Synthesis of Zeolite A by Ultrasound Irradiation Technique

[1]H. M. Razif, [1]N. H. N. Hadzuin, [2]T. Sugeng and [1]A. J. Aishah.

[1]Faculty of Chemical and Natural Resources Engineering,
[2]Ibnu Sina Institute for Fundamental Science Studies,
Universiti Teknologi Malaysia, 81310 UTM Skudai, Johor, Malaysia

Email: aishah@fkkksa.utm.my

Abstract

Synthesization of Zeolite A from colloidal silica by ultrasound irradiation technique was compared with the material obtained by hydrothermal technique. The composition was at molar ratio of 1 Al_2O_3: 0.85 SiO_2: 3.0 Na_2O: 200 H_2O. X-Ray diffraction (XRD) and Scanning Electron Microscope have been used to characterize the products. Although, XRD pattern of both samples showed typical peaks of zeolite A, intensity of peaks were higher for ultrasound irradiation technique compared to the hydrothermal technique. Moreover, SEM analysis indicated the crystal size of zeolite A synthesized by ultrasound irradiation technique (1.7-5.2μm) was smaller compared to the hydrothermal technique (1.9-5.5 μm). Thus, ultrasound irradiation has a potential to be applied in synthesizing zeolite A.

Keywords: Zeolite A, Ultrasound irradiation technique, XRD, SEM

Introduction

Nowadays, much effort has been made to synthesize zeolite A by using various methods. However, all of these methods have their advantages and disadvantages. Most of the studies reported in the literature were on hydrothermal technique (Sathupunya, et al., 2003; Jansen, 2001; Rollman et al., 1999). The disadvantages of this method were due to the process itself which takes more time to react and the amount of zeolite produced was low and the exact yields were often not given (Jansen, 2001).

The introduction of ultrasound irradiation in the synthesis of zeolites offers some advantages over the conventional synthesis in terms of higher crystallization rate, yield enhancement and eventually a shorter crystallization time compared to hydrothermal technique (Zhang, et al., 2006). Recently, there has been an effort to synthesize zeolite A via ultrasound technique. Synthesis of zeolite A from clear a clear-to-the-eye sodium aluminosilicate solution was carried out in the presence of ultrasound at different temperatures and time and the results were compared to those obtained by performing conventional static synthesis under similar conditions. In hydrothermal technique, spontaneous mixing of the reaction mixture may occur through conviction or mechanical stirring and can be employed to homogenously distribute the reactants and the temperature throughout the reaction vessel. The highest temperature that can be achieved is limited to the boiling point of the particular mixture which higher boiling solvent must be used. In contrast, it was possible to obtain highly crystalline zeolite A in the presence of ultrasound, which would influence the types and stability areas of the phases that were formed during metastable phase transformations. Nucleation and crystallization rates, as well as the yield of zeolite A increased as a result of the application of ultrasound. The result showed the optimum conditions were at temperature of 50°C and 7 to 8 hours reaction times under ultrasound irradiation technique. (Andac, et al., 2006).

In the present work, zeolite A has been synthesized from colloidal silica under ultrasound irradiation and compared with hydrothermal technique. To the best of our knowledge, zeolite A was first time synthesized from colloidal silica by using ultrasound irradiation and has never been reported before.

Materials and Method

Experimental

Synthesis mixtures have the following molar ratio: 1 Al_2O_3: 1.96 SiO_2: 3.165 Na_2O: 128 H_2O. Sodium aluminate, NaOH and distilled water were mixed together and the solution was boiled until it was

clear. Once the solution had cleared up, the colloidal silica and NaOH was slowly added under stirring and the mixture stirred for one hour at 95°C. The prepared solution with the substrate was transferred into Teflon vessel and placed into ultrasound to get radiation for 1-3h at 60°C and 80°C. The substrate was filtered until pH <9 in order to eliminate any impurities and an excess of NaOH and dried for 25min at 110°C. Synthesis zeolite A by hydrothermal technique was the same procedure as mentioned before. The prepared solution with the substrate were placed in the oven for treatment at 110°C for 4h and dried for overnight.

