Jurnal Rekayasa Kimia & Lingkungan
(Journal of Chemical Engineering and Environment)
Volume 19, Number 1, Page 45–53, June 2024
ISSN 1412-5064, e-ISSN 2356-1661
https://doi.org/10.23955/rkl.v19i1.33921

Synthesis of Zeolite from Fly Ash and Bottom Ash and Application for Biodiesel Transesterification

Rika Putri[1*], Fadjar Goembira[1], Maria Ulfah[2], Muhammad Andrifar[1], Rati Yuliarningsih[1], Rizki Aziz[1]

[1]Enviromental Engineering Departement, Faculty of Engineering, Universitas Andalas, Limau Manis, Padang 25163, West Sumatera, Indonesia
[2]Chemical Engineering Departement, Faculty of Engineering, Bung Hatta University Jl. Gajah Mada No.19, Padang 25143, Sumatra Barat, Indonesia
Email: rika.putri18@yahoo.com

Article History
Received: 28 August 2023; Received in Revision: 20 February 2024; Accepted: 28 February 2024

Abstract

Burning coal in a Coal-Fired Power Plant produces by-products like fly ash and bottom ash. Zeolite synthesized from the ash in Teluk Sirih Coal-Fired Power Plant was applied as a catalyst in the biodiesel transesterification reaction. Zeolite synthesis used the hydrothermal method with acid pretreatment. The operating conditions for fly ash zeolite are a 2.4-molar ratio SiO2/Al2O3 and a crystallization time of 6 hours. The bottom ash zeolite used a 2.0-molar ratio SiO2/Al2O3 and a crystallization time of 8 hours. The performance test of the synthesized catalyst was carried out in the transesterification reaction using waste cooking oil as a raw material with a free fatty acid content of 0.7%. The synthesized catalyst was characterized using x-ray diffraction, scanning electron microscope, and Bruneuer-emmet-teller. The biodiesel with the highest yield was analyzed based on SNI 7182: 2015. The synthesis results of the catalyst produced type A zeolite, shown by the typical X-ray diffraction pattern and supported by the morphological test results using a cube-shaped. The surface area of zeolite fly ash and bottom ash is 12.87 m2/g and 5.13 m2/g. The test showed fly ash zeolite had the highest biodiesel yield of 89.66%. Based on the characterization using SNI 7182: 2015, the color and free glycerol met the standards, while density 40 ˚C, kinematic viscosity 40 ˚C, acid number, total glycerol, methyl ester content, and water content did not meet the standards.

Keywords: biodiesel, fly ash and bottom ash, transesterification reaction, waste cooking oil, zeolite a

1. Introduction

Biodiesel as an alternative fuel has attracted global attention to replace conventional diesel fuel in recent years. Biodiesel is environmentally friendly and easily degraded, as indicated by its clean and non-toxic combustion properties, and also produces less carbon and SO_2 emissions (Mahlia et al., 2020). In addition, biodiesel has characteristics similar to diesel (Da Costa et al., 2018). Raw materials for biodiesel can use free fatty acids (FFA), animal fats, and vegetable oils (Đặng et al., 2017).

More than 95% of the feedstock for biodiesel is dominated by edible oil (Roschat et al., 2017). Using food oil as a raw material for biodiesel can impact food oil scarcity and high prices (Suzihaque et al., 2022). One alternative to replace food oil is waste cooking oil (WCO).

As a raw material for biodiesel, WCO can be a solution to solving environmental problems and it can reduce 60-70% of the final cost (Farooq and Ramli., 2015). In addition, WCO is also easier to find, such as on a household and restaurant scale, and can also be collected from small and medium enterprises. Based on survey results, the potential for WCO in Indonesia reaches 4,000,000 tons per year (Renilaili., 2022). Biodiesel can be produced through a transesterification reaction between triglycerides and alcohol with the help of a catalyst. The catalysts used in biodiesel transesterification can be homogeneous catalysts, enzymes, and heterogeneous catalysts. Currently, research on the transesterification of biodiesel mainly concentrates on the use of heterogeneous catalysts. In addition to regenerating a heterogeneous base catalyst, it performs 4,000 times faster than an acid catalyst

To cite this article: Rika Putri, Fadjar Goembira, Maria Ulfah, M. Andrifar, Rati Yuliarningsih, Rizki Aziz. (2024) Synthesis of Zeolite from Fly Ash and Bottom Ash and Application for Biodiesel Transesterification, *Jurnal Rekayasa Kimia & Lingkungan,* 19(1), 45-53. https://doi.org/10.23955/rkl.v19i1. 33921

(Sharma et al., 2008). Several heterogeneous catalysts have been studied in the transesterification of biodiesel, such as CaO (Kesserwan et al., 2020), TiO_2 (De & Boxi., 2020), SrO (Li et al., 2020), and including zeolite (Manique et al., 2017).

Zeolite is a porous crystalline aluminosilicate material with a three-dimensional framework structure with uniform molecular dimensional pores (Indira & Abhitha., 2022). The waste from burning coal-fired power plants (CFPP), called fly ash and bottom ash (FABA), rich in SiO_2 and Al_2O_3, can be used as a source of raw materials for zeolite because it has a high surface area, adjustable pore size, and customizable framework (Bhandari et al., 2015). Utilizing fly ash as raw material for zeolite synthesis will not only improve ecological balance but also add value to FABA waste. Teluk Sirih coal-fired power plant in Padang city produces around 5,000 tonnes of FABA waste per month (Putri et al., 2019).

Zeolite synthesis generally uses a hydrothermal process. The source of the raw material influences the composition of the zeolite synthesis. Manique et al. (2017) have studied the synthesis of zeolite by utilizing fly ash waste from Candiota-Brazil CFPP, with a Si/Al 3-molar ratio, 24-hour crystallization time, and 100ºC crystallization temperature to produce sodalite (Manique et al., 2017). However, very few researchers study the use of zeolite A from fly ash and bottom ash as a catalyst in the biodiesel transesterification reaction.

In this research, the synthesis of zeolite A from FABA waste from the Teluk Sirih CFPP was carried out using the hydrothermal method to be used as a catalyst in the transesterification process of biodiesel from waste cooking oil. The synthesized zeolite was characterized using X-Ray diffraction (XRD), Scanning Electron Microscope (SEM), and Brunauer-Emmet-Teller (BET). The resulting biodiesel was evaluated using SNI 7182: 2015.

2. Methodology

2.1. Materials

Fly ash and bottom ash were collected from the coal-fired power plant of Teluk Sirih in Padang City. WCO was collected from Sahabat Alam Waste Bank in Pariaman City. Other materials such as distilled water, NaOH, $NaAlO_2$, methanol, and ethanol were supplied by CV. Vahana Scientific.

2.2. