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THE EFFECT OF AGING TIME ON ZSM-5 PRODUCTION FROM SILICEOUS PALM OIL FLY ASH

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ABSTRACT

Factories used palm oil solid waste in empty bunches, shells, and fiber (coir) as boiler fuel. The combustion process in this boiler produces by-products in the form of fly ash and bottom ash. Oil palm fly ash and bottom ash content were 37.98% (100 kg/week) and 64.36% (3-5 tons/week), respectively. After refluxing of fly ash, the SiO2 content of 96.8% is used to synthesize ZSM-5. The synthesis was carried out with the aging variation of 6, 12, and 24 h, and without aging time at the temperature of 60 °C. The hydrothermal was carried out at a temperature of 150 °C for 10 and 18 h. The results show an amorphous phase over 10 h hydrothermal, and ZSM-5 has formed over 18 h hydrothermal. ZSM-5 was formed and sodalite phase without aging time for 18 h hydrothermal. Analcime and sodalite formed for 6 h aging time. ZSM-5, analcime, faujasite, and sodalite formed over 12 and 24 h aging time. With a longer time of aging, the analcime phase dominated. It caused the metastable MFI structure to open to transform into alumina-rich structures. The morphology of ZSM-5 synthesized with hydrothermal 18 h with 6 h of aging is spheric and trapezohedron.

Keywords: fly ash, silica, ZSM-5, hydrothermal, aging time.

INTRODUCTION

Indonesia is one of the largest agricultural countries in the world, with palm oil as its flagship commodity. Based on data from the Central Statistics Agency (2018), the total area of oil palm plantations in Jambi Province in 2016 was 476.413 ha with a total production of 1.031.215 tons/year. The palm oil processing industry negatively impacts the form of large organic waste-garbage in the form of empty bunches, shells, and fibers or fibers. Shells and fibers or fibers are used as fuel in the boiler. Boilers (boilers) produce steam power. Burning palm kernel shells and fibers produce ash in 2 types, namely bottom ash and fly ash [1].

Oil palm fly ash from boiler combustion waste is grayish-white to blackish. Silica is the main ingredient in fly ash. Fly ash from palm oil industry combustion waste contains 50.57% SiO_2. After preparation, the silica content increased to 80.92% [2]. In the XRF analysis conducted by Ranjbar et al. [3], oil palm fly ash has a higher silica content, namely 64.169%. Saputra et al. [4] reported that fly ash from palm oil industrial waste could produce silica precipitation with a high SiO_2 content of 92.288%. Fly ash was refluxed with NaOH solution, and then the filtrate was added with 10 N HCl to get silica precipitate. High silica from fly ash can be used as a source of silica in the synthesis of zeolites such as ZSM-5 [5].

Zeolite Socony Mobil-5 (ZSM-5) is a zeolite classified as a zeolite with high Si content. So that ZSM-5 is widely used as a hydrocracking catalyst and has high porosity [6]. Also, ZSM-5 has a long life, organophilic properties, and three-dimensional structure that makes it more selective to the formation of hydrocarbons with C ≤11 chains and resistant to heat and acid treatment [7].

The synthesis of zeolite ZSM-5 carried out by Kardatos *et al*. [8] used rice husk ash as a source of silica. The silica content in rice husk ash used was 92.3%. The synthesis was carried out with a molar ratio of 12NaOH: $30SiO_2$: $1Al_2O_3$: 6TPABr: $1800H_2O$. TPABr was used as an organic template to direct the formation of ZSM-5 crystals. The hydrothermal process was carried out at a temperature of 150 °C for 1 day. The hydrothermal product was calcined to remove TPABr. Based on the XRD diffractograms, the peak of zeolite ZSM-5 at 2θ of 8° and 24°. These are the specific peaks of ZSM-5. The ZSM-5 produced was a crystal with an average size of 0.8 µm at low temperature and high temperature 2-8µm with a specific surface area of 375 m^2/g. FT-IR absorption at wavenumbers 1100, 800, and 450 cm^{-1} showed vibrations from internal Si, tetrahedral AlO_4 and observed in silica, quartz, and cristobalite. Furthermore, the absorption at wavenumbers 550 and 1230 cm^{-1} showed the double tetrahedral ring's vibrations and the asymmetrical stretching of Si and AlO_4 in the zeolite framework.

Research conducted by Hartanto *et al*. [9] by aging at a temperature of 60 °C using a variation of 6, 12, 24 h, and without aging. This study used a silica source from TEOS, an Al source from sodium aluminate, and TPAOH as a template. The results showed ZSM-5 resistance increased with increasing aging time. The ZSM-5 has a pore size of about 3.8 nm. Increasing the aging time can increase the number of ZSM-5 precursors that are formed. At 24 h of aging, the highest crystallinity was 65%.

Hartati *et al*. [10] synthesized ZSM-5 from metakaolin. The synthesis was carried out at hydrothermal temperatures ranging from 120, 150, and 170 °C. The synthetic optimum temperature was obtained at a temperature of 150 °C. The result of synthesis at 120 °C for 72 h has not yet formed ZSM-5 because the XRD diffractogram shown is the widened peak for amorphous solids. At a temperature of 150 °C, ZSM-5 was formed, but with increasing hydrothermal time, other phases were

found, such as mordenite and quartz. The longer the hydrothermal time causes the reduction in the typical peak of ZSM-5. At a temperature of 150 °C, the optimal hydrothermal time is 24 h. At a temperature of 170 °C with a hydrothermal time of 24, 48, and 60 h, there was no peak showing the characteristics of ZSM-5. The higher the hydrothermal temperature, the metastable phase ZSM-5 will form a more stable phase such as mordenite and quartz.

Based on the description above, silica extraction from fly ash from the palm oil industry is used as a source of Si for ZSM-5 synthesis. The synthesis was carried out using the hydrothermal method at 150 °C using the TPABr template. This study aims to study aging time and hydrothermal time on ZSM-5 synthesis from oil palm bottom ash.

MATERIAL AND METHODS

The materials used in this study were Fly Ash from palm oil industry (PT BBIP, Mendahara, Tanjung Jabung Timur, Jambi Province, Indonesia), sodium hydroxide pellets (Merck, NaOH, ≥97%), HCl, aluminum sulfate (Merck, $Al_2(SO_4)_3.16H_2O$), Sosium Aluminate (Sigma-aldrich, $NaAlO_2$), Tetrapropylammonium bromide (Sigma-aldrich, TPABr).

Extraction of Silica from Fly Ash

The preparation of fly ash follows the procedure carried out by Ali, *et al.* [11]. The bottom ash was washed, filtered and calcined at temperature of 750 ºC for 6 h. The gray fly ash (Figure-1a) changes to white ash (Figure-1b). 12 g of white ash was refluxed for 4 h using 4 M NaOH. The reflux results are filtered, so we get a yellowish filtrate which is sodium silicate. The filtrate was added with HCl to pH 9, so that a white gel is formed which is then allowed to stand for 2 days. The white gel is filtered and washed to neutral pH. The solid is dried and crushed. The yield of white silica powder in this sample ash was 45.7%, as shown in Figure-1c. The chemical composition of palm oil bottom ash was showed in Table-1.

Figure 1. a. Fly ash before calcination, (b) fly ash after calcination and (c) silica presipitation

Synthesis of ZSM-5 via Template

Synthesis of zeolite ZSM-5 using 3.01 g of silica from palm oil industry fly ash extraction and 0.57 g of NaOH dissolved in 15 mL of distilled water (solution A). 2.46 g of TPABr and 0.28 g of sodium aluminate were dissolved in 35 mL of distilled water (solution B). Solution B is added to solution A and then put into a stainless steel autoclave [8]. Furthermore, the mixture was preserved using a stainless steel autoclave with variations of 6, 12, and 24 h at a temperature of 60 ºC [9]. After the aging process, it is followed by a hydrothermal process at a temperature of 150 ºC for 18 h. After that, it was filtered and washed with distilled water until the filtrate pH was 8 and dried at 110ºC, followed by calcination at 550ºC [4].

Instrumental Techniques

The Fourier transform infrared (FT-IR) spectra were recorded on a Shimadzu-IRPrestige21 FT-IR, single beam spectrometer with a resolution of 2 cm^{-1}. The samples were ground with KBr (1:100 ratio) as a tablet and mounted to the sample holder in the cavity of the spectrometer. The measurements were recorded at room temperature in the region at 4000–400 cm^{-1}. XRD diffraction patterns of ZSM-5 were measured by a PANalytical XPert MPD diffractometer with a Cu-K*f*Ñ as source of X-ray operating at 40 kV and 30 mA. Samples were scanned in the range 2θ of 5-90º with a step size of 0.017 and scan step time of 10.15 s. The existence of the crystalline phase of ZSM-5 was confirmed from the diffraction patterns recorded on 2θ = 5-80º. SEM instrument (model:EVO MA10) was used to study the surface morphology of ZSM-5 equipped with energy-dispersive X-ray analysis spectroscopy (EDX) to determine the Si/Al composition. Chemical composition of POBA and silica solid was examined by Xray fluorescence spectroscopy (XRF: Philips, PW1400).

RESULTS AND DISCUSSIONS

Based on Table-1, an increase in SiO_2 levels occurred from 22.9% to 96.8% after chemical treatment. The SiO_2 contained in fly ash can dissolve in an alkaline state which comes from NaOH. Soluble SiO_2 forms sodium silicate. HCl was added to the natrium silicate filtrate to pH 9 to form silica gel. Gel formation occurs through the formation of Si-O-Si siloxane bonds [7].

Table-1. The chemical composition of fly ash.

Compounds	Composition (wt.%)		
	Before Calcination	After Calcination	After Extraction
SiO_2	22.9	30.9	96.8
P_2O_5	2.1	7.28	-
SO_3	0.2	3.1	-
K_2O	4.44	11.2	2.26
CaO	64.5	38.5	0.35
TiO_2	0.17	0.34	-
MnO	0.17	0.31	0.036
Fe_2O_3	2.17	3.19	0.068
CuO	0.092	0.16	0.064
MoO_3	2.5	3.1	-
MgO	0.2	1.1	-
Al_2O_3	0.39	0.65	-
BaO	0.1	0.09	-
Na_2O	-	-	-

Fourier Transform Infrared Spectroscopy

Based on Figure-2, it can be seen that the sample, 6; 12; 24 h and without aging, has a wavenumber close to commercial ZSM-5. ZSM-5 samples without aging have an absorption band at wavenumber 443.64, 609.53, 744.55, 1014.59, 1637.62, and 3487.42 cm^{-1}. The absorption at wavenumber 443.64 cm^{-1} shows the bending vibration of T-O (Si-O or Al-O). At 18 h hydrothermal, a double ring has been formed, indicated by the absorption at 609.53 cm^{-1} [5, 12]. It is also interpreted that zeolite has been formed, shown by the absorption band at 744.55 cm^{-1}, the zeolite fingerprint area [13]. Symmetric stretching vibration of Si-O and or Al-O in the absorption band 744.55 and 1014.62 cm^{-1}. The absorption at wavenumber 1637.62 cm^{-1} shows the OH mechanical vibration of Si-OH. In addition, there is also absorption at wavenumber 3487.42 cm^{-1} which shows the stretching vibration of OH from Si-OH [9].

The absorption band formed from the interpretation of the functional groups of the ZSM-5 sample with an aging time of 6 h, at 439.78 cm^{-1}, shows the bending vibration of T-O [13]. 613.38 cm^{-1} forms an absorption band interpreted to form a double ring of ZSM-5 [5, 12]. In addition to forming the double ring, there is also an absorption band representing the zeolite fingerprint area at wavenumber 742.62 cm^{-1}. The absorption indicates the stretching vibration of Si-O and or Al-O at wavenumbers 1018.45 and 742.62 cm^{-1}. The absorption band also shows OH vibrations from Si-OH with bending vibrations at 1647.26 cm^{-1} and stretching vibrations at wavenumber 3483 cm^{-1} [9, 13].

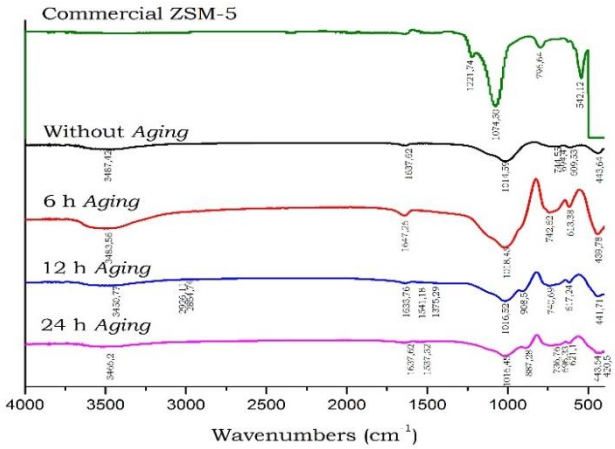


Figure-2. Synthesis of ZSM-5 infrared spectra with aging time variation.

The ZSM-5 sample absorption with an aging time of 12 h at a wavenumber of 441.71 cm^{-1} showed a bending vibration of T-O. The absorption at wavenumber 617.24 cm^{-1} is interpreted as forming a double ring [12]. The absorption at wavenumber 740.69 cm^{-1} is the area of the zeolite fingerprint [13]. The absorption bands formed at wavenumbers 1016.52, 908.50, and 740.69 cm^{-1} showed a stretching vibration of Si-O and/or Al-O symmetry [5]. A wavenumber 1633.76 cm^{-1} and absorption show the OH bending vibration of Si-OH and the stretching vibration at 3483.56 cm^{-1}. [9]. In this sample, there is a new absorption band at wavenumber 1542.18 cm^{-1} which is interpreted as the presence of a C-H bond and the stretching vibration absorption C-H at wavenumbers 2854.74 and 2926.11 cm^{-1}. The presence of the C-H bond indicates that the template remains in the zeolite sample. TPABr organic sites do not decompose completely in the calcination process [13].

Synthesis of ZSM-5 with an aging time of 24 h has an absorption band at a certain wavenumber. In this sample, the calcination after synthesis cannot completely decompose the template. This is known from the absorption at the wavenumber 1537.32 cm^{-1} which shows the C-H bond of the template [13]. The absorption at wavenumber 420.50 cm^{-1} shows the bending vibration of T-O. The zeolite formation in this synthesis is known from the absorption at wavenumbers 738.76 and 696.33 cm^{-1}, the zeolite fingerprint areas, and double rings shown at 621.10 cm^{-1}. The presence of symmetrical stretching vibrations from Si-O and or Al-O at the absorption of 110.45, 887.28, and 736.76 cm^{-1}. Absorption at wavenumber 1637.62 cm^{-1} shows the OH bending vibration of Si-OH and the stretching vibration at 3466.20 cm^{-1} absorptions [5, 9, 12-13].

The four samples of ZSM-5 have an absorption band that indicates zeolite formation, the absorption band in the fingerprint area (700-800 cm^{-1}). The uptake from the fingerprint of zeolite showed variations in Si-O and Al-O. It is strengthened again by absorption at the wavenumber 500-650 cm^{-1}, indicating a pentasile double ring from ZSM-5. The four ZSM-5 samples with a hydrothermal time of 18 h with an aging time of 6 h, the FTIR spectra

were closer to the commercial ZSM-5. The ZSM-5 sample with 6 h aging time had a higher absorption intensity, seen from the sharper absorption band. The more functional groups of zeolites that are formed, the higher the intensity of the absorption band [5].

