwave heating time

Since, in the course of crystallization, the solid product is a mixture of zeolite and unreacted amorphous solids, the crystallinity of the product increases with time.

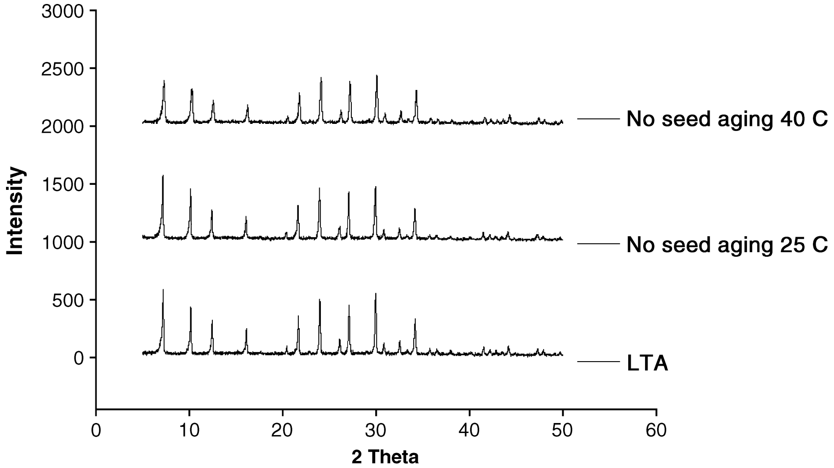

Figure 6. XRD spectra of synthesized NaA zeolite using $SiO_2 : Al_2O_3 : xNa_2O{:}410H_2O$, 110 °C microwave heating for 2 h 20 min with aging at 25 and 40 °C without seeding.

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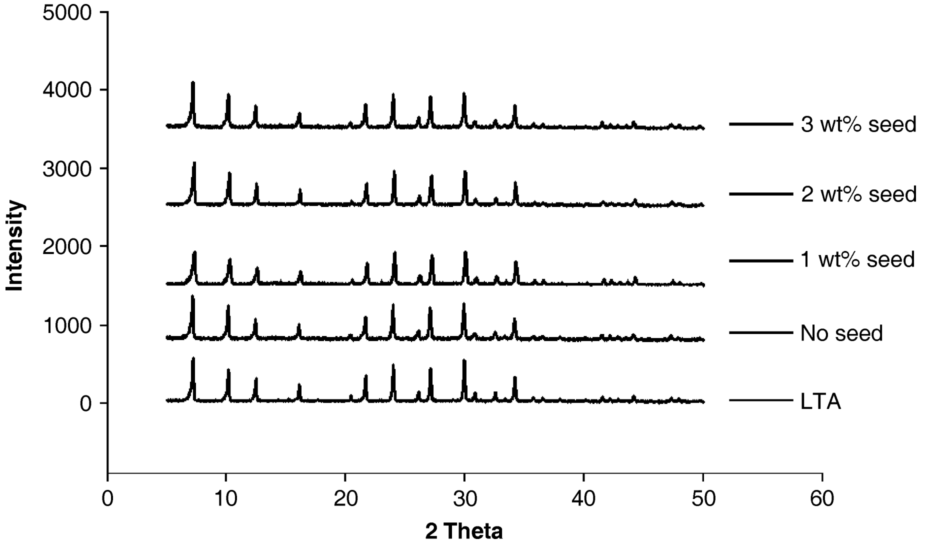

Figure 7. XRD spectra of synthesized NaA zeolite using $SiO_2 : Al_2O_3 : x Na_2O:410H_2O$, 110 °C microwave heating for 2 h 20 min at aging temperature 25 °C with 0, 1, 2, 3 wt% seed.

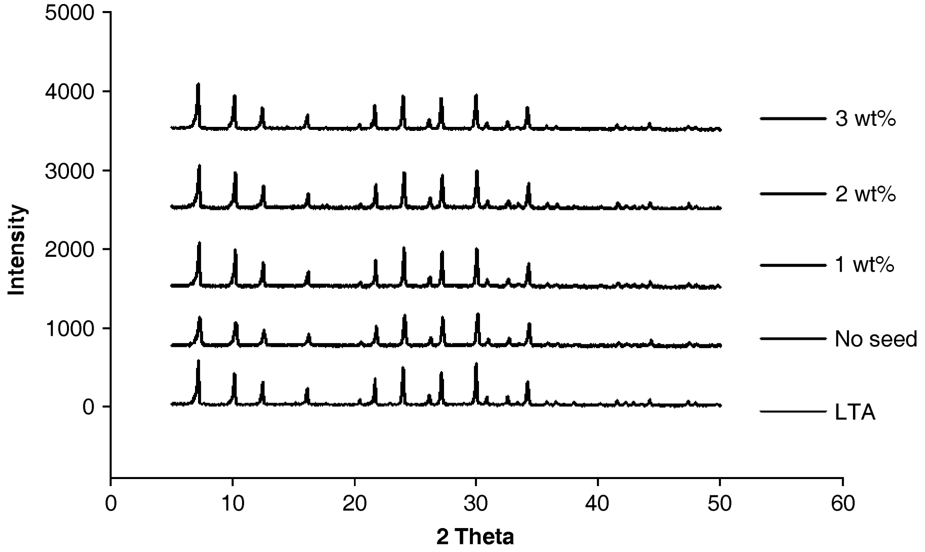

Figure 8. XRD spectra of synthesized NaA zeolite using $SiO_2 : Al_2O_3 : x Na_2O:410H_2O$, 110 °C microwave heating for 2 h 20 min at aging temperature 40 °C with 0, 1, 2 and 3 wt% seed.

Zeolite crystallization is governed by the occurrence of successive phase transformations. Thermodynamically, the least favorable phase crystallizes first, and is replaced by more stable phases.[19] In general, crystallization is an activated process, within certain limitations temperature having a positive influence on the zeolitization process. Higher temperature results in shorter crystallization time.[20] Moreover, the effect of variables, viz. increasing temperature, hydroxide concentration or adding crystal seeds, can be applied to minimize the crystallization time. Not only the temperature, hydroxide concentration, but also the addition of crystal seeds influences the crystallization time.[4] In our system, we fixed the composition at $SiO_2 : Al_2O_3 : 3Na_2O:410H_2O$ with 3 wt% seeding, aging for half a day, and microwave heating at 110 °C for 2 h 20 min. The results (Fig. 9) indicate that, when the temperature was increased to 110 °C, it provided uniform and homogeneous (0.5–0.6 μm) crystals. However, the lower temperature at longer time, such as, 100 °C for 4 h or 90 °C for 5 h, gives the smaller crystal size as 0.5 or 0.3–0.4 μm, respectively. The most uniform and smallest crystals, 0.1–0.2 μm of zeolite product were observed at 80 °C for 6 h because the higher temperature affects the reduction of induction time, speeding the crystal growth rate.[21]

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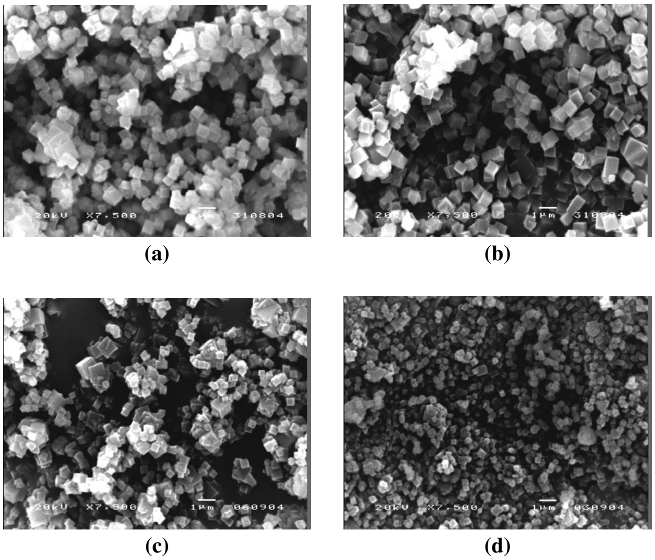
(a) (b)

(c) (d)

Figure 9. SEM micrographs of NaA zeolite synthesized using $SiO_2 : Al_2O_3 : 3Na_2O:410H_2O$, 3 wt% crystal seed at microwave heating temperatures and times of (a) 110 °C for 2 h 20 min, (b) 100 °C for 4 h, (c) 90 °C for 5 h, and (d) 80 °C for 6 h.

Effect of seed wt%

Seed crystals are added to the reaction mixture for reducing the synthesis time and directing the synthesis towards a desired phase.[22–24] The role of seeds in promoting the zeolite crystallization rate is due to the formation of new nuclei on the seed crystal surface, and subsequently the growth of these new crystallites of the same phase.[11] In the sol-gel system, according to Prabir's work,[25] the seed provides the nuclei for crystal growth while the gel's role then is to provide a source of nutrients for nuclei growth. For this present work, we thus fixed the composition at $SiO_2 : Al_2O_3 : 3Na_2O:410H_2O$, aging at half a day, varying seed at 1–3 wt%, and microwave heating at 80 °C for 6 h (see Fig. 10). Uniform and small sized crystals were observed, especially in the 3 wt% seed. The higher amounts of seed promoted a higher nucleation rate, generating more nuclei, while the rate of both nucleation and growth are known to decrease as the temperature is lowered.[26,27] Since the growth rate seems to vary more strongly than the nucleation rate with temperature, as the temperature is lowered, the result is the preparation of small zeolite crystals. SEM-EDS results show the Si : Al ratio equals about 1. The phase of NaA crystal was confirmed by XRD (Fig. 11) compared with the commercial one. Only one phase was observed.

Surface area measurement

The physico-chemistry of all samples is shown in Table 1, which indicates that many samples give high surface area compared with a commercial one. Many effects (such as hydroxide ion concentration, seed wt%, microwave temperature and time) on the surface area of as-synthesized products are studied. It is well known that hydroxide concentration enhances the dissolution of amorphous gel and accelerates the crystal growth. When hydroxide ion concentration is increased to 3 with 3 wt% seed, at 80 °C microwave heating for 6 h, the average crystal size is 0.1–0.2 µm. When hydroxide ion concentration is increased to 4, with the same conditions, the average crystal size is 0.3–0.4 µm. A crystal size of ~0.5 µm was obtained when the hydroxide ion concentration was 5. Seed wt% is the incorporation with NaOH to generate small size NaA zeolite.

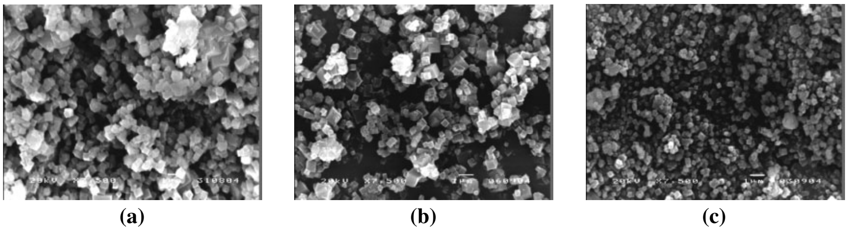
(a) (b) (c)

Figure 10. SEM micrographs of synthesized NaA zeolite using the $SiO_2 : Al_2O_3 : 3Na_2O:410H_2O$ at 80 °C heating temperature for 6 h with seed amount of (a) 1, (b) 2 and (c) 3 wt%.

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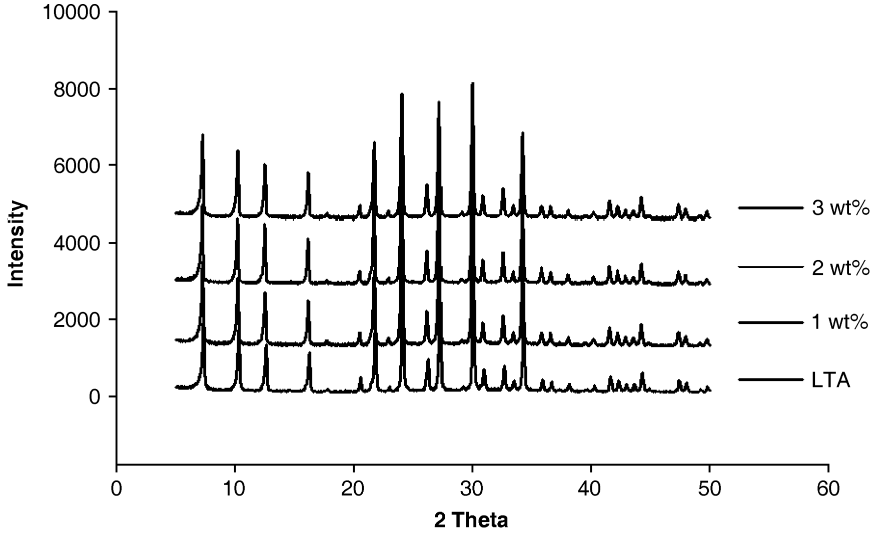

Figure 11. XRD spectra of synthesized NaA zeolite using $SiO_2 : Al_2O_3 : 3Na_2O : 410\ H_2O$ at 80 °C heating temperature for 6 h with seed amounts of 1, 2 and 3 wt% compared with the commercial one.

