Caspian J. Chem. 2(2013) 1-7

Caspian Journal of Chemistry

http://caschemistry.journals.umz.ac.ir

Using Taguchi Experimental Design to Develop an Optimized Synthesis Procedure of Sodalite Prepared by Microwave and Ultrasonic Assisted Aging

Seyed Naser Azizi*, Abdolrauf Samadi-Maybodi, Mohammad Hossein Fatemi, Neda Asemi

Analytical division, Faculty of Chemistry, University of Mazandaran, Babolsar, Po Box: 47 416–95447, Iran

* Corresponding author phone and fax number: *+981125342393*, +98 1125342350.

E-mail address: azizi@umz.ac.ir

Received 7 April 2013|Received in revised form 10 June 2013|Accepted 12 July 2013

Abstract: Perlite or pearl stone is a natural glass which has formed by rapid cooling of viscous lava or magma. Perlite is mainly composed of silica, aluminum, potassium and sodium. In this experiment, perlite was used as a low-cost source of Si and Al to synthesis of sodalite zeolite using the alkaline hydrothermal treatment. A statistical Taguchi design of experiments was employed to evaluate the effects of the process variables such as type of aging, aging time and hydrothermal crystallization time on the crystallnity of synthesized sodalite zeolite. The statistical analysis of the experimental results using Taguchi method show that the optimum conditions for maximum crystallinity of sodalite zeolite were obtained as microwave-assisted aging, 60 min aging time and 8 h hydrothermal crystallization time. The synthetic samples were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM) techniques. The results showed that the zeolite formation time was drastically reduced in the case of synthesis of sodalite zeolite using microwave assisted aging.

Key words: perlite, sodalite, Taguchi experimental design, microwave technique

1. Introduction

Perlite is a rhyolitic glass making up of more than 70% of silica and 13% of alumina occurring in various types and forms [1]. After heating, perlite becomes a light weight material and is typically light grey to white in color and is known as ‘‘expanded’’ perlite [2]. Perlite of different types (raw and expanded) and origins have various properties because of differences in composition. Among the leading countries producing perlite, Iran is estimated to be one of the largest producers of raw and expanded perlite [3]. Perlite is commercially low in price due to its abundant utilization such as ceiling tile, pipe insulation, gypsum wallboard, cryogenic insulation and filter media, etc [4]. The conversion of perlite to zeolitic materials (crystalline aluminosilicates) occurring naturally under humid atmosphere and low to medium temperatures (75–250 °C) has been reported and hence revealed a novel way in enhancing the commercial value of the mineral [5-7].

The synthesis of zeolites from low-cost silica and alumina sources has been the aim of many experiments [8]. Several groups of researchers have prepared zeolites from perlite in laboratories such as zeolite-X, gismondine, heulandite, ZSM-5 and etc [9], but to the best of our knowledge, so far there have been no reports on the synthesis of zeolite from perlite using ultrasonic and microwave-assisted aging techniques and study of effective parameters on the crystallinity of synthesized sodalite using Taguchi design of experiment.

Conventional experimental procedures involve altering of one factor at a time keeping all other factors constant, resulting in assessing the impact of those particular factors, these are time consuming, require more experimental sets and are unable to provide mutual interacting information of the factors. Design of experiments (DOE) is one approach, which helps to gain information about the optimized levels, by taking large number of variables [10]. The Taguchi method for optimization offers a good design of experiments concerned only with the main effects of the parameters. In principle, it is used for getting information from a minimum number of experiments such as the main and interaction effects of the design parameters.

The objectives of the Taguchi method for parameter design were to find out the best combination of design parameters and to reduce the variation for quality [11, 12]. Conventional heating is a routine and time consuming method for zeolite synthesis. Ultrasonic and microwave assisted (UA and MA) techniques significantly reduce the synthesis time from several days to several hours compared to conventional hydrothermal synthesis techniques [13-15]. The most important benefit of combining hydrothermal crystallization with the microwave heating technique is that it can promote faster crystallization, smaller crystal size and shorten the synthesis time [16-19].

On the other hand, employing Ultrasound prior to hydrothermal crystallization can increase dissolution processes, chemical reactions, and growth of precipitates and change the rate of synthetic process [20, 21]. Therefore, Taguchi design of experiments was used to investigate the effective parameters on the crystallinity of synthesized sodalite obtained from Iranian perlite using UA and MA aging and the other characteristics of as-synthetized zeolites sodalite was compared with each other using XRD and FE-SEM techniques.

