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Synthesis and characterization of NaY zeolite from Bayat natural zeolite: effect of pH on synthesis

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Abstract. Herein, NaY zeolite was synthesized by a hydrothermal method via seeding using Bayat natural zeolite with a Si:Al ratio of 6.26, which comprised the silica and alumina sources as well as sodium aluminate as the additional alumina source. Poorly crystalline natural zeolite contains several impurities; thus, activation and purification are crucial for removing some impurities and widening the zeolite pores. Before synthesis, Bayat natural zeolite was subjected to dealumination to remove some alumina and fragmentation to break the zeolite framework into their monomers. In addition, the effect of pH on synthesis was investigated. NaY zeolite was characterized via X-ray diffraction (XRD) and Fourier transform infrared (FTIR) spectroscopy equipped with energy-dispersive X-ray spectroscopy (EDX). The XRD patterns revealed that a pH of 11–12 is typical for synthesizing NaY zeolite. Different FTIR spectra were observed for the raw, fragmented, and as-synthesized NaY. The EDX data revealed that NaY zeolite synthesized at pH values of 11–13 exhibits Si:Al ratios of 5.35, 3.72, and 2.96, respectively.

Keywords: NaY zeolite, natural Bayat zeolite, fragmentation method, seeding method, pH effects

1. Introduction

NaY zeolite exhibits a faujasite-type framework with saturated sodium (Na⁺) cations, which plays an important role in the petrochemical industry because it exhibits some favorable properties, including high catalytic activity, atomic surface ratio, specific surface area, and surface energy [1]. NaY zeolites have been widely used in separation and ion exchange as well as acid catalysts for size- and shape-selective catalytic reactions related to their molecular-sized microporosity [2]. In this study, Bayat natural zeolite was used as the main silica and alumina source with a dominant mordenite structure [2]. Poorly crystalline natural zeolite contains some minerals, which are unevenly distributed, with several impurities [4]. The presence of these impurities can decrease the activity of zeolite; hence, it is crucial to activate and purify zeolites. Before synthesis, Bayat natural zeolite was subjected to dealumination and fragmentation. pH is a crucial factor during synthesis [3]. At an extremely high pH, the solubilities of the Si and Al sources can increase, thereby possibly accelerating the polymerization rates of polysilicates and aluminate anions. This in turn leads to an increase in the amount of time required for shorter nucleation in order to accelerate the crystallization time. However, extremely high crystallization rates may lead to the formation of an imperfect zeolite framework or other zeolite frameworks [4]. Therefore, Bayat natural zeolite is synthesized before its conversion into NaY zeolite and the effect of pH on synthesis is investigated.



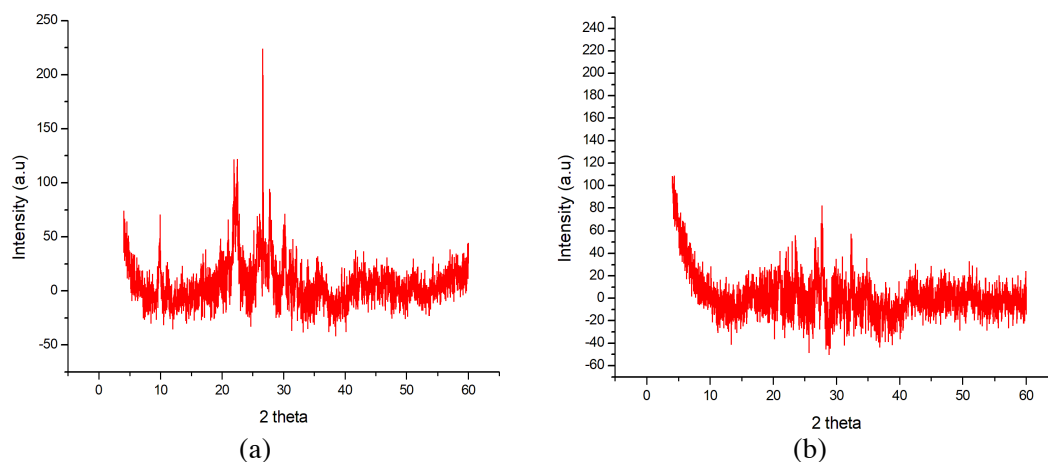


Figure 1. (a) Raw Bayat natural zeolite and (b) fragmented Bayat natural zeolite

2. Experimental

2.1. Synthesis of Bayat natural zeolite

2.1.1. Activation. Bayat natural zeolite was dispersed in deionized (DI) water at a ratio of 1:3 (w/v) and heated at 300 °C for 3 h to separate zeolite from impurities.

2.1.2. Purification. To dissolve carbonate salts, zeolite was dispersed into a pH 5 sodium acetate buffer at a ratio of 1:3 (w/v) at 80 °C for 1 h. Next, the solution was decanted and stirred again by the dropwise addition of DI water and a 30% H₂O₂ solution with simultaneous heating (in case of bubbles, 0.1 M of an acetic acid solution was added until bubbles were not observed) [5]. The mixture was decanted, and a precipitate was obtained by the addition of a Na-citrate-bicarbonate solution and sodium dithionite and stirring at 80 °C for 30 min. The precipitated zeolite was washed using DI water and then dried at 105 °C [6].

2.1.3. Dealumination. This step involves the reduction of the levels of alumina in zeolite. The purified zeolite was dissolved with 2-M HCl at a ratio of 1:10 (w/v) and stirred at 100 °C under reflux for 2 h.

2.1.4. Fragmentation. Zeolite and NaOH were homogenized at a ratio of 1:2, followed by the addition of DI water, stirring until homogeneity was attained, and heating at 250 °C for 2 h.

2.2 Synthesis of NaY zeolite

The seed gel was synthesized with a molar ratio of 10.67 Na₂O:Al₂O₃:10 SiO₂:180 H₂O [7]. A mixture of the seed gel was stirred in a polypropylene (PP) bottle and then aged for 24 h at room temperature. For synthesizing NaY zeolite, Bayat natural zeolite was used as the silica and alumina source and sodium aluminate was used as an additional alumina source. For this synthesis, the molar ratio was 6.25 Na₂O:Al₂O₃:10 SiO₂:180 H₂O. A mixture of Bayat natural zeolite, sodium aluminate, and DI water was stirred for 48 h, with the addition of varying volumes of glacial acetic acid (pH 11, 12, and 13). Next, the seed gel was inserted into a PP bottle and heated at 100 °C for 24 h. After heating, two phases were formed. Then, the precipitate phase was washed with DI water until a pH of 9 was attained, following by drying at 60 °C.

3. Results and discussion

3.1 Characterization of Bayat natural zeolite

The X-ray diffraction (XRD) patterns of the raw and fragmented Bayat natural zeolite (figure 1) revealed poor crystallinity for the raw zeolite; peak shifts at 19.10°, 20.74°, and 20.86° after fragmentation; and the loss of some peaks at 21.94°, 22.48°, and 26.60°. The XRD patterns also revealed the presence of a high amount of amorphous materials, indicating successful fragmentation.

