Paper 11-Teguh Kurniawan

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Effect of Intermittent Agitating During Hydrothermal Synthesis on Mordenite Properties and Ammonium Adsorption

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Abstract. Mordenite is well-known as a commercial catalyst in oil refineries. Conventionally, mordenite is produced through hydrothermal method under static condition for long period of time. In this investigation, the effect of intermittent agitating of the solution during the hydrothermal process on the mordenite properties and ammonium sorption was studied. The synthesized mordenite samples were characterized by X-ray diffraction (XRD) and Scanning Electron Microscope (SEM). The XRD pattern of the intermittent agitated sample showed broader peaks with similar intensity peaks as compared to the non-intermittent agitated sample. According to the SEM images, the average particle size of the intermittent agitated sample was 1.8 μm, while the non-intermittent agitated sample was larger with 13 μm. Both of samples show similar morphology as ellipsoid like. The sample of mordenite from the intermittent agitated sample showed a lower bulk density and high particle dispersion in water. The ammonium adsorption on the mordenite samples were tested by using various volume of ammonium solution. The ammonium adsorption capacity of the intermittent agitated sample slightly higher than the non-intermittent agitated mordenite. This simple intermittent agitated action which favored smaller particle size of mordenite could be beneficially applied for other applications dealing with bulky molecules due to the lower mass transfer limitation.

INTRODUCTION

Mordenite is a type of zeolite which has 12 Membered Ring (MR) with 8 MR and another 8 MR bridging the 12 MR and the 8MR. Despite it has three types of interconnected pores, mordenite is considered as 1 dimensional pore due to non-accessible 8 MR pores [1]. Mordenite has a wide sorption application such as removal Zn^{2+}, NH_4^+, Cu^{2+}, Ni^{2+}, Co^{2+}, and Fe^{2+} [2][3][4]. Mordenite firstly coined by How after the place where it was found in Morden, Nova scotia, Canada [5]. The natural mordenite is easily found in Indonesia such as in Sumatera, Java, Nusa Tenggara and Sulawesi. Mordenite can also be synthesized from precursors silica, alumina, and sodium hydroxide. Mordenite is synthesized by using silica gel which could be obtained from non-renewable and renewable sources [6]. It is Barrer was who was invented the hydrothermal method to synthesize mordenite in 1948 [7].

Hydrothermal synthesis is conventionally executed by heating the gel in a static condition in the oven for 2-4 days. Long period of hydrothermal process favored a dense precipitated solid which inhibited mass transfer between the precursors in solution and the solid. Recently, Chen et.al. reported that the intermediate stirring was triggered the rapid crystallization of mordenite [8]. The fast crystallization was occurred because of the loosen packed solid phase improve dissolution of aluminosilicate promoted crystallization. However, further comparison on application of the mordenite with and without intermittent stirring was not investigated. Mordenite could be applied in small aquaculture

and intensively-stocked transport and holding units for aquatic animals for minimizing ammonium content [9]. Ammonium in water and wastewater contribute to eutrophication in lake, river, and others body water [10][11]. Mordenite as a porous material adsorbs ammonium mainly through ion exchange principle. The cations attached on the negatively charge. In this investigation, the effect of intermittent stirring during hydrothermal synthesis on the synthesized mordenite properties was studied. Furthermore, the mordenite was tested for ammonium adsorption application by varying the volume of ammonium solution.

EXPERIMENTAL

Mordenite Synthesis

Mordenite was synthesized by hydrothermal method with molar ratio of $30SiO_2$:Al_2O_3:$6Na_2O$:$780H_2O$ at 170 °C. An amount of 1.231 g NaOH (Merck) was dissolved in 37.140 g demineralized water. An amount of 0.630 g sodium aluminate (Sigma Aldrich) which consist of 53% Al_2O_3 and 42% Na_2O was added into solution. An amount of 14.76 g colloidal silica 40% (Sigma Aldrich) was added into the solution. The hydrothermal synthesis was conducted in autoclaves. Three samples were treated in oven for 48 h with and without stirring interruption. Prior to stirring, the sample was crushed using spatula. After that, the mixing was accomplished by magnetic stirrer for 5 min. Sample A was hydrothermally treated in the oven for 48 h without interrupting of agitation. Sample B was agitated at 17 h and return into the oven until 48 h. Sample C was agitated at 17 h and 24 h and keep in the oven until 48 h. The solid separated from the liquid and washed several times until pH less than 10.