2. Experimental

2.1. Synthesis of sodalite zeolite

The starting material used was raw perlite with a grain size of 40 µm (79.79 wt.% SiO_2; 10.66 wt.% Al_2O_3). The starting material (perlite) reacted with KOH 5 M and with the ratio of solid to liquid 1:40 (w/v) and the temperature was used at 100 ^oC at various times [4], in a 80 ml stainless steel reactor. The chosen effective factor for Taguchi analysis was as type of aging techniques (microwave, ultrasonic assisted and without aging), aging time (20, 40 and 60 min) and hydrothermal crystallization time (24, 12 and 8 h) while hydrothermal crystallization temperature of 100 ºC and the ratio of perlite to KOH (5 M) solution 1:40 w/v were kept as constant input parameters. The solid products were characterized by XRD and FE-SEM.

Table 1. Parameters and their values corresponding to their levels to be studied in crystallinity of synthesized zeolites

Factors	**Levels**		
	Level 1	**Level 2**	**Level 3**
***X*₁:Type of aging**	without aging	Ultrasonic aging	microwave aging
X_2:Aging time (min)	20	40	60
X_3:Hydro. Crystal. time[a] (h)	8	12	24

[a]Hydrothermal crystallization time

Table 2. Orthogonal array (L9) of designed experiments and their results.

Run	**X_1**	**X_2**	**X_3**	**Y_1 (%)**	**Y_2 (%)**	**S/N**
1	1	1	1	56	58	35.113
2	1	2	2	58	59	35.342
3	1	3	3	61	63	35.844
4	2	1	2	70	69	36.839
5	2	2	3	71	73	37.144
6	2	3	2	79	79	37.952
7	3	1	3	75	74	37.442
8	3	3	1	80	81	38.115
9	3	3	2	81	82	38.222

2.2. Employing Taguchi method

The process parameters, as well as their levels, are given in Table 1 and experimental data was statistically analyzed using Qualiteck-4 software. The orthogonal array of L9 type was used, and is represented in Table 2. The values of X in Table 2 indicate the levels of a factor. Taguchi recommends analyzing the mean response for each run in inner array and analyzing the variation using an appropriately chosen signal-to-noise ratio (S/N) can be calculated by formula:

$$\frac{S}{N} = -10\log\log\Sigma\frac{\frac{1}{y_i^2}}{n}$$

Where Y_i is the characteristic property (percent of crystallinity) and n is the replication number of the experiment. The S/N ratios are different according to the type of characteristics. "The bigger, the better" response is considered with the aim to maximize crystallinity (%). Crystallinites were calculated by comparing the sums of intensities of the XRD characteristic peaks appeared at 14.16° and 31.99° of 2θ in the synthesized sodalite zeolite to those found in fully crystalline reference sodalite zeolite which is hydrothermally synthesized for 48 h at 100ºC using Iranian perlite (was not shown).

3. Results and discussion

Table 3 shows the raw data for the average value of the S/N ratio for each parameter at three levels. It was shown that the parameters of aging type, aging time and hydrothermal crystallization time significantly affect the crystallinity of the synthesized zeolites. The order of importance of these factors are as follows: type of aging>aging time>hydrothermal crystallization time. Therefore, it can be concluded that the aging step plays an important role in shortening the crystallization time in synthesis of sodalite zeolite. After that, aging time is important because if UA and MA pretreatments were employed in the synthetic process for 1 h, the crystallization time could be shortened.

The response curves for the individual effects of the various parameters on the S/N ratio are given in Figure 1.

Table 3. Response (average value of *S/N* ratio) for Taguchi analysis

Factors	Level 1	Level 2	Level 3
Type of aging	35.433	37.311	37.926
Aging time (min)	36.465	36.867	37.339
Hydro. Cryst. Time[a] (h)	37.06	36.801	36.810