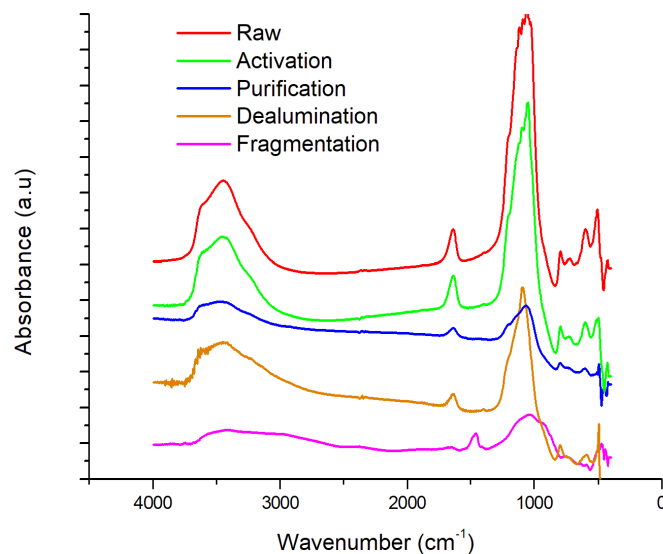


Figure 2. Fourier transform infrared (FTIR) spectrum of Bayat natural zeolite

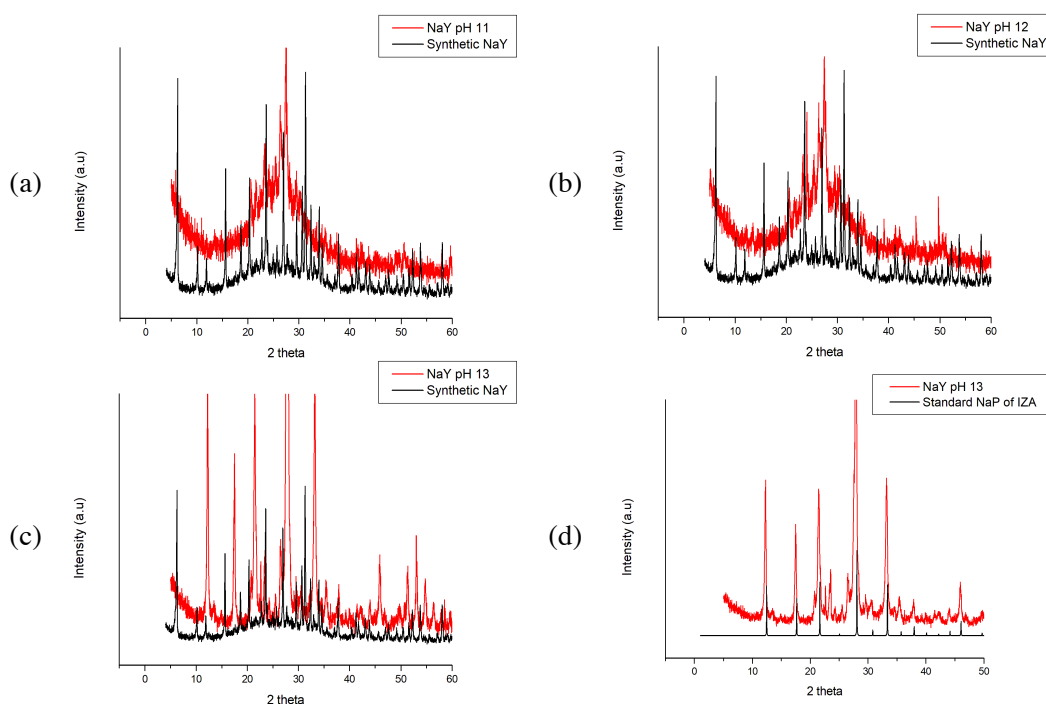


Figure 3. Comparison of NaY zeolite at (a) pH 11, (b) pH 12, (c) pH 13, and (d) pH 13 with that of standard NaP zeolite from the IZA database.

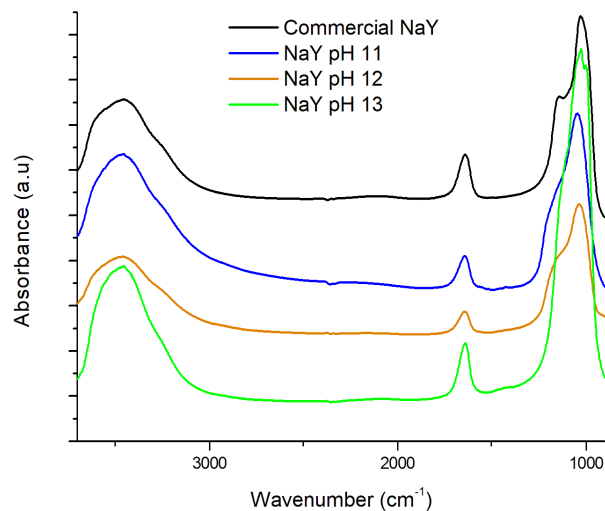
The Fourier transform infrared (FTIR) spectrum of the raw Bayat natural zeolite was compared with that of the fragmented natural zeolite (figure 2). Raw zeolite adsorbed a considerable amount of water, as can be observed from the peaks at 4000–3000 cm^{-1} and 1620 cm^{-1} , corresponding to the molecular water adsorbed on the zeolite surface. After fragmentation, a small peak was observed at 1500 cm^{-1} , corresponding to an aluminum oxide compound. This result indicated that the alumina-silicate framework collapses, affording smaller fragments. The FTIR analysis results supported the XRD data.