Characterization

X-Ray powder diffraction pattern of the sample was recorded on a JEOL X-Ray Diffractometer JDX-3500 with a Cu-K□□ 40kV, 45 mA radiation sources. The diffraction spectrum range was 20-90° and the scanning speed and step were 1 sec and 0.050. The size of the particle of zeolite A was determined by JSM-6300F scanning electron microscopic analysis (SEM).

Results and Discussion

Effect of temperature

Zeolites which were crystallized by both ultrasound irradiation and hydrothermal technique were analyzed by X-ray diffraction. XRD patterns of zeolites obtained by means of a hydrothermal technique and by the ultrasound irradiation are reported in Fig.1. It can be seen that the typical diffraction peaks of zeolite A obtained by both samples at 60°C. However, the intensity of the diffraction peaks of the zeolite A crystal increases as the temperature rises from 60°C to 80°C under ultrasound irradiation compared to hydrothermal technique, which implied that more zeolite A crystals were grown on the substrate.

Fig. 2 illustrates the scanning electron microscope of zeolite A samples synthesized by both microwave and hydrothermal technique. SEM analysis indicates the crystal size of zeolite A synthesized at 60°C by ultrasound irradiation technique (1.7-5.2μm) was smaller in comparison to the hydrothermal technique (1.9-5.5 μm). However, there was only slight crystal size different when the temperature of ultrasound increase to 80°C, in a range 1.9-5.7 μm. This discrepancy is attributed to different nucleation processes in the different temperature.

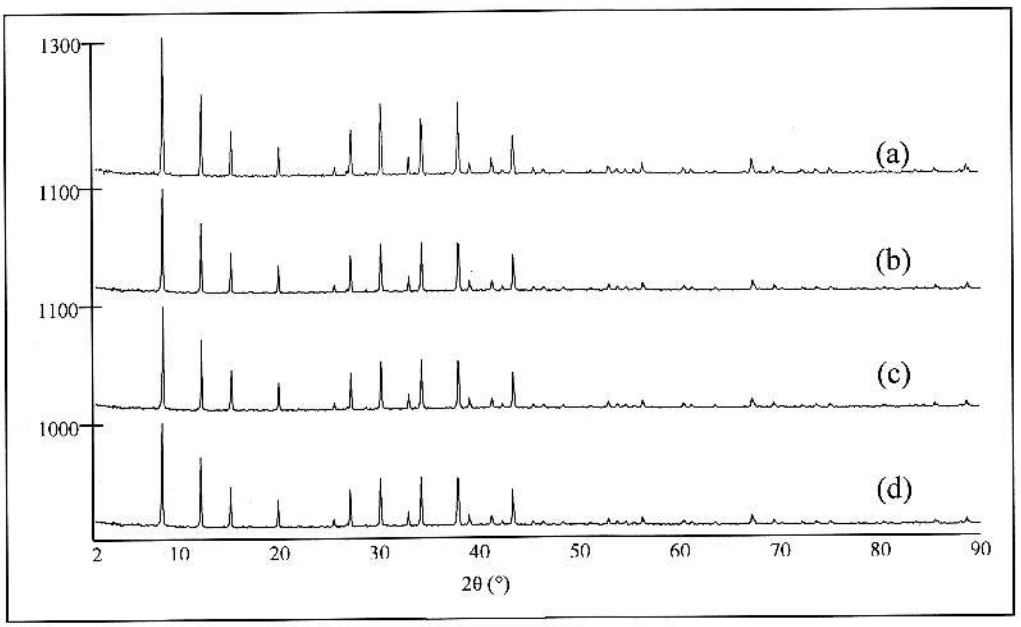
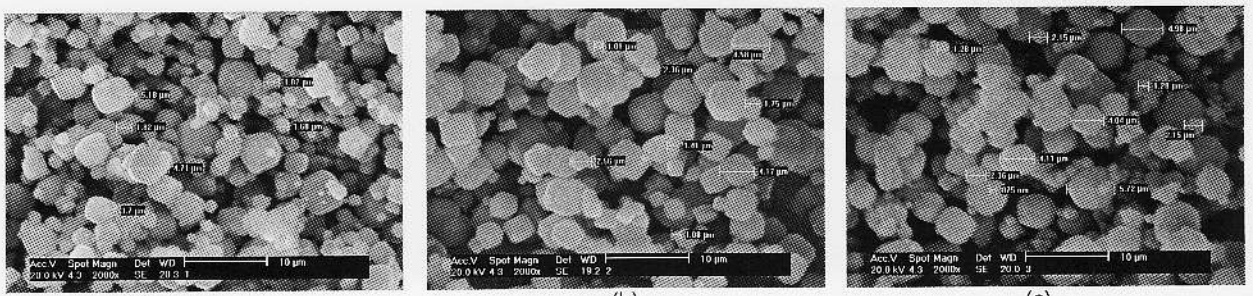