Characterization

Mordenite was observed visually by dispersing in water to study qualitatively the degree of solubility. Bulk density was determined by weighing some amount of mordenite in a fixed volume of vial. X-ray diffraction (XRD) was performed to study the crystallinity and zeolite phase. The morphology and particle size were determined by Scanning Electron Microscope (SEM).

Ammonium Adsorption

Ammonium solution with low concentration 23 ppm was prepared by dissolving NH_4Cl in water. Adsorption test was performed over the samples by using various ammonium solution volume, i.e. 10 mL, 150 mL and 250 mL. The mordenite sample was 0.5 g. Time for adsorption test was 2 h. Ammonium concentration was tested by using colorimeter method (Hanna).

RESULTS AND DISCUSSION

Characterization

Figure 1 presented the observation of dispersing mordenite samples in water. At the beginning of dispersion, all samples were homogeneous as presented in Figure 1.a. Sample C showed a well dispersed particle in the water followed by sample B, and sample A. After 150 min, sample A showed clearer solution followed by sample B and sample C as presented in Figure 1.b. The sample A is much easier to settle as compared to sample B and C. It is most likely that particle of sample A has a large size as the crystallization step during the hydrothermal process was not interrupted by stirring. On the other hand, sample B and sample C has smaller particle because of the stirring step during hydrothermal synthesis at t=17 h and 24 h were performed. Chen et al, concluded that the intermediate stirring improve the dissolution of aluminosilicate and enhance the crystallization as well as reduce the particle size [8].

(a) (b)

FIGURE 1. (a) Dispersion of mordenite in water at t=0 and (b) after t= 150 min.

Bulk density of mordenite samples is presented in Table 1. As we expected, sample A showed the highest bulk density. This is in agreement with the dispersion observation which showed that sample A was the fastest sample to settling. On the other hand, sample C showed the lowest bulk density. The intermittent stirring of sample C at 17 and 24 h lead to a lighter particle. The number of crystallization nuclei are much higher hence the crystallized particle formed smaller than the sample without interrupted stirring. During the hydrothermal time, the precipitated solid formed favours a low mass transfer from the solution into the solid.

