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Synthesis of zeolite-Y from rice husk silica and food grade aluminum foil using modified hydrothermal method

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Abstract. In this investigation, zeolite-Y prepared from rice husk silica (RHS) and food grade aluminum foil using modified hydrothermal method. In this proposed method, rice husk silica was dissolved in sodium hydroxide solution to obtain sodium silicate solution and then small pieces of aluminum foil were directly mixed with silicate solution, producing zeolite precursor in the form of gel. The gel was transferred into PTFE lined autoclave and aged for 24 hours and subsequently subjected to the crystallization process for 24, 48, 72, 96, and 120 hours at fixed temperature of 100°C. The solid product was washed with distilled water and dried in an oven at 80°C and calcined at 550°C for 6 hours. Characterization using XRD and SEM confirms the formation of zeolite-Y, and showed that crystallization time influenced both structure and surface morphology of the zeolites quite significantly.

Keywords: Zeolite-Y, rice husk silica, food grade aluminum foil, hydrothermal method, PTFE lined autoclave.

1. Introduction

Zeolite-Y is a versatile hollow molecule of faujasite family with three dimensional pore structure and solid acidity that makes it potential as ion exchange [1,2], adsorbent [3,4], and catalyst [5,6]. As catalyst for example, this zeolite has been used in transesterification reaction for biodiesel production [7] and biomass pyrolysis for liquid fuel production [8-10]. Due to its wide application, zeolite-Y continuously attracts interest from scientific society, with the main focus on preparation from various raw materials such as kaolin [11], rice husk ash [12], and rice husk silica [13].

Of various raw materials that have been studied, rice husk silica is of particular interest for several reasons. According to research by Harsono [14], it is known that the content of rice husk consists of about 20% inorganic material with silica as the main component. Rice husk silica is noted to dissolve in alkaline solutions such as sodium hydroxide and potassium hydroxide solutions so that it can be extracted easily using alkaline extraction methods [15] and produce silica with the purity in the range of 85-97%. Rice husk silica is also recognized as amorphous and reactive material that makes it suitable as raw material for preparation on various silica based products. These characteristics of rice husk silica are the reasons to use this material for the synthesis of zeolite-Y in this study. Another component of synthetic zeolite is alumina. Some sources of alumina commonly used in zeolite manufacturing include aluminum sulfate [16], aluminum nitrate [17], sodium alumina [18], and other materials containing aluminum. In this study, aluminum foil was used as a source of alumina since it is easily obtained at a relatively cheap cost with purity 98.35-99.2% and has high solubility in alkaline solutions.

To prepare synthetic zeolite, two methods commonly used are solvothermal [19,20] and hydrothermal process [21-23]. In principle, these two methods involve similar process, with the only difference is the type of solvent used. In the hydrothermal method, water is used as a solvent while in the solvothermal method, non-aqueous substances such as alcohol, inorganic, and organic are used. The hydrothermal process is considered more practical as it uses water as a solvent. In general, most synthetic zeolites are also synthesized using the hydrothermal method because it produces zeolites with a high level of purity and good thermal properties.

In previous studies [11,24] preparation of zeolite-Y from rice husk ash was reported to involve rather complicated process, in which the procedure was started by producing seed gel and feedstock gel and then the two gels were mixed to produce overall gel. This final gel was then aged for 24 hours, followed by crystallization process for 24 hours at 110°C and finally, the product was filtered, washed thoroughly with distilled water, and dried at 110°C in the oven.

In this study, a simpler procedure for synthesis of zeolite-Y was proposed. The procedure was initiated by preparing sodium hydroxide solution. Then, rice husk silica was dissolved in the sodium hydroxide solution to produce sodium silicate solution. Next, the food grade aluminum foil that has been cut into small pieces was added to the sodium silicate solution and stirred for 3 hours to form a homogeneous gel. The gel was transferred into PTFE lined autoclave and then aged for 24 hours to produce crystal seed [24]. The sample was placed in the oven for crystallization process at 100°C for different times. After the completion of the crystallization process, the product was washed with distilled water until the filtrate has neutral pH and dried in the oven at 80°C for 24 hours. Finally, the sample was subjected to calcination treatment at 550°C for 6 hours. The sample was characterized using X-Ray Diffraction (XRD) to analyze the crystallography of zeolites and Scanning Electron Microscopy (SEM) to determine the surface morphology of the sample.

2. Materials and methods

2.1. Materials and Instruments

The following is a list of materials' sources that were used in this study, they are rice husk obtained from rice field in Pringsewu regency contained silica with the purity is in the range of 85-97%, reagent grade nitric acid (HNO_3) and sodium hydroxide (NaOH) pellet purchased from Sigma Aldrich, and food grade aluminum foil purchased from local supplier in Bandar Lampung city. The instruments include hotplate stirrer, laboratory glassware, oven, and Polytetrafluoroethylene (PTFE) lined stainless steel autoclave for crystallization process. The calcination of zeolite-Y was done using Nabertherm electrical furnace (Lilienthal, Germany). XRD characterization was analyzed by PANalytical type Empyrean diffractometer. Scanning electron microscope Zeiss EVO MA 10 type was performed for SEM characterization.

2.2. Rice Husk Silica Extraction

The rice husk silica was prepared through alkali extraction procedure described in our prior study [15]. Initially, 50 g dried rice husk was extracted in 500 mL of 1.5% sodium hydroxide (NaOH) solution and heated to boiling for 30 minutes. The mixture was then allowed to cool at room temperature and left for 24 hours. After that, it was filtered to separate rice husk from the filtrate that contains silica. The formation of silica gel was reached by adding the dropwise of 10% nitric acid (HNO_3) solution slowly to the filtrate until the condition was neutral (pH of 7.0-8.0). It was aged for 24 hours. After that, the gel was washed with distilled water until the color was clear. Sodium hypochlorite solution then was added to maximize the process of whitening the gel. It was left for 24 hours more and rinsed until clean and odorless. The dried rice husk silica could be obtained by drying the silica gel in oven at 80° C for 24 hours and ground it into powder.

2.3. Zeolite-Y Preparation

Zeolite-Y was synthesized using hydrothermal method with composition of $4Na_2O$: Al_2O_3: $4.8SiO_2$: $120H_2O$ prepared from a mixture of sodium silicate solution and food grade aluminum foil. The zeolite-Y synthesis was started by weighing 11.11 grams of sodium hydroxide (NaOH) pellets, 1.875 grams of food grade aluminum foil, and 10 grams of rice husk silica. The sodium hydroxide (NaOH) pellets were then dissolved with 120 mL of distilled water and slowly added with rice husk silica while stirring for 4 hours in a boiling state. After all the rice husk silica was dissolved, the sodium silicate solution was filtered using filter paper to separate it from impurities that were still present in the solution. Next, the food grade aluminum foil that has been cut into small pieces was added to the sodium silicate solution by distributing it evenly and then stirred for 6 hours to form a homogeneous gel. After that, the gel was moved into PTFE lined autoclave and then aged for 24 hours in a closed state to grow the zeolite crystal core [24]. Next, the sample was placed into the oven to undergo the crystallization process at 100°C for 24, 48, 72, 96, and 120 hours. After the process was finished, the sample was gently filtered with filter paper to separate the zeolite from the filtrate in the autoclave and rinsed with distilled water until the filtrate was neutral (pH 8). Finally, zeolite-Y was dried in an oven at 80°C for 24 hours and calcined at 550°C for 6 hours.

2.4. Zolite-Y Characterization

PANalytical type Empyrean diffractometer with CuKα (λ = 1.54 Å) radiation and the energy of 40 kV and current of 100 mA was performed to get the XRD pattern for phase identification. The pattern was analyzed over the 2θ range of 5–60°. Match! Version 2.4.7 was used to identify the phases formed further and confirmed by comparing the pattern with the standard data of the International Zeolite Association (IZA). The surface morphology of zeolite-Y was verified by SEM ZEISS EVO MA 10. Using an electron acceleration voltage of 20 kV, the instrument was controlled at 30 KV. Zeolite-Y then was scanned at different magnifications to acquire micrographs with better image of the surface.

3. Results and discussion

3.1. Characterization using XRD

Zeolite-Y synthesized at different crystallization times, 24, 48, 72, 96, and 120 hours were characterized using X-Ray Diffraction (XRD). The diffraction data provides information about the structure and phase based on the angle of 2θ (in degrees) and can be used to study the effect of crystallization time on zeolite formation. To identify the phases formed, the diffractogram of each sample was then analyzed further using the Match! Software Version 2.4.7, and were confirmed with reference diffractograms established by the International Zeolite Association (IZA) with the basic principle that there are at least three major peaks of the sample diffractogram overlapping with the corresponding reference peaks. The result of zeolite-Y diffractograms crystallized at different times is shown in Figure 1.

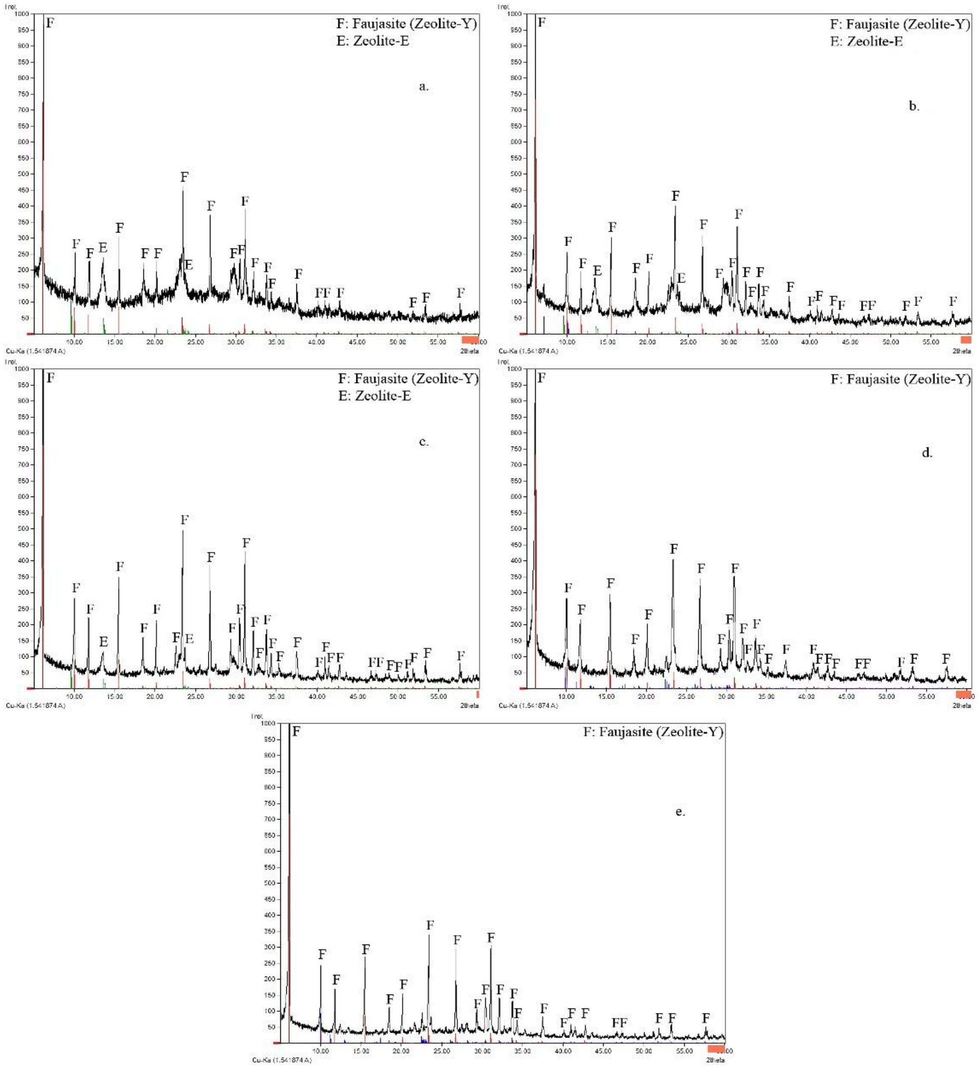

Figure 1. XRD diffractograms of zeolite-Y with different crystallization times: (a) 24 h; (b) 48 h; (c) 72 h; (d) 96 h; and (e) 120 h.

The pattern presented in Figure 1 shows that all samples are dominated by the crystalline phases which are marked by sharp peaks in the range of 2θ, although there are still a few amorphous phases formed. The diffractograms above clearly illustrate the reaction between sodium silicate and food grade aluminum foil has occurred during the crystallization process to produce the crystalline phase. With the aid of Match software! Version 2.4.7, it can be seen that all samples are dominated by faujasite phase which is the primary phase forming the zeolite-Y. As for the second phase formed, zeolite-Y crystallized at 24, 48, and 72 hours show the results of the presence of zeolite-E. However, the second phase is a minority phase with very low intensity so that the zeolite-Y resulting from 24 to 72 hours crystallization period time obviously still meet the reference standards set by the International Zeolite Association (IZA), even the formation can take place in the crystallization time of 24 hours. While the second phase is not found in zeolite-Y with the crystallization time of 96 and 120 hours. These results suggest that in the crystallization process for 96 hours, the zeolite-E has been completely converted into zeolite-Y. As shown in the diffractograms that the longer the crystallization time, the lower the intensity of zeolite-E. Therefore, from the crystallization time of 96 hours, zeolite-Y practically has formed a single phase (*monophase*).

Further evaluation of the XRD data was carried out by comparing experimental data with the International Zeolite Association (IZA) data. The results are presented in Figure 2 (a-e), while the 2θ data and the intensity values of the main peaks are emphasized in Table 1.

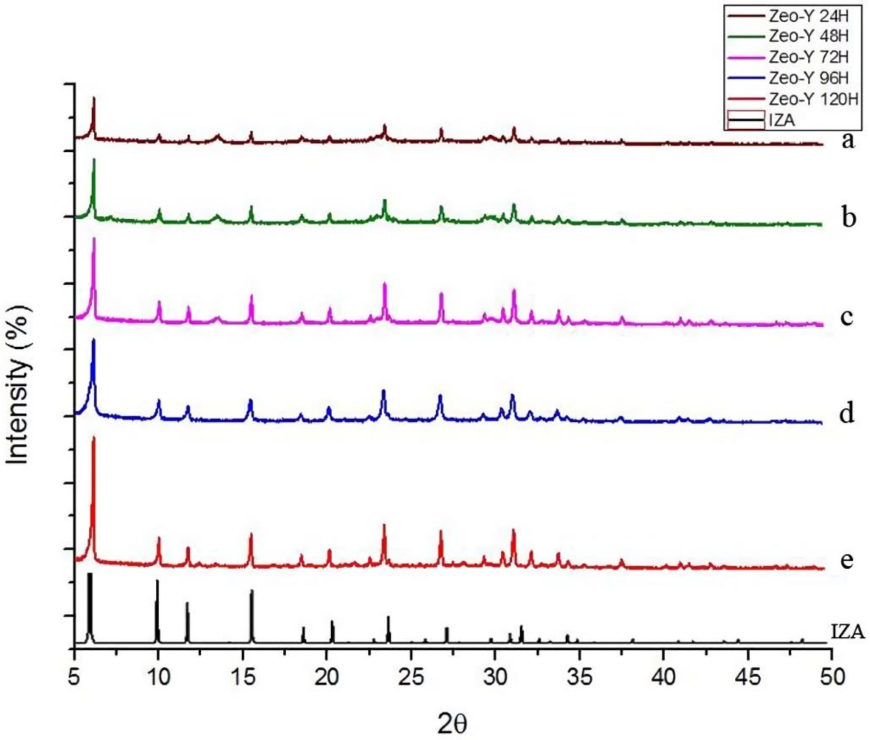

Figure 2. XRD patterns of IZA standard; (a) zeolite-Y 24 h; (b) zeolite-Y 48 h; (c) zeolite-Y 72 h; (d) zeolite-Y 96 h, and (e) zeolite-Y 120 h.

The data in Table 1 show that there are 6 peaks which are the main characteristics of zeolite-Y according to IZA standard found in all synthesized samples. Even though the relative intensities in the samples are not the same, the highest intensity of 100% in all samples is obtained in the 2θ range of 6° which complies with the IZA standard.

Table 1. XRD data comparison of zeolite-Y IZA, zeolite-Y 24 h, zeolite-Y 48 h, zeolite-Y 72 h, zeolite-Y 96 h, and zeolite-Y 120 h.

IZA		Sample with various crystallization times									
		24 hours		48 hours		72 hours		96 hours		120 hours	
2θ (°)	Intensity (%)	2θ (°)	Intensity (%)	2θ (°)	Intensity (%)	2θ (°)	Intensity (%)	2θ (°)	Intensity (%)	2θ (°)	Intensity (%)
6.183	100	6.149	100.00	6.155	100.00	6.152	100.00	6.137	100.00	6.131	100.00
10.104	9.09	10.049	20.37	10.045	23.96	10.039	15.11	10.025	23.72	10.026	21.62
11.854	7.34	11.783	13.46	11.786	19.09	11.780	14.48	11.765	16.17	11.760	14.59
15.600	10.46	15.502	25.66	15.506	32.08	15.500	23.86	15.474	26.66	15.486	25.65
18.621	4.1	18.493	12.42	18.505	12.8	18.476	13.21	18.456	8.58	18.472	8.87
26.947	3.87	26.766	26.9	26.766	37.97	26.754	30.58	26.701	34.51	26.741	29.36

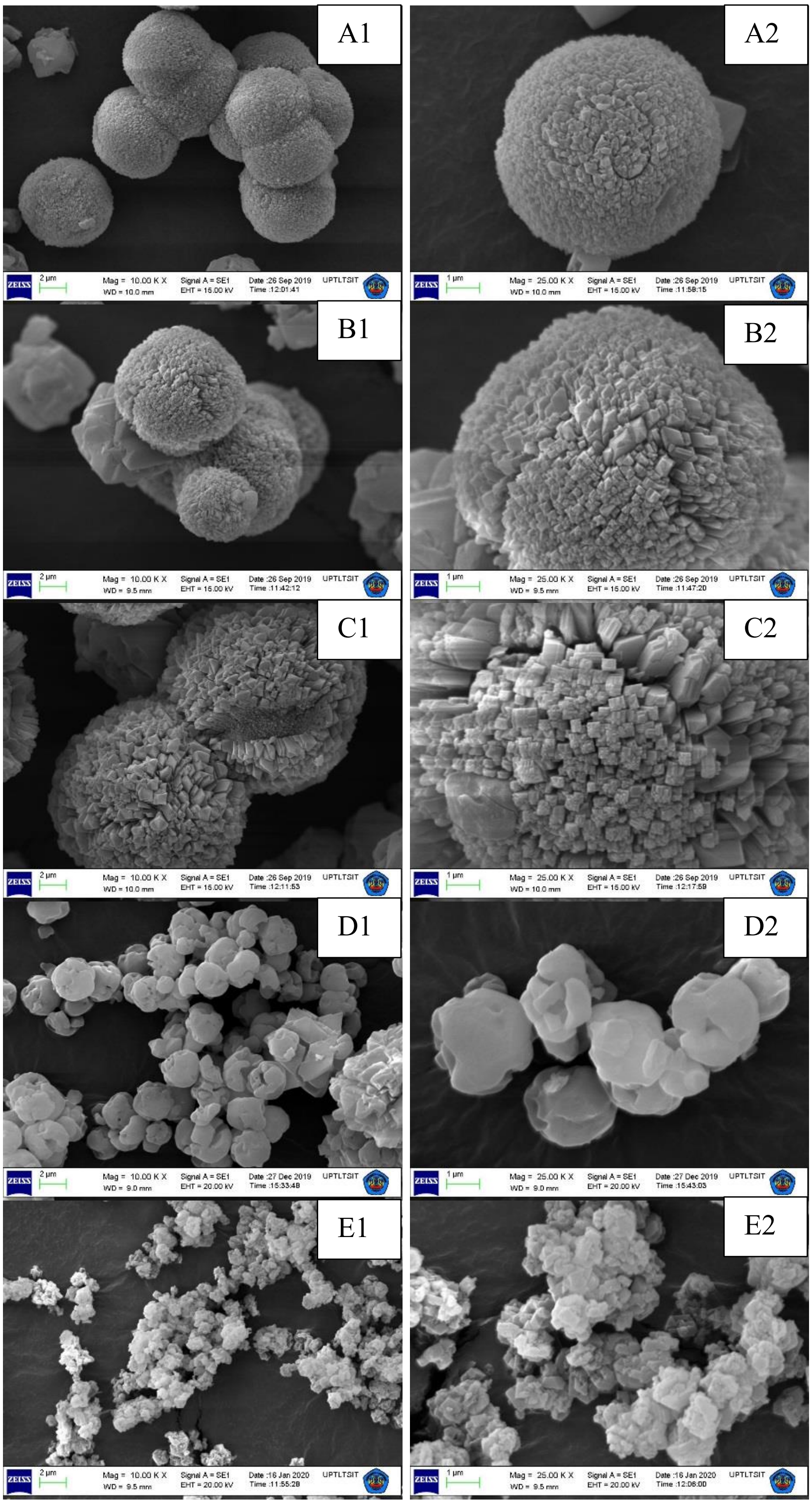

Figure 3. SEM micrograph of (a) zeolite-Y 24 h; (b) zeolite-Y 48 h; (c) zeolite-Y 72 h; (d) zeolite-Y 96 h; and (e) zeolite-Y 120 h with magnification of 10.000 and 25.000 times.

3.2. Characterization using SEM

Characterization using SEM aims to obtain information about the surface morphology, particle shape, and distribution patterns of particles on the surface. SEM analysis was carried out on all samples synthesized at different times, 24, 48, 72, 96, and 120 hours and calcined at 550°C with different magnifications, 10.000 and 25.000 times, producing SEM micrographs as presented in Figure 3. Based on Figure 3, it appears that the surface morphologies of all samples are almost similar, spherical formed. Nevertheless, if being observed further, zeolite-Y synthesized at 24, 48 and 72 hours have more similar shape characteristics and in accordance with the reference [25], that is a circle which is assembled from a collection of crystal blocks that grow sticking out from the center of the circle. However, as crystallization time increases, the three samples show that the crystal size is getting bigger so that the diameter of the formed circle has expanded. Whereas the zeolite-Y synthesized at 96 and 120 hours show a more similar shape, it is biconcave with a very dominating crystal phase, but zeolite-Y 120 hours shows more irregular shape. It is affected by the duration of crystallization time which turned out to destruct the samples' surface morphology. This fact is proven by the gradual change from the size of the crystal that enlarges to disintegration at 96 hours and increasingly irregular at 120 hours. So from the SEM results of the five samples, it can be concluded that the maximum crystallization time of zeolite-Y is 72 hours.

4. Conclusion

The results obtained in this investigation demonstrate successful synthesis of zeolite-Y from rice husk silica and food grade aluminum foil using modified hydrothermal process proposed. Characterization using XRD and SEM confirmed the formation of zeolite-Y, and at the same time also revealed a quite significant effect crystallization time on crystallite structure and surface morphology of the zeolite produced.

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