X-Ray Diffraction

Based on the diffraction pattern in Figure-3, the ZSM-5 diffraction pattern has distinctive peaks at 2θ 7-8° and 22 to 25° [10]. Commercial ZSM-5 has a diffraction pattern with characteristic peaks at 2θ of 7.9444°, 8,8184°, 22.1866°, and 23.0439°.

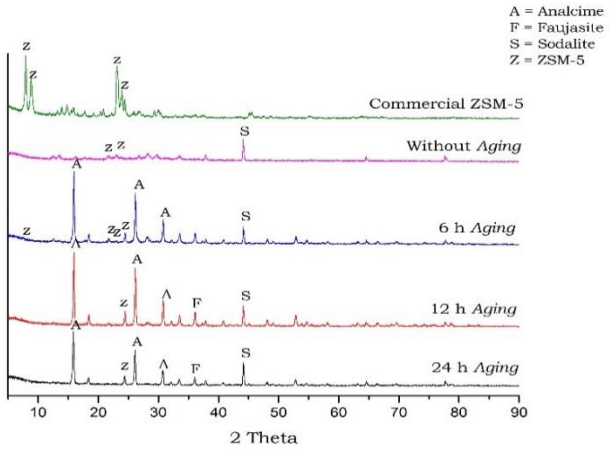


Figure-3. XRD Diffractograms of commercial ZSM-5, without Aging, 6, 12 and 24 h aging.

The zeolite sample without aging has a peak with very high intensity at 2θ of 44.12°, which is the peak of sodalite and other peaks with low intensity. Among them are the peaks with low intensity at 2θ of 22.98° and 23.73°. The small peaks are the peaks of ZSM-5. Samples with an aging time of 6 h had peaks that represented the formation of ZSM-5, namely at 2θ of 7.47°, 21.80°, 23.05°, and 24.45°. Also, peaks formed at 2θ of 15.96°, 26.17°, and 32.13° which are the peaks of analcime. The peak at 2θ of 44.14° is the peak of sodalite [14-15]. Samples with an aging time of 12 h had peaks that represented the formation of ZSM-5 at 2θ of 24.46°. Also, peaks formed at 2θ of 15.96°, 26.15°, and 30.82° which are the peaks of analcime [16]. At 2θ of 36.04°, there is a peak that indicates the identity of the faujasite. The peak at 2θ of 44.14° is the peak of sodalite [15].

Samples with a 24 h aging time had peaks that represented the formation of ZSM-5 at 2θ of 24.46° [8]. Peak also formed at 2θ of 15.84°, 26.14°, and 30.81°, which are the peaks of analcime. At 2θ of 36.06°, there is a peak that indicates the identity of the faujasite. The peak at 2θ of 44.14° is the peak of sodalite [15].

In the zeolite sample without aging, besides forming ZSM-5, also formed sodalite. Likewise, the three other samples also formed sodalite, and there are also other types of zeolites, analcime and faujasite. As the aging time increases, the intensity of the formation of ZSM-5 decreases. This is because the ZSM-5 structural framework formed is less stable so that it dissolves again and builds a more stable zeolite framework. In the zeolite sample diffraction with an aging time of 12 and 24 h, it can be seen that the dominant zeolite formed is analcime. There is a disproportion where the ZSM-5 that should be formed decomposes into zeolites with a higher alumina content, such as the analcime phase [17].

The formation of other phases besides ZSM-5 is also due to the transformation of the zeolite framework and the possible binding of other cations to the zeolite framework [18]. Aging factors affect the formation of the analcime phase in this synthesis. Very long aging caused the metastable MFI structure to transform into structures rich in silica and alumina, such as analcime [19]. It also occurred in Putro and Prasetyoko [16] research, besides ZSM-5, which also formed an analcime phase. The analcime phase contains low SiO_2/Al_2O_3 [6].