Table 1. Energy-dispersive X-ray spectroscopy (EDX) data of as-synthesized NaY.

Element (%wt)	Raw	NaY		
		pH 11	pH 12	pH 13
Si	37.84	38.02	35.09	30.87
Al	6.04	7.1	9.42	10.42
Na	0.42	4.02	6.4	6.63
Si/Al ratio	6.26	5.35	3.72	2.96

Table 2. Water adsorption capacity of as-synthesized NaY.

Zeolite	% Weight loss	Weight after adsorption of water (g)
NaY pH 11	3.92	0.5043
NaY pH 12	4.83	0.5244
NaY pH 13	13.82	0.5351

**Figure 4.** FTIR spectrum of as-synthesized NaY.

3.2. Characterization of NaY zeolite

The XRD patterns of NaY as-synthesized at pH 11–13 revealed the formation of crystalline NaY compared with that observed for synthetic NaY zeolite. This result was confirmed by comparing with the XRD pattern of standard NaY and NaP zeolite obtained from the International Zeolite Association (IZA) database (figure 3). NaY zeolite was not completely formed at pH 11 and 12, but some similarities to the synthetic NaY zeolite XRD pattern at 2θ values of 6.73° , 10.91° , 23.23° , and 27.47° were observed. This result is probably related to the presence of cations other than Na, such as K^+ , Mg^{2+} , Ca^{2+} , and Fe^{2+} , which were not removed during purification [8]. In addition, an uneven baseline was observed in the XRD patterns of NaY at pH 11 and 12, indicating a mixed amorphous fraction in zeolite. However, for NaY prepared at pH 13, the XRD pattern revealed an extremely high crystalline structure, but this structure was found to be similar to that of P zeolite based on its comparison with the XRD pattern of the standard NaP zeolite from the IZA database because of the typical peaks observed for P zeolite. Nevertheless, the XRD patterns revealed that the Bayat natural zeolite structure was successfully destroyed and converted into a different zeolite structure.

A comparison of the FTIR spectra of NaY prepared at pH values of 11, 12, and 13 with that of commercial NaY is shown in figure 4. From the band observed at 1600 cm^{-1} , NaY prepared at pH 13 exhibited the highest H–OH intensity. This result was in agreement with the energy-dispersive X-ray spectroscopy (EDX) data.

The EDX data (table 1) revealed that the lower the Si:Al ratio, the higher the Na ratio. Thus, NaY prepared at pH 13 exhibited high polarity and can adsorb a considerable amount of water.

The water adsorption capacity of as-synthesized NaY was investigated (table 2). From the % weight loss of zeolite after activation at 200 °C was in agreement with the EDX data.

4. Conclusions

In this study, NaY zeolite was prepared from Bayat natural zeolite, which was subjected to dealumination and fragmentation. A pH of 11–12 was successful for the synthesis of NaY zeolite, which was verified via the XRD pattern and the fact that Si:Al ratio was similar to that of synthetic NaY. However, at pH 13, multiple hydroxyl (–OH) groups were present, leading to the dissolution of silica and the formation of the NaP framework. Furthermore, a low Si:Al ratio in NaY leads to a high polarity, implying a high ability for adsorbing water.

Acknowledgements

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