TABLE 1. Bulk density of mordenite samples

Sample	**Bulk density (g/cm^3)**
A	0.47
B	0.27
C	0.21

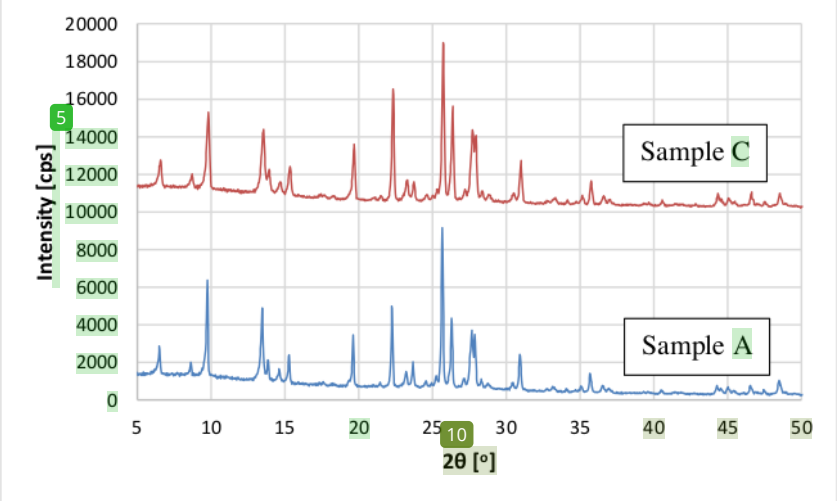
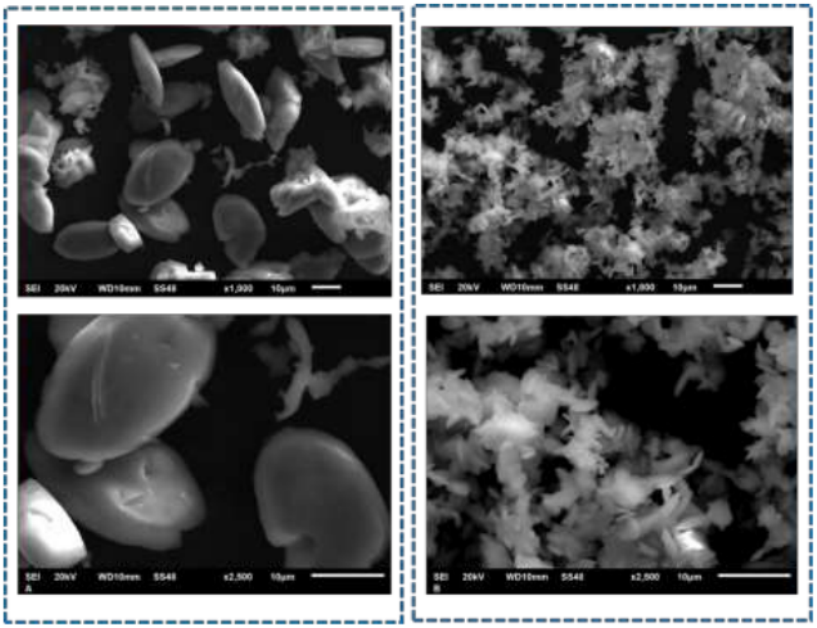


Figure 2. XRD pattern of mordenite synthesized without and with intermittent stirring.

Figure 2 presents the diffraction pattern of sample A and C. Both of the samples show a pure mordenite crystal as compared with the XRD pattern of simulated mordenite [12]. The peak width of sample C is broader than sample A. Scherrer equation is presented in equation 1.

$$B(2\theta) = \frac{K\lambda}{Lcos\theta} \qquad (1)$$

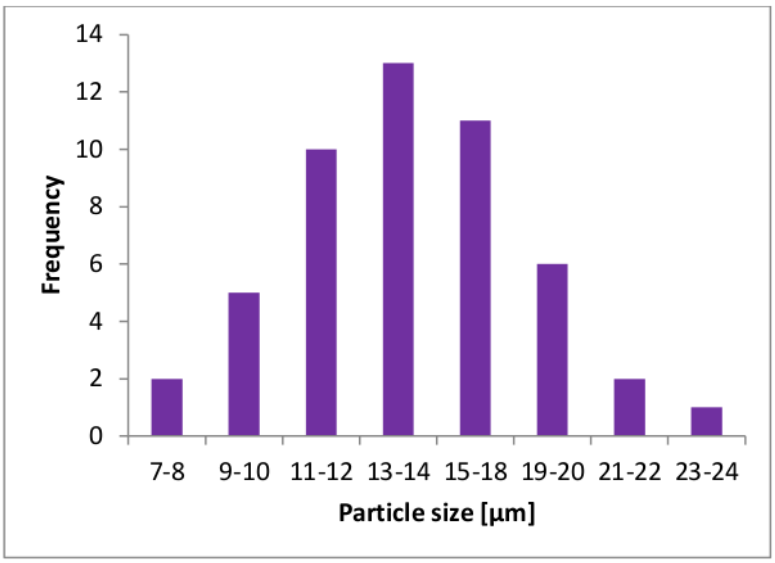
B is peak width and L is crystallite size. According to Scherrer equation. Peak width is inverse proportionally to the crystallite size. The peaks of sample C shows a broader peak width as compared with sample A which indicated that the crystallite size of sample C smaller than sample A.