Based on the results of XRD characterization, the sample with an aging time of 6 h and hydrothermal 18 h had 5 diffraction peaks representing the peaks of ZSM-5. Although the peaks formed are still in low intensity, these peaks prove that ZSM-5 has been formed. According to Putro and Prasetyoko [16] (2007) and Prasetyoko et al. [9], the peaks formed at 2θ of 7.46°, 21.96°, 23.05°, and 24.45° indicate the formation of MFI formation from ZSM-5 (Figure-4). XRD and hkl diffractogram of ZSM-5 synthesized at 6 h aging time with 18 h hydrothermal time. ZSM-5 peaks formed at 2θ of 7.46°, 21.96°, 23.05°, and 24.45° have the hkl (011), (023), (031) and (313) fields. These results are consistent with the hkl field in the Collection of Simulated XRD Powder Patterns for Zeolite, which states that XRD ZSM-5 peaks are formed at 2θ of 7.9°, 8.9°, 21.8°, 22.2°, 23.1°, 23.98°, and 24.61° with the hkl (011), (200), (023), (340), (051), (033) and (313) fields [15].

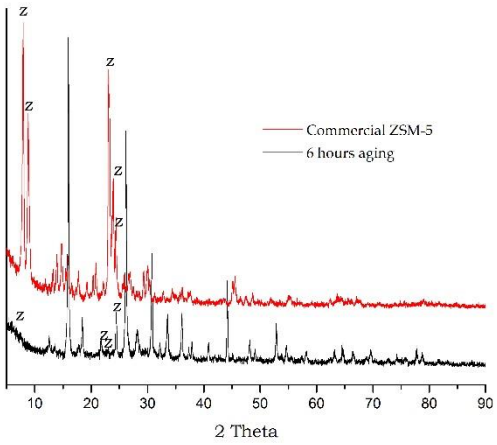
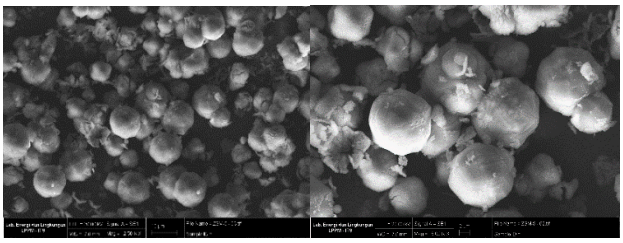


Figure-4. XRD Difractogram of commercial ZSM-5 and 6 h aging.

Scanning Electron Microscope-Energy Dispersive X-Ray Spectroscopy (SEM-EDX)

Characterization to see surface morphology and elemental composition of materials that have been

synthesized using Scanning Electron Microscopy-Energy Dispersive X-Ray Spectroscopy (SEM-EDX). In Figure-5, it can see characterization results with the magnification of 2500 and 5000 times showing the surface morphology of the synthesized ZSM-5 at 18 h hydrothermal with 6 h of aging.



a) b)

Figure-5. The morphology of ZSM-5 Synthesis in hydrothermal 18 h and aging 6 h a) Magnification of 2500x and b) 5000x.

The surface morphology of synthetic ZSM-5 has a round shape with a size that is not uniform. This is following the research of Panpa and Jinawath [20]. The ZSM-5 synthesized has a morphology in the form of round crystals. In Figure-5, you can see that there are still impurities around the crystal. Hydrothermal 18 h with 6 h of aging formed the Trapezohedron phase. These results are the same as described by Sari and Agustini [21]. ZSM-5 was synthesized using the TPABr template. It indicated the analcime phase. In Figure-5, you can see that there are still impurities around the crystal. The crystal particle size was not uniform, with size variations ranging from 4 µm to 14 µm.