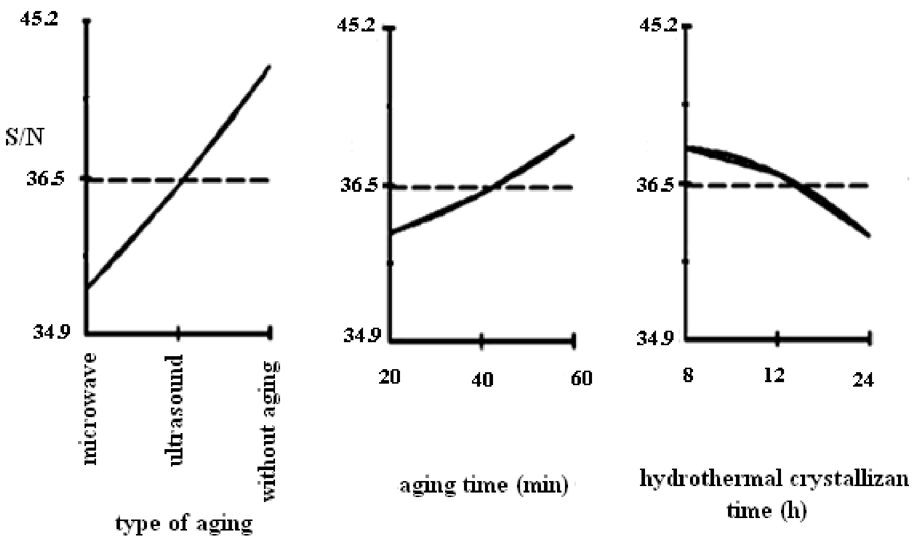

Figure 1. Effect of parameters on S/N ratio

3.1. Optimum Condition Predicted By Taguchi Method

Table 4 indicates that the first level of hydrothermal crystallization time, the third level of aging time and aging type have higher average values of crystallinity (= 82.56 %) than they have in the other levels. Furthermore, two confirmatory experiments were conducted using the optimized parameters (Table 4); the mean value of crystallinity is found to be 83.50% (Figure 2C). As it can be seen in Table 5, the crystallinity of the synthesized sodalite zeolite in the case of expected and confirmatory experiments are in good agreement with each other at optimum condition.

Table 4. Optimum conditions and their contribution

Factors	Values	Levels
Aging type	microwave	3
Aging time	60 min	3
Hydro. Cryst. time	8h	1
Expected result at optimum condition=83.23%		
Confirmatory results= 81.56%		

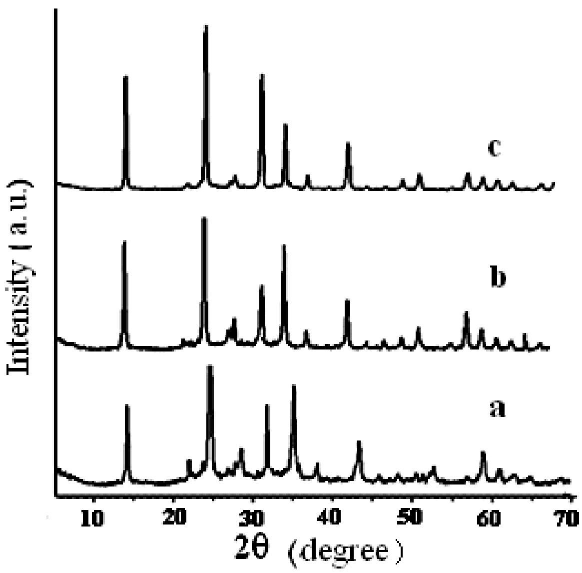

Figure 2. X-ray diffraction of synthesized sodalite zeolite (a) using 24 h crystallization time without any aging, (b) 12 h crystallization time with ultrasonic assisted and (c) 8 h crystallization time with microwave assistedaging techniques (optimum condition).

3.2. Comparision of XRD and FE-SEM Images of Synthesized Zeolite

The XRD patterns of synthesized zeolites are matched to the sodalite zeolite [9]. The XRD patterns of synthesized zeolite which were proposed using Taguchi design are shown in Figure 2. From the obtained results, it can be concluded that the aging step plays an important role in shortening the crystallization time in synthesis of sodalite zeolite. If no aging of the solution in the synthetic process was performed, crystallization time of 24 h would be required to obtain highly crystalline zeolite (Figure 2a and Table 2; run=3). In the cases of ultrasound-assisted and microwave-assisted aging for 1 h prior to heat treatment, the crystallization time could be efficiently shortened and reduced to 12 h (Figure 2b and Table 2; run=6) and 8 h (Figure 2c; Table 4; run=8). Also, the study of FE-SEM images of synthesized zeolite indicate that the morphology of synthesized zeolite has been changed when the aging techniques was used (Figure 3). The FE-SEM image of microwave assisted sample clearly indicates that the particle size of crystalline sodalite is ultrafine and within a range of 30-180 nm.

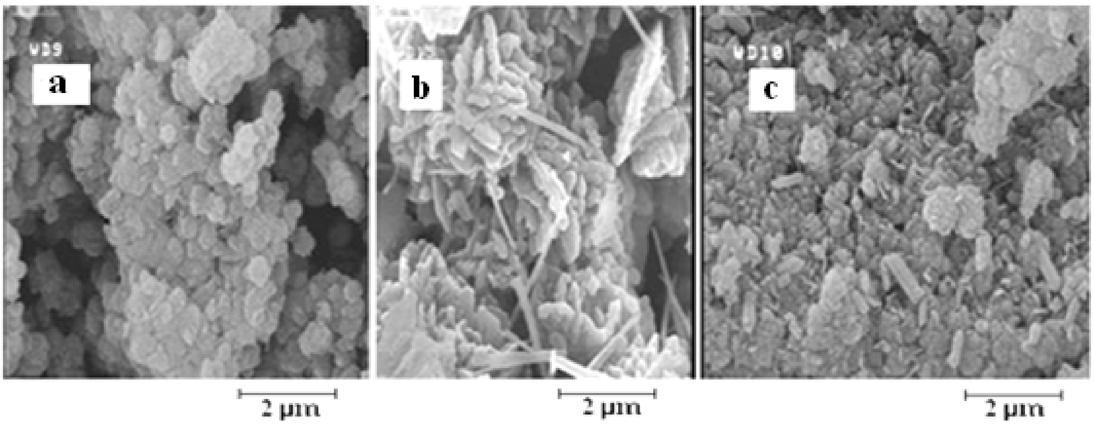

Figure 3. SEM images of synthesized sodalite zeolite (a) using 24 h crystallization time without any aging, (b) 12 h crystallization time with ultrasonic assisted (c) and 8 h crystallization time with microwave assisted aging techniques (optimum condition)

4. Conclusion

In the synthesis experiments with KOH solution of perlite, sodalite zeolite were synthesized. The XRD patterns of synthesized zeolites which were proposed using Taguchi experimental design show that the aging step plays an important role in shortening the crystallization time in synthesise of sodalite zeolite. If no aging of the solution in the synthetic process was performed, crystallization time 24 h would be required to obtain highly crystalline zeolite (Figure 1a). In the cases of ultrasound-assisted aging for 1 h and microwave-assisted aging for 1 h prior to heat treatment, the crystallization time could be efficiently shortened and reduced to 12 and 8 h (Fig 1b and 1c) and the study of FE-SEM images of synthesized zeolite indicates that microwave assisted aging led to the aggregation of smaller crystal size, probably due to the creation of more nuclei by this aging techniques (Figure 2). By Taguchi design of experiments we have been able to experimentally improve the crystallinity of sodalite zeolite, which requires a long crystallization period under the conventional method without any pretreatment. Microwave aging technique enhances the rate of nucleation and increase the nucleation rate during the crystallization process. The optimum conditions for maximum crystallinity of sodalite zeolite were obtained as microwave-assisted aging, 1h aging time and 8 h hydrothermal crystallization time from statistical analysis of the experimental results using Taguchi design.

5. References

[1] Department of Minerals Resources, Interesting Data. http: //www. dmr. go. th/ Interest/ Data/ TI2 peri D.htm. Accessed 21 October 2002.

[2] D. Shackley, M. J. Allen, International Institution of Mining and Metallurgy Bullten,1008 (1992) 13.

[3] H. Talebali, A. Farzinpour, International Journal of Poultry Science, 5 (2006) 432.

[4] W. P. Bolen, Amer. Ceram. Soc. Bull., 73 (1994) 117.

[5] J. W. Noh, J. R. Boles, Clay Clay Miner., 37 (1989) 47.

[6] A. Rujiwatra, Mater. Lett., 58 (2004) 2012.

[7] S. Khodabandeh, M. E. Davis, Micropor. Mesopor. Mat., 9 (1997) 161.

[8] P. Kongkachuichay, P.Lohsoontorn, Science Asia., 32 (2006) 13.

[9] S.Tangkawanit, K. Rangsriwatananon, Suranaree J. Sci. Technol., 12 (2005) 61.

[10] Taguchi, G., System of Experimental Design. UNIPUB Kraus, International Publications, New York, 1987.

[11] S. T. Kim, H. M. Park, Sens. Actuators B., 102 (2004) 253.

[12] C.B. Raj, C. H. L. Quen, Chem. Eng. Sci., 60 (2005) 5305.

[13] P. Wang, B. Shen, D. Shen, T. Peng, J. Gao, Catal. Commun., 8 (2007) 1452.

[14] Y. Hu, , C. Liu, Y. Zhang, N. Ren, Y. Tang, Micropor. Mesopor. Mat., 119 (2009) 306.

[15] W. C. Conner, G. Tompsett, K. H. Lee, K. S. Yngvesson, J. Phys. Chem. B., 108 (2004) 13913.

[16] M. Abrishamkar, S. N. Azizi, H. Kazemian, Chem. Met. Alloys, 3 (2010) 12.

[17] P. Chu, F. G. Dwyer, J. C. Vartuli, (Mobil Oil Corporation). US Patent 4778666, 1988.

[18] A. Arafat, J. C. Jansen, A. R. Ebaid, H. Van bekkum, Zeolites., 13 (1993) 162.

[19] C. S. Cundy, R. J. Plaisted, J. P. Zhao, Chem. Comm., (1998) 1465.

[20] C. G. Wu, T. Bein, J. Chem. Soc. Chem. Comm., 8 (1996) 925.

[21]Y. S. Li, Q. G. Yin, Ultrasonic Sonochemistry, Science Press, Beijing, 1995.