Jurnal Teknologi

Full paper

The Characteristic of Synthesized Zeolite Rice Husk Particles via Different Routes

Nurafiqah Rosman*, Zawati Harun, Mohd Riduan Jamalludin

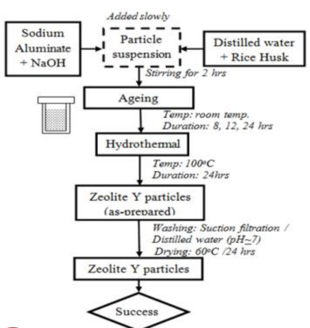
Advanced Material and Manufacturing Centre, Faculty of Mechanical Engineering, Universiti Tun Hussein Onn Malaysia, Batu Pahat 86400, Johor, Malaysia

*Corresponding author: afiqahrosman@gmail.com

Article history

Received :15 September 2013
Received in revised form :
17 November 2013
Accepted :15 January 2014

Graphical abstract



Abstract

Zeolite particles were synthesized through in-situ extraction of silica from rice husk ash without seeding method in the absence of organic template by a static hydrothermal condition at 100ºC/24h at varied ageing 8-24h. The effect of amorphous silica content on ageing time of synthesis products was evaluated with X-ray diffraction (XRD), Fourier transform infrared (FTIR) and field emission microscopy (FESEM). The present of zeolite Y without seeding method by XRD analysis show a mixture of zeolite A, Y and P. The transformation of amorphous rice husk particle gradually increased as early at 8 hours in ageing and increased with increase in ageing reaction. The vibration bands at around 974 cm^{-1} indicated Si-OH vibrations of the surface silanols, which is the characteristic of mesoporous silica. The FESEM images showed cubic-shaped morphology of the powders increased with increase in ageing reaction.

Keywords: Rice husk ash; zeolite Y; hydrothermal; ageing duration

■1.0 INTRODUCTION

Rice husk (RH) is one of the most abundance agricultural products in Malaysia. A million tons of rice husks, which can be considered as agricultural waste, are thrown as a waste every year and usually ends up with open burning process that causes environmental pollution and disposal problems. Discovering and recovering high value materials from RHs is becoming a potential research to be explored since burning and decomposition of RH will produce a value silica compound. In fact, due to growing environmental concern, and the need to conserve energy and resources, effort have been made to utilize the resultant ash as for a building proposes, semiconductor industry and additive in membrane applications. [1-3] Basically, RHA is an active catalyst and a good material for catalyst support due to high surface area [4] and can be used as an alternative cheap source of amourphous silica for zeolite preparation. The extensive usage of RH in the preparation zeolites for separation processes such as sorbent, as well petrochemical processes is widely explored and studied. [5]

In general, zeolites are hydrated crystalline aluminosilicates composed of tetrahedral TO_4 units (T=Si or Al) which are linked together by sharing oxygen atom to form regular intracrystalline cavities and channels of aromic dimensions.[6] The zeolites are conventionally prepared by the hydrothermal method of the gel containing silica, alumina, cation, template and water. [7] However, the silica element can be modified and replaced with different silica sources. Due to excellent features by RHA, many researchers have established different method and routes to synthesis different zeolites.[8,9] By use RHA as silica source, Khemthong *et al*.[10] able to synthesized Zeolite LSX by hydrothermal process, while Katsuki *et al*.[11] obtained ZSM-5 by conventional- and microwave-hydrothermal method. Yet, Zeolite beta was also obtained from rice husk ash by hydrothermal process.[7]

Theoretically, the formation mechanism of zeolite under hydrothermal condition is very complex.[12] It involves several important parameters such as agitation time, agitation temperature, ageing time, ageing temperature, crystallization time and crystallization temperature on the zeolite preparation.[13] However, the optimization and discussion regarding to ageing time is rarely emphasized in synthesis process. Khabuanchalad *et al*.[14] reported, ageing time did not have a great effect on the procedure of zeolite synthesis. In contrast with Al-Zaid [15] reported a deformation in of Na-Y is obviously shown with the increased of ageing period. As reported by him, the intensities of peak is obviously seen and the effect of ageing becomes an interest in the mechanism of crystal growth and the results show that the process expected to perform better when gel for a zeolite hydrothermal synthesis is proceed rapidly. In this present work, static hydrothermal method is applied for the synthesis of Zeolite Y powders. The intermediate reaction steps for the extraction of silica from RHA could be omitted to provide a simple and easy steps, followed by the investigation of aging effect.

■2.0 EXPERIMENTAL

2.1 Materials

Rice husk ash, pellets of sodium hydroxide (NaOH) (Emory), Sodium aluminate ($NaAl_2O_3$) (Riedel-de Haën), and distilled water were used as the starting materials in the initial mixture for the synthesis of zeolite Y. The silica source used for the experiments was a rice husk obtained from Malaysia rice mill (Jelapang Selatan). The received rice husk was burning at 700°C for 5h in atmosphere, to obtain the silica contain as well as eliminating the amount of carbon. Figure 1 shows schematically the preparation of zeolite Y powders. In the hydrothermal synthesis, the sodium aluminate solution was prepared by dissolving sodium aluminate powder (1.888g) with NaOH solution (2.361g in 26mL water). Silicate solution prepared by adding RHA (0.18g) into 270 mL water and left for 2hr in stirred condition. The progressive addition of silcate solution and sodium aluminate was carried out with stirring using magnetic stirrer until they dissolved, to homogenize the reaction gel for 2 hours. The molar composition of the reaction gel was $3.4Na_2O$: Al_2O_3: 9.5 SiO_2: $136H_2O$.[16] The reaction gel was poured into Teflon-lined stainless steel autoclaves and left for ageing process in 8,12 and 24 hour. Then, it was heated at 100°C for 24 hour at autogenic pressure. After hydrothermal reaction, the as-prepared powder were collected by centrifugation (5000rpm) followed by washing with distilled water repeatedly until the washing liquid become almost neutral as observed by the pH meter. The powders were dried at 60°C for 24 hours.

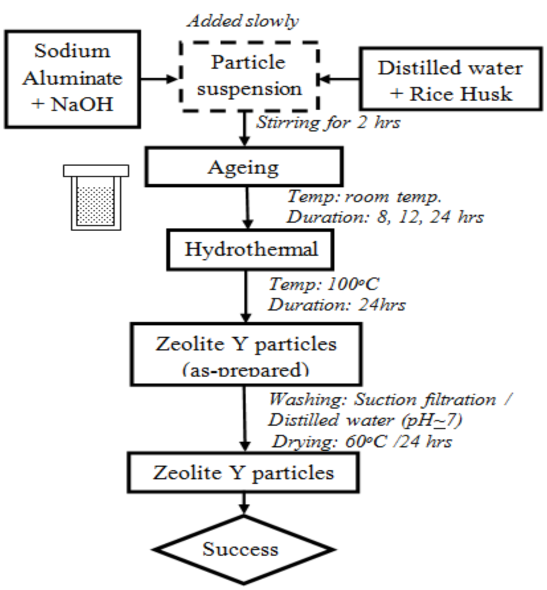


Figure 1 A schematic representation for the synthesis of ZeoliteY powders using rice husk ash as silica source

2.2 Characterizations

The crystal phases of the powder were identified using powder diffraction technique by Bruker XRD (Model: D8 Advance) with Ni-filtered Cu-K_a radiation (λ=0.15418nm), operating at 40kV and 40mA. The morphology of the synthesized particles was examined by FESEM (Model: JEOL JSM 7600F) operating with an accelerating voltage of 1kV. The characteristic vibration bands of the particles were confirmed by FTIR (Perkin Elmer) at resolution of 4cm^{-1}.

■3.0 RESULTS AND DISCUSSION

3.1 Characterization of Zeolite Y by XRD.

The effect of ageing time in the formation of zeolite Zeolite-Y on the type and composition of zeolite samples were investigated. The XRD diffractograms were presented in Figure 2. which indicates the characteristic peaks of powders obtained hydrothermally at 100°C/24hr at different ageing duration.

The XRD patterns show that transformation of silica phase to crystalline phase of zeolite occurred as early 8 hours in ageing reaction. The increased of ageing time will result in increasing of intensity peak as displayed in Figure 2 which revealed the reduction of zeolite crystallization process.[17] In fact, the synthesis of zeolite Y without seeding method in this present work produce a mixture of zeolite A, Y and P. Analysis of zeolite Y formation was referred to JCPDS number 38-0240 data by matching the diffractograms pattern of all synthesized samples. Due to the fact that zeolite Y is metastable, some of the crystallites were transformed to zeolite P with JCPDS number 39-0219 as clearly can be seen in Figure 2.[18] In fact, the result also shows the existence of Zeolite type A with JCPDS number 40-1646. According to Turnbull[19] zeolite A, Y and P have the structure of faujasite where all of these type are naturally exist as mineral, and are commonly denoted as FAU. All these three types of zeolite are formed of solidate cages. Besides, the small zig-zag peaks refer to presence of some amorphous material.[20]

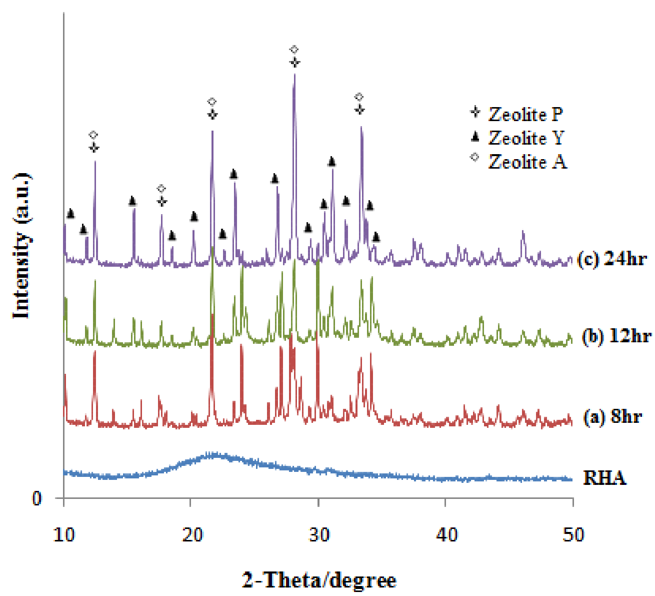


Figure 2 XRD patterns of Zeolite-Y powders synthesized at 100°C at varied ageing duration for (a) 8, (b) 12, and (c) 24 h

3.2 Characterization of Zeolite Y by FTIR.

Figure 3 shows the FTIR spectra of the powders prepared at 100°C for 24 hr at various ageing duration (a) 8, (b) 12, and (c) 24 h. All the plots show a similar pattern and characteristic of absorption bands but with slightly different in formation peak. Theoretically, the characteristic of absorption bands for zeolite are around 3351, 1635, 972, 667 and 576 cm^{-1}. The peak at 576 cm^{-1} is attributed to the double ring of external linkage peak which assigned zeolite Y [21, 22] and peak at 667cm^{-1} is assigned to carbon dioxide. [23] Meanwhile, the peak at 972 cm^{-1} is assigned to Si-OH vibrations due to the effect of surface silanols, which is the characteristics of mesoporous silica [24] and peak around 3351cm^{-1} is given to hydroxyl groups of zeolite. The existence of peak at 1635cm^{-1} indicates chemisorbed pyridine (Py) characteristic that show the dealumination of the sodium form on zeolite.[25] As obviously shown in the FTIR spectra pattern, the complete pattern of zeolite was slowly established with the increased of ageing time.

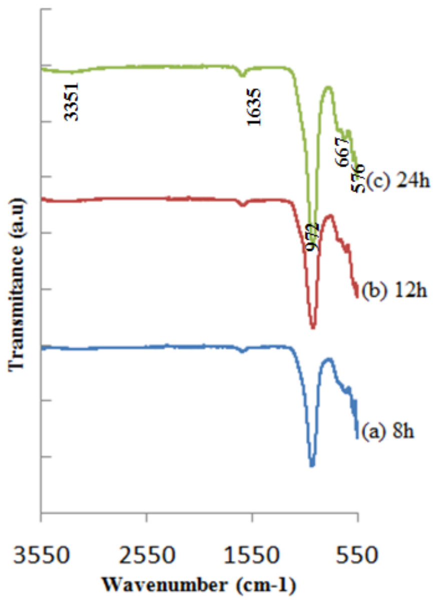
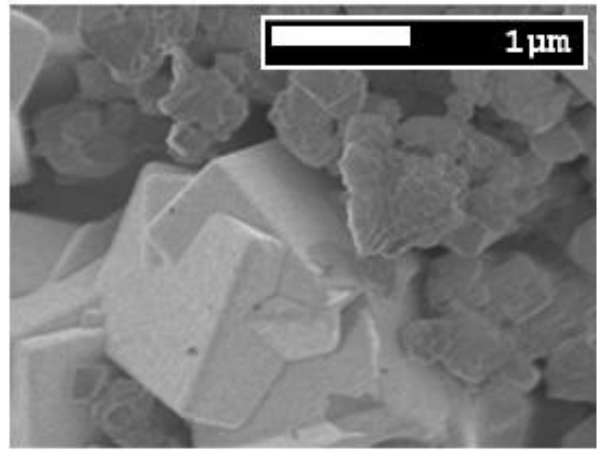


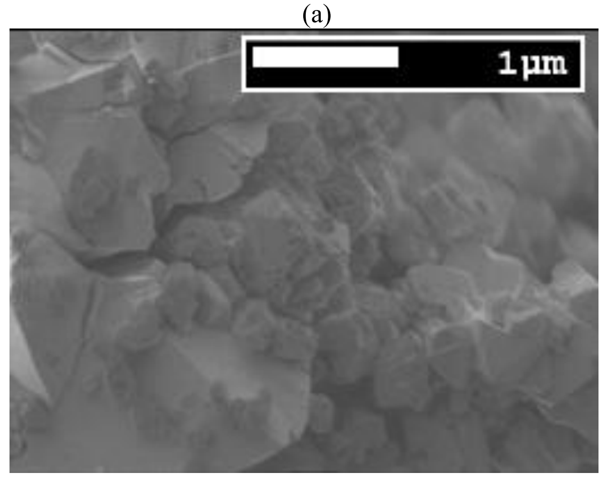
Figure 3 FTIR spectra of Zeolite Y powders synthesized at 100°C at varied ageing duration for (a) 8, (b) 12, and (c) 24 h

3.3 Characterization of ZeoliteY by FESEM

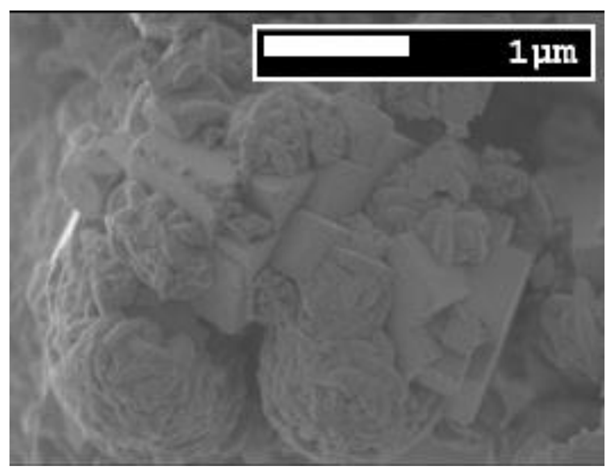
Figure 4 shows the FESEM microstructure of Zeolite Y particles obtained at 100°C/24 hr at varied ageing duration (a) 8, (b) 12 and (c) 24 h with magnification of 10,000. It reveals characteristic of cubic shape particles of ZeoliteY and it was noticed that the particles were agglomerated in crystal.



(a)



(b)



(c)

Figure 4 FESEM-image of Zeolite Y powders synthesized using rice husk ash as a silica source without seeding method at varied ageing duration (a) 24, (b) 12 and (c) 8 h

The cubic-shape Zeolite Y powder is clearly seen with increase in ageing duration due to the reaction between RHA and sodium aluminate. Al Zaidi[15] reported that the deformation of zeolite crystals increased with increasing of ageing period. The FESEM morphology in Figure 2(c) shows the cubic shape of Zeolite Y covered with amorphous silica which can be clearly seen compared to others. The effect of reducing ageing duration delays the transformation of crystal zeolite.

4.0 CONCLUSION

The zeolite-Y powder was synthesized by static hydrothermal condition by using of rice husk ash as silica source at 100ºC/24 hr. The transformation of metastable zeolite crystalline phase from amorphous RHA was observed as early 8 hours in ageing reaction at room temperature. Microstructure observation show the formation of Zeolite Y in increase of ageing time increase the solid formation of crystalline structure. FTIR results obviously peak at around 974 cm^{-1} indicated Si-OH vibrations of the surface silanols, which is the characteristic of mesoporous silica. As proved with FESEM analysis, the image of cubic shape growth is clearly seen the figure. Preparations of zeolite Y without seeding method also give a mixture of zeolite A and P.

Acknowledgement

The author would like to thank to Ministry of Higher Education Malaysia, Post Graduate Incentive Research Grant (GIPS) and Universiti Tun Hussein Onn Malaysia for their Long Term REsearch Grant Scheme (LRGS vot A022) for support in providing the grant implement "High Performance of Polymeric Materials" project.

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