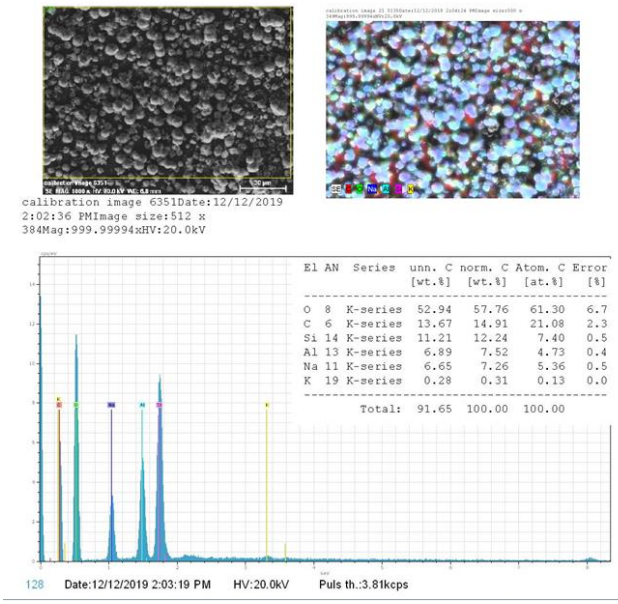


Figure-6. SEM-EDX results of hydrothermal synthesis of ZSM-5 at 18 h and aging for 6 h.

The SEM-EDX analysis results in Figure-6 show the elemental composition of ZSM-5, which was synthesized under hydrothermal conditions of 150 °C for 18 h and with 60 °C aging for 6 h. Based on Figure-6, it can be seen that the elements contained are in the form of a zeolite framework, namely O, Si, Al, and Na, but there are also elements of K and C. Each composition of O, Si, Al, and Na in the synthetic zeolite is 61.3%, 7.4%, 4.73%, and 5.36%. The presence of element K (0.13%) comes from silica extracted from oil palm fly ash. Element C, which is quite large (21.08), comes from the organic container used, namely TPABr. Element C should not be found if post-hydrothermal calcination is complete. Perfect calcination will decompose the whole TPABr. However, the presence of element C is assumed to be less long in calcination. In the study by Krisnandi *et al*. [22] that calcination was carried out at 500 ° C for 8 h, the results obtained by the ZSM-5 did not contain C flour. The distribution of each element in the ZSM-5 sample, it is also seen in Figure-6. The Si/Al ratio is around 1.56.

CONCLUSIONS

Silica extracted from fly ash containing of 96.8% SiO_2 has been used to synthesize ZSM-5. According to the results, the synthesis was successfully carried out by utilizing fly ash as a source of silica under hydrothermal conditions at 150 °C for 18 h, with aging at 60 °C for 6 h. The results showed the characteristics of ZSM-5 in XRD analysis, which was characterized by a peak at 2θ 7.46°, 21.96°, 23.05°, and 24.45°. Based on SEM analysis, the morphology of ZSM-5 was crystals with spheric. The crystalline form of analcime is trapezohedron.

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REFERENCES

[1] Pahan I. 2007. Panduan Lengkap Kelapa Sawit. Jakarta: Penebar Swadaya.

[2] Kongnoo, A., S. Tonsisirin., P. Worathanakul., C. Phalakornkule. 2017. Surface Characteristics and CO_2 Adsorption Capacities of Acid-Activated Zeolite 13X Prepared From Palm Oil Mill Fly Ash. Fuel. 193: 385-394.

[3] Ranjbar N., M. Mehrali., A. Behnia., U. J. Alengaram., M. Z. Jumaat. 2014. Compressive Strength and Microstructural Analysis of Fly Ash/Palm Oil Fuel Ash Based Geoplymer Mortar. Material and Design. 1-26.

[4] Saputra R., E. Saputra., Yelmida. 2018. Sintesis ZSM-5 Menggunakan Silika Presipitasi dari Fly Ash Pabrik CPO. Jom FTEKNIK. 5(1).

[5] Zahrina I., Yelmida., F. Akbar. 2012. Sintesis ZSM-5 dari Fly Ash Sawit Sebagai Sumber Silika dengan Variasi Nisbah Molar Si/Al dan Suhu Sintesis. Jurnal Rekayasa Kimia dan Lingkungan. 9(2): 94-99.

[6] Trisunaryanti, W. 2018. Konversi Fraksi Aspal Buton Menjadi Fraksi Bahan Bakar. Yogyakarta: Gadjah Mada University Press.

[7] Handayani P. A., E. Nurjanah dan W. D. P. Rengga. 2015. Pemanfaatan Limbah Sekam Padi Menjadi Silika Gel. Jurnal Bahan Alam Terbarukan. 4(2): 55-59.