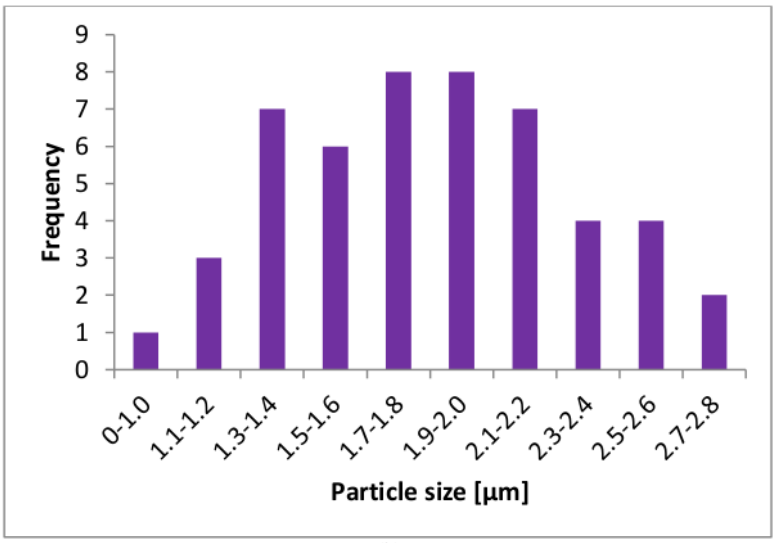
(a) (b)

Figure 3. SEM images of synthesized mordenite samples (a) without intermittent stirring (Sample A) and (b) with intermittent stirring (Sample C).

Mordenite morphology of all samples were ellipsoid like as presented in Figure 3. The difference between sample A and C is on the particle size. It can be seen clearly that particle size of sample with intermittent stirring is smaller than particle size of sample without stirring. Figure 4 shows that the average particle size of sample C is 1.8 μm while average particle size of sample A is 13 μm.



(a)



(b)

Figure 4. Particle size distribution (a) without intermittent stirring (Sample A) and (b) with intermittent stirring (Sample C).

Ammonium Adsorption

Ammonium concentration after adsorption using mordenite samples in various solution volume were presented in Figure 2. Ammonium sorption is affected by mass of zeolite, ammonium concentration, pH [2][13]. The ammonium was adsorbed completely when using only 10 mL of solution for all mordenite samples. At this lower solution volume,

the effect of intermittent stirring on the ammonium sorption was not significant. However, at higher volume 150 and 250 mL the samples with intermittent stirring show a higher ammonium sorption than the sample without intermediate stirring. It is most likely because of the sample with intermediate stirring has a high dispersion in solution hence much easier to contact with the ammonium. The zeolite prepared by continuously rotating autoclave show a much smaller and pure crystal zeolite [14].

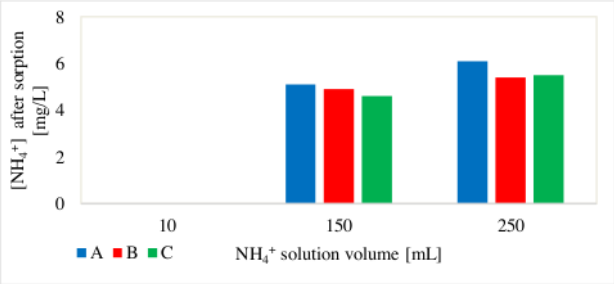


FIGURE 7. Ammonium concentration after adsorption with mordenite samples for 2 h at various solution volume.

CONCLUSION

Mordenite has been successfully synthesized by hydrothermal method with and without intermittent stirring during the heating process in the oven. The sample of mordenite from the intermittent stirring shows a smaller particle size, lower bulk density, high dispersion in water, and higher ammonium adsorption capacity than mordenite without intermediate stirring. The mordenite sorption was fast and remove all the ammonium at lower solution volume. . This simple intermittent agitated action which favored smaller particle size of mordenite could be beneficially applied for other applications dealing with bulky molecules due to the lower mass transfer limitation.

ACKNOWLEDGMENTS

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