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Effect of Microwave Irradiation on the Zeolite Properties Synthesized from Rice Husk Ash

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Abstract. Indonesia is an agricultural country with the main product was rice. The abundance of rice farming contributes to the amount of rice husks every year. Today, the rice husk has been widely used as a source of silica in the preparation of synthetic zeolite in many researches. In this work, increasing characteristics of synthetic zeolite from rice husk ash by microwave irradiated method is investigated. The prepared zeolite samples were characterized using X-Ray Diffraction (XRD) to determine crystallinity and zeolite components. In application, synthetic zeolite is used in improvement of waste cooking oil quality. Waste cooking oil quality was determined by peroxide value. Modification of zeolite by microwave irradiation fixed in 100 W and 180 W variable of power for 60 minute. Both treated and untreated sample formed high crystallinity based on the XRD traces that is above 90%. In this project, the wasted cooking oil sample is taken from the rest of the frying fish. Adsorption was carried out at 60 ℃ with constant stirring in each sample. The peroxide value was analyzed by iodometric method, directly. The initial peroxide value of waste cooking oil is 9 meq/kg, this value decreased since adsorption process by prepared zeolite. Among the adsorption processes, 180 W modified zeolite yield highest decrease of peroxide value i.e 2 meq/kg, follow by 100 W modified which is 5 meq/kg, and the least decrease was reached by unmodified zeolite adsorption i.e 6 meq/kg.

1. Introduction

Paddy is a commodity with the largest amount of production in Indonesia compared to other food crops such as corn, soybeans, peanut, etc. The amount of paddy production tends to increase every year, significantly. This can be seen from the amount of rice production in 2015 that is reached 75 tons, while rice production in 2014 amounted to 70 tons [1]. The abundance of paddy in Indonesia will have a serious impact on the environmental pollution, due to rice husk from residue milling. Thus, potent technique should be applied to utilize rice husk, effectively. In the process of milling, paddies produce 20-30% of rice husk of total weight [2][3]. The result of burning rice husk as rice husk ash is a potential source of silica. Rice husk contain approximately 20-25% of silica [4][5]. Rice husk is one of the most underutilized biomass, most of the rice husk is just burned. Many studies have been conducted to increase the economic value of rice husk, so that rice husk can be used as one of renewable material sources. Hydrothermal is commonly used method used in the manufacture of synthetic zeolites. In the past year, a new technique of microwave-utilize has been widely introduced as a novel energy. By its properties, microwave is able to heat objects rapidly, quickly and effectively [6]. Several methods in the zeolite manufacturing process have been performed to produce zeolites with superior characteristics. Heat is regenerated thoroughly by rotate motion, it might cause the shorten heating time in chemical reactions [7][8]. Previous study mentioned that the synthesis process

of zeolite-A from CFA using microwave irradiation obtained shorten the crystallization time compared with conventional heating method i.e. from 8-89 hours to 10-30 minutes [9]. The microwave utilization also showed a positive response on the synthesis of zeolite Na-p1 from CFA [10]. Arafat et al [11] concluded that the process of synthesizing zeolite-Y and ZSM-5 by involving microwave result shorten time from 10-50 hours to 10 minutes on zeolite-Y and from several days to minutes (30 minutes) of zeolite ZSM-5. We have already known that amorphous silica hold great promise in promise in heavy metal stabilization [12]. In this work, the obtained both modified and unmodified zeolite were used to tread decreasing peroxide present in waste cooking oil. The effect of microwave irradiation on zeolite sorption capability will be investigate further.

2. Experimental Procedure

2.1 Sample Preparation

The initial stage in the synthesis of zeolites is initiated from the making of rice husk ash. Previously rice husk was thoroughly washed to remove soil and other impurities. Water contained in rice husks is evaporated by oven at 100ºC for 4 h. The dried rice husk is then burned in the furnace for 3 hours at 600ºC. The obtained rice husk ash is then washed with HCl to remove the metal content. After that, rice husk ash was washed by dissolving in distilled water until reach neutral pH. The neutral pH of rice husk ash let dried in an oven at 150℃ for 4 h. The white ash product from this process was named silica.

2.2 Sodium Silicate Preparation

The neutral silica was pounded carefully to obtain fine particle [13]. After that, as much as 10 g of silica which was dissolved into 82.5 ml NaOH 4 M was boiled under constant stirring, and let the solution evaporated and form dark gel. Formed gel was poured to the petri dish thoroughly and then anneal it at elevated temperature (500℃) for 3 h. The final Filtrate that was produced in this step was called sodium silicate (Na_2SiO_3).

2.3 Sodium Aluminate Preparation

A number of sodium hydroxide (20 g) was incorporated to 100 ml distilled water, briefly. After that place the solution to the constant stirring for 1 h and 100℃ heating. Then poured 8,5 g aluminium oxide to the sodium hydroxide solution gradually.

2.4 Zeolite Preparation

Both of Sodium aluminate and sodium silicate solution were blend homogeneously by constant stirring, homogeneous mixture was then fed into the microwave at various power of 100 W, 180 W, for 60 min. The unmodified sample was treated by stirred in constant stirring for 5 h, in 80℃ of heating. The next stage was neutralized the zeolite using distilled water thoroughly. The neutral zeolite was then anneal at 400ºC before it was characterized.

2.5 Characterization

X-ray diffraction is a common method for analyzing the structure of a crystal. In this study sample was analyzed by Shimadzu-7000. The testing was start by interacting the sample with electromagnetic waves of X-rays that have a wavelength λ= 0.5-2.5 Å [14].

3. Result and Discussion

Effect of microwave irradiation on zeolite morphology also has investigated in our previous study [15]. Compare the unmodified zeolite, treated zeolite by microwave irradiation proved to be more effective in formed crystalline structure in time preparation. Microwave irradiation process was carried out the object with rapidly and selectively heating process. It's one of equipment that can be rotate and spread out of the energy to the object. Table 1 shows the crystallinity of both modified and unmodified zeolite. Zeolite without irradiation had highest percentage of crystallinity compare others. In this case unmodified zeolite under 5 h stirring and heating, large more time compare to modified

zeolite that was needs 1 h preparation. Transformation silica contained in ash was deeply depend on annealing temperature. In this work sample of zeolite was burnt in temperature of 600-700℃, this condition was high enough to form crystalline structure on zeolite. This result slightly different which is explained by Nair and Co-workers, the amorphous silica can be produced by combustion process at 500-700℃, greater temperature was form crystallinity on zeolite [16]. This condition might be caused by different kind of paddy-utilize in the research.

Table 1. The crystallinity of modified and unmodified zeolite.

Sample of zeolite	Crystallinity (%)
Unmodified zeolite	93,684
Treated 100 W	90,807
Treated 180 W	90,212

Effect of irradiation power is studied in 0 W, 100 W, and 180 W, 1 h. Both sodium silicate and sodium aluminate were mixed in constant stirring for several minutes before heating process. The influence of microwave irradiation on the sample is analyzed by XRD method to determine the percentage of each component. High crystallinity was indicated by strong peak at 2θ: 10-11, 22-24 and 29-31. Auerbach et al [17] identified range of crystallinity zeolite with cube shape in measurement 2θ:7-8, 10-11, 22-24, and 29-31. The XRD date was analyzed by software Match to determine component of prepared zeolite. Untreating zeolite resulted in the least percentage of silica (3.3 %), Unmodified sample contain of 3.3% silica and 96.7% alumina, these indicate the blending process of sodium aluminate and sodium silicate produces less bind of each compound. It was slightly different to the treated zeolite that is resulting more contain of silica. Microwave irradiation of 100 W result of the highest (16.1 wt %) amount of SiO_2, follow by treated zeolite 180 W which is contain of 5.8 wt % of SiO2. The result of the investigation tells us that microwave irradiation contribute to the binding process among sodium aluminate and sodium silicate. This result shows microwave irradiation affect to the zeolite properties.

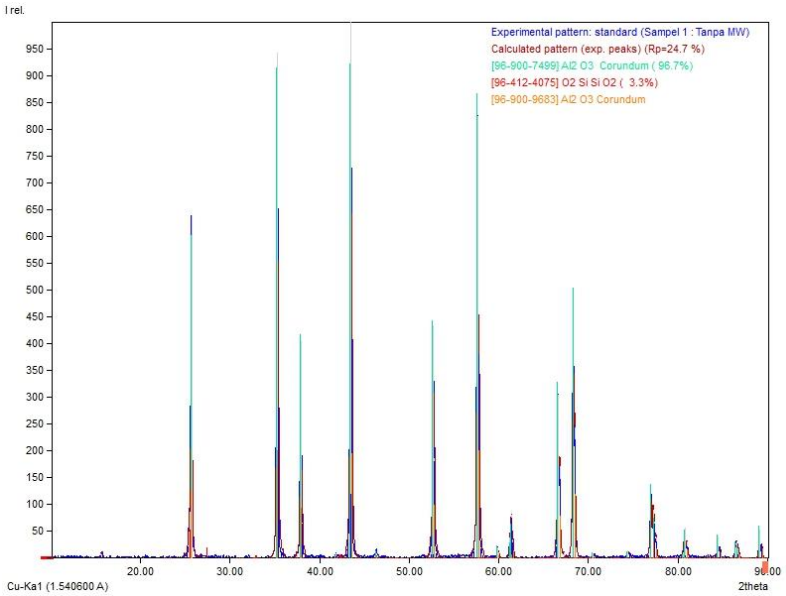

Figure 1. Unmodifed sample (0 W).

According to the previous studied, crystallinity forming was start at 60 min in zeolite preparation by microwave irradiation [18]. XRD trace of modified zeolite running under 100 W shown in Fig 2. Based on the strong peak of XRD diffractograms, sample resulting high crystallinity. According to the Match calculation, sample with 100 W irradiation for 1 h has highest silica value i.e. 16.1wt %. These

conditions indicate that irradiation process gives some prospective method than the conventional method.

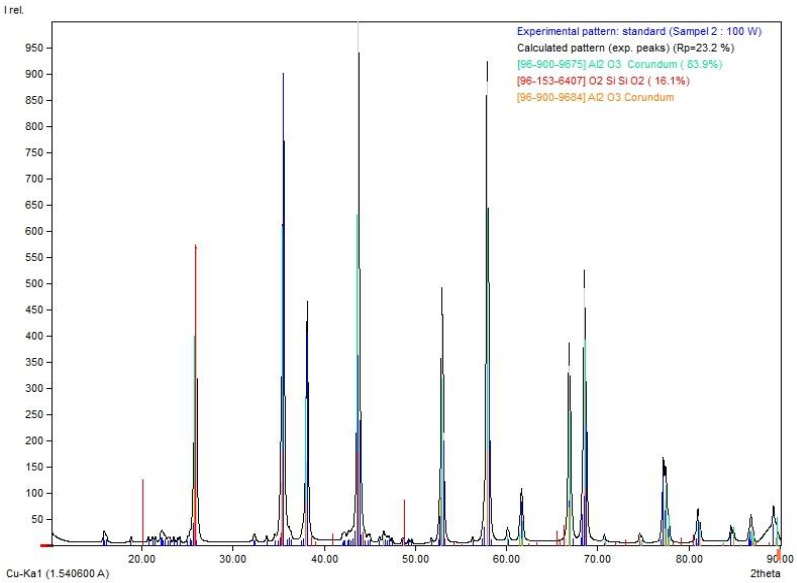

Figure 2. Modified Sample 100 W.

The main structure of modified sample 180 W can be seen in Fig 3. XRD diffractograms tells the same crystallinity of modified sample in 100 W. This result indicates that the power modification from 100 W to 180 W did not gave significant effect. The data from Match analyze give the fact that sample contains of 5.8% silica and 94.2% alumina. These might be caused by the leaching process to get a neutral pH.

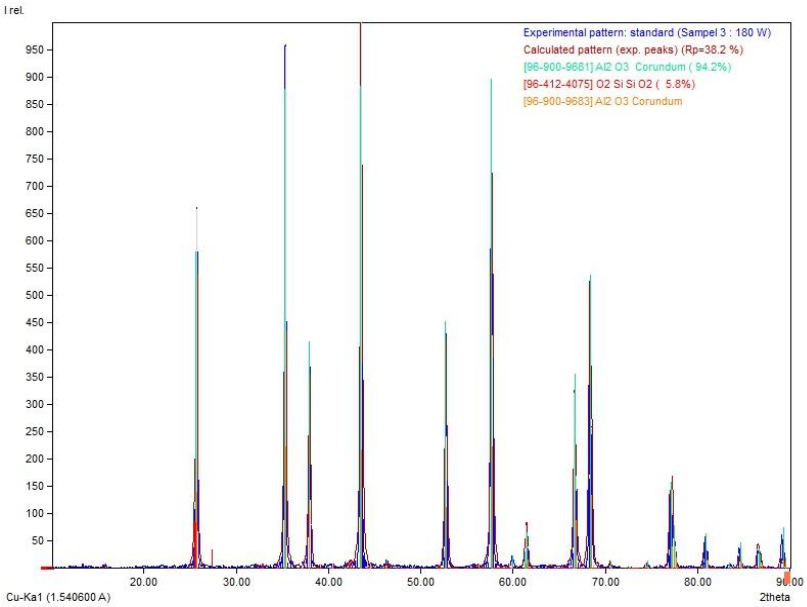

Figure 3. Modified sample 180 W.

XRD date showed that both modified and unmodified sample resulting high crystallinity. Furthermore, it is also found that these can be expressed by each single correlation line. Hence, the crystalline phase of the generated zeolite does not depend on the kind microwave irradiation. The reason behind this statement is the time sample preparation without any irradiation was take longer than modified. This experimental result support the hypothesis that the microwave irradiation contributes to high crystallinity in short time.

Effect of microwave irradiation in purification process

Effect of zeolite adsorbent capability coupled with microwave irradiation is studied by adsorption process of waste cooking oil, which is, has 9 meq/kg of peroxide value. The peroxide value is fixed as parameter waste cooking oil quality. Process of adsorption is carried out by constant stirring and 60℃ of heating in ratio 1:10 zeolite and waste cooking oil. The number of peroxide value each sample was analyzed by iodometric method. Result of peroxide value after adsorption process shows in table 2 as follows.

Table 2. The Peroxide Value before and after adsorption.

Sample	Peroxide Value (meq/kg)
Waste cooking oil	9
Unmodified zeolite	6,4
100 W adsorption	5
180 W adsorption	2

Microwave irradiation contribute in adsorption capability, it is indicated by decreasing the number of peroxide of wasted cooking oil. Increasing of annealing temperature will affect to the zeolite morphology i.e. surface area. Pakpahan and Co-workers (2017) [19] have been reported that increasing of temperature lead the decreasing of surface area and increasing particle size of zeolite. This morphology was contribute to the adsorption capability.

4. Conclusion

Zeolite with high crystallinity was success prepared by microwave irradiation. Comparing with conventional method, microwave irradiation increasing crystallinity of zeolite synthetic in short time (1 h). Unmodified sample (without irradiation) needs more preparation time (5 h). Microwave heating more effective in zeolite synthetic of rice husk ash than conventional heating. Microwave irradiation also increase the capability of zeolite synthetic applied in waste cooking oil purification. The peroxide value for waste cooking oil was decreased by adsorption with modified zeolite.