Fig1: XRD pattern for zeolite A synthesized by a) hydrothermal, 80°C; b) Ultrasound Irradiation, 80°C; c) hydrothermal, 60°C; d) Ultrasound irradiation, 60°C



(a) (b) (c)

Fig2: SEM analysis for zeolite A synthesized by a) hydrothermal, 60°C; b) Ultrasound Irradiation, 60°C; c) Ultrasound irradiation, 80°C

Effect of time

In order to investigate the optimum condition for crystallization of zeolite A under ultrasound irradiation, these experiments were carried out at 80°C for 1h, 2h and 3h. XRD diffractogram in Fig. 3 showed that the intensity of zeolite A for 1h is quite similar with 3h of synthesizing time under ultrasound irradiation. In contrast, the result of XRD for 2h in synthesizing zeolite A gave the lowest intensity compared to the other samples. This is due to the pressure that may be varies in different batch testing. Therefore, 1h has been considered as an optimum duration for crystallization of zeolite A under ultrasound irradiation.

Fig. 4 illustrates the scanning electron microscope of zeolite A samples synthesized in three different duration, 1h, 2h and 3hr at 80°C. SEM images have shown almost no-different in crystal size of zeolite A synthesized by all of these samples. Thus, the crystallization of zeolite A by ultrasound irradiation has not increased rapidly with increasing the time.