Table 1. Surface area of as-synthesized NaA zeolite by varying hydroxide ion concentration (3, 4, 5), seed wt% (1, 2, 3) and microwave temperature and time (80°/6 h and 90 °C/5 h) with composition $SiO_2 : Al_2O_3 : xNa_2O{:}410H_2O$; $3 \leq x \leq 5$

NaA synthesis condition			BET surface area (m^2/g)
NaOH (mole ratio)	Seed wt%	Microwave temperature/time	
3	1	80 °C/6 h	7.3
3	2	80 °C/6 h	9.4
3	3	80 °C/6 h	11.9
3	1	90 °C/5 h	5.0
3	2	90 °C/5 h	6.2
3	3	90 °C/5 h	9.0
4	1	80 °C/6 h	7.3
4	2	80 °C/6 h	7.4
4	3	80 °C/6 h	8.8
4	1	90 °C/5 h	3.8
4	2	90 °C/5 h	5.1
4	3	90 °C/5 h	8.7
5	1	80 °C/6 h	2.7
5	2	80 °C/6 h	3.1
5	3	80 °C/6 h	7.9
5	1	90 °C/5 h	1.1
5	2	90 °C/5 h	1.8
5	3	90 °C/5 h	6.2
Commercial NaA (Sigma Aldrich Co. Ltd)			5.9

For hydroxide ion concentrations equal to 3, 4 and 5, as more seed is applied, small size NaA zeolite is obtained due to the seeds providing their surface area to assimilate material from the solution for nuclei formation, leading to the small crystal. From Table 1, with the same hydroxide ion concentration, the surface area of NaA zeolite increases when greater seed wt% is applied. An example is that, at a hydroxide concentration equal to 3 with 1, 2 and 3 seed wt%, higher surface areas of 7.3, 9.4 and 11.9 m^2/g, respectively, at a microwave heating temperature of 80 °C for 6 h, are shown. Temperature and time can alter the surface area of the NaA crystal. Temperature affects factors in zeolite synthesis, reducing the length of induction time and enhancing the crystallization rate when temperature increases. For instance, with a hydroxide concentration equal to 3 with 3 wt% seed, the higher temperature of 90 °C for 5 h provides a larger crystal (0.3–0.4 μm) compared with the small crystal (0.1–0.2 μm) obtained from the same condition, but with heating at 80 °C for 6 h. Thus, the surface area of the crystal from 80 °C/6 h (11.9 m^2/g) is higher than the one from 90 °C/5 h (9.0 m^2/g). Most of the synthesized NaA zeolites display high surface areas compared with the commercial one (Sigma Aldrich Co. Ltd).

Moisture absorption testing

Moisture absorption of the small size (0.1–0.2 μm, having surface area of 11.9 m^2/g) synthesized zeolite, using $SiO_2 : Al_2O_3 : 3Na_2O{:}410H_2O$ at 80 °C for 6 h was determined by the method described elsewhere.[6] The results of water absorption of NaA zeolite synthesized was determined to be 0.1338 g H_2O/g zeolite, which is higher than the 0.1186 g H_2O/g zeolite obtained from the commercial one (5.9 m^2/g). The reason is that the smaller size of the synthesized NaA (0.1–0.2 μm) involves higher surface area for the absorption of water, more than the commercial one (2 μm).

NaA zeolite is hydrophilic and it has potential for the dehydration of an organic mixture. NaA zeolite is thus used for membrane applications. Xu *et al.*[28] synthesized a NaA zeolite membrane by the microwave technique in which uniform and small sized zeolite was formed from nuclei on the support surface homogeneously and in short duration. A thin layer of NaA zeolite membrane was received, resulting in increasing permeation rate.

CONCLUSIONS

Nanosize NaA zeolite can be successfully synthesized from silatrane and alumatrane by the sol–gel process and microwave techniques. The composition of $SiO_2 : Al_2O_3 : 3Na_2O{:}410H_2O$ with 1–3 wt% of seed crystals obtained using reaction temperatures ranging from 80 to 100 °C provides uniform and narrow size distribution of 0.1–0.2 μm NaA zeolite crystals.

Acknowledgments

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