[8] Kardatos K., A. Ntziouni., L. Illiadis., V. K. Rigopoulou. 2013. Utilization of Amorphous Rice Husk Ash for the Synthesis of ZSM-5 Zeolite Under Low Temperatures. Journal Mater Cycles Waste Management. 15: 571-580.

[9] Hartanto D., T. E. Purbaningtias., H. Fansuri., D. Prasetyoko. 2011. Karakterisasi Struktur Pori dan Morfologi ZSM-5 Mesopori yang disintesis dengan Variasi Waktu Aging. Jurnal Ilmu Dasar. 12(1): 80-90.

[10] Hartati H., A. K. Widati., A. Purwaningsih., A. N. Kristanti. 2018. Suhu dan Waktu Hidrotermal pada Sintesis ZSM-5 dari Metakaolin. Jurnal Kimia Riset. 3(1): 38-46.

[11] Ali I.O., Hasan A., Shabaan S., El-Nasser K.S. 2013. - Synthesis and characterization of composite catalysts Cr/ZSM-5 and their effects toward photocatalytic degradation of p-nitrophenol. Arabian Journal of Chemistry. S2106-S2114.

[12] Sauqi R., F. Akbar., Yelmida. 2014. Sintesis ZSM-5 dari Fly Ash Sawit dengan Variasi Waktu Sintesis dan Waktu Kalsinasi. Jurnal FTeknik. 1(2): 1-10.

[13] Octaviani, S. 2012. Sintesis dan Karakterisasi Zeolit ZSM-5 Mesopori dengan Metode Desilikasi dan Studi Awal Katalis Oksida Metana. Universitas Indonesia.

[14] Metta A., S. B. Ginting., H. Saputra. 2014. Sintesis ZSM-5 dari Coal Fly Ash (CFA) dengan Sumber Silika Penambah yang Berasal dari Abu Sekam Padi: Pengaruh Rasio SiO_2/Al_2O_3 terhadap Kristanilitas Produk. Jurnal Rekayasa Proses. 8(2): 62-68.

[15] Treacy M. M. J., J. B. Higgins. 2001. Collection of Simulated XRD Powder Patterns for Zeolites. United Kingdom: Elsevier.

[16] Putro A. L., D. Prasetyoko. 2007. Abu Sekam Padi sebagai Sumber Silika pada Sintesis Zeolit ZSM-5 Tanpa Menggunakan Template Organik. Akta Kimindo. 3(1): 33-36.

[17] Cundy C. S., P. A. Cox. 2005. The hydrothermal of Zeolites: Precursor, Intermediates and Reaction Mechanism. Microporous and Mesoporous Material. 83: 1-78.

[18] Hanifah, R. D. Puryanto., A. Muttaqin. 2018. Pembuatan Zeolit Sodalit dari Abu Dasar Batubara dengan Variasi Konsentrasi Larutan $NaAlO_2$ Menggunakan Metode Peleburan Alkali Hidrotermal. Jurnal Fisika Unand. 7(3): 229-232.

[19] Jacobs P. A., J. A. Martens. 1987. Synthesis of High-Silica Aluminosilicate Zeolites. United Kingdom: Elsevier

[20] Panpa W., S. Jinawath. 2009. Synthesis of ZSM-5 Zeolite and Silicalite from Rice Husk Ash. Applied Catalysis B: Enviromental. 90: 389 - 394.

[21] Sari E. F., A. P. Agustina. 2018. Perbandingan Pengaruh Sintesis ZSM-5 dengan Template Organik dan tanpa Template organik. Review Artikel. Surabaya. Institut Teknologi Sepuluh Nopember.

[22] Krisnandi Y. K., F. M. Yanti., S. D. S. Murti. 2017. Synthesis of ZSM-5 Zeolite from Coal Fly Ash and Rice Husk: Characterization and Application for Partial Oxidation of Methane to Methanol. IOP Conference Series: Materials Science and Engineering.