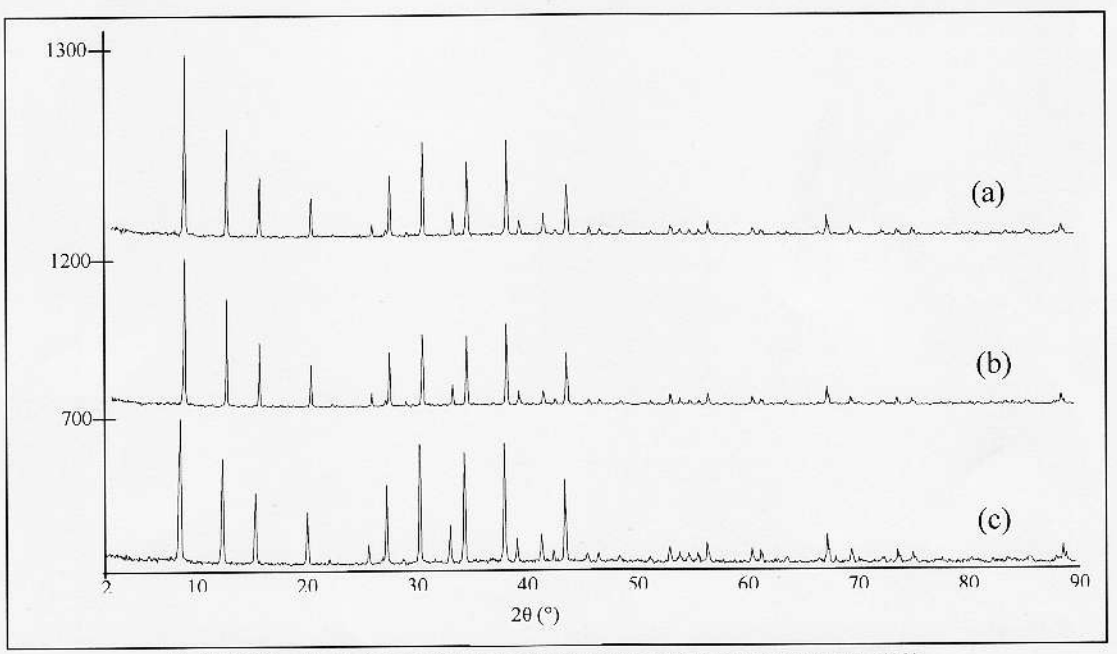
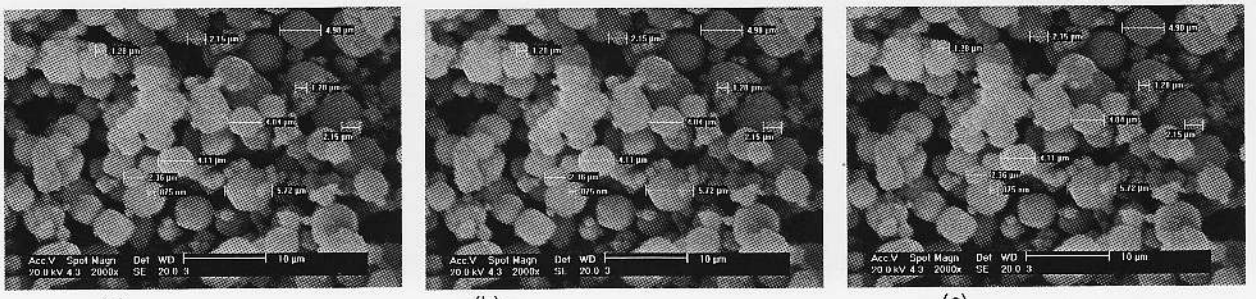


Fig3: XRD pattern for zeolite A synthesized at 80°C for a) 1h; b) 3h; c) 2h



(a) (b) (c)

Fig4: SEM analysis of zeolite A synthesized by ultrasound irradiation at 80°C for a) 1h; b) 3h; c) 2h

Conclusion

Zeolite A from colloidal silica has been synthesized for the first time by ultrasound irradiation with a significant shortening in the synthesis duration compared to hydrothermal technique. This has been proved by XRD analysis where the typical diffraction peak of zeolite A obtained. The intensity of zeolite A synthesized by ultrasound irradiation increased when the temperature is increased compared to hydrothermal technique. SEM analysis did not show any big variation of particle size when synthesized of zeolite A in different temperature and time. It is proved that the optimum condition for synthesized zeolite A under ultrasound irradiation is at 80°C and 1h reaction time. As a conclusion, ultrasound irradiation has a potential to use in synthesized zeolite A from colloidal silica.

Acknowledgements

This study was supported by Ibnu Sina Institute for Fundamental Science Studies, Universiti Teknologi Malaysia and Chemical Reaction Engineering Laboratory, Faculty of Chemical and Natural Resources Engineering, Universiti Teknologi Malaysia.

References

Andac, O., Tather, M., Sirkecioglu, A., Ece, I. and Erdeem-Senatalar, A. **2006**. *Microporous and Mesoporous Materials*. 79: 225-233.

Jansen, K., Dawyer, J. and Cundry, C. **2001**. Studies In Surface Science and Catalysis. *Fuel,* 105.

Roolmann, L., Schlenker, L., Lawton S., Kennedy, C. and Doranj D. **1999**. *Physc Chem.* 104: (721-725).

Sathupunya, M., Gulari E. and Wongkasemjit, S. **2003**. *Journal of The European Ceramic Society*. 23: 1293-1303.