5. Reference

[1] Badan Pusat Statistik 01 March 2016 Accessed 2017 from *https://www.bps.go.id/brs/view/1271*
[2] Prasertsan S and Sajjakulnukit B 2006 Biomass and biogas energy in Thailand: Potential, opportunity and barriers *Renewable Energy* **31** pp 599-610
[3] Chandra S 1996 *Waste Materials Used in Concrete Manufacturing* (Westwood: William Andrew)
[4] Real C, Alcala M and Criado J.M. 1996 Preparation of silica from rice Husks *Journal of the American Ceramic Society* **79** pp 2012–16
[5] Patel M, Karera A and Prasanna P 1987 Effect of thermal and chemical treatment on carbon and silica contents in rice husk *Journal of Material Science* **22** pp 2457-64
[6] Kunihiro Fukui, Keiji Kanayama, Tetsuya Yamamoto and Hideto Yoshida 2007 Effects of microwave irradiation on the crystalline phase of zeolite synthesized from fly ash by hydrothermal treatment *Advanced Powder Technol* **18** pp. 381–393
[7] Yanagida S, Nikawa Y, Katsuki H, Kato K, Takizawa H and Tokuda M 2003 *New Industrial Technology of Microwave* pp. 4–8 (Japan: NTS)
[8] Somiya S and Roy R 2000 Hydrothermal synthesis of fine oxide powders *Bull. Mater. Sci*. **23** pp 453–460
[9] Querol X, Alastuey A, López-Soler A, Plana F, Andrés JM and Juan R. 1997 A fast method for recycling fly ash:microwave-assisted zeolite synthesis *Environ. Sci. Technol.* **31** pp 2527-33.
[10] Inada M, Tsujimoto H and Eguchi Y, Enomoto N and Hojo J 2005 Microwave-assisted zeolite synthesis from coal fly ash in hydrothermal process *Fuel* **84** pp 1482–6
[11] Arafat A, Jansen J C, Ebaid A R and Van Bekkum H 1993 Microwave preparation of zeolite Y and ZSM-5 *Zeolites* **13** pp 162-5

[12] Ponsot I, Bernardo E, Bontempi E, Depero L E, Detsch R, Chinnam R K and Boccaccini A R 2015 Recycling of pre-stabilized municipal waste incinerator fly ash and soda-lime glass into sintered glass-ceramics *J. Clean. Prod.* **89** pp 224-230
[13] Muljiyanti D R, Nuryono and Kunarti E S 2010 Sintesis dan karakterisasi silika gel dari abu sekam padi yang dimobilisasi dengan 3-(trimetoksisilil)-1 propantiol (Yogyakarta: Sekolah Pasca Sarjana Universitas Gadjah Mada Yogyakarta)
[14] Wahyuni T E 2003 *Hand Out Metode Difraksi Sinar X* (Yogyakarta: Laboratorium Kimia Analitik FMIPA UGM)
[15] Suryani P E, Candra A D 2017 Pengaruh Iradiasi Microwave Terhadap Luas Permukaan Spesifik Pada Zeolit Sintetis Dari Abu Sekam Padi *Seminar Nasional Hasil Penelitian (SNHP)-VII* Semarang
[16] Nair D G, Fraaij A, Klaassen A A K and Kentgens A P M 2008 A structural investigation relating to the pozzolanic activity of rice husk ashes *Cement and Concrete Research* **38** pp 861–869
[17] Auerbach S M, Carrado K A and Dutta P K 2003 *Hand book of zeolite science and technology Marcel Dekker* (New York: Basel,Switzerland)
[18] Fuadi A M, Malik Musthofa, Kun Harismah, Haryanto and Nur Hidayati 2012 Pembuatan Zeolit Sintetis Dari Sekam Padi *Simposium Nasional RAPI XI FT UMS-2K012*
[19] Pakpahan, Jennifer Kapriati, Karo, Pulung Karo, Suroto and Bambang Joko 2017 Studi Luas Permukaan Spesifik Zeolit Akibat Pengaruh Mikrostruktur dan Potensinya Sebagai Elektrode Superkapasitor *Jurnal Teori dan Aplikasi Fisika* **5** 1 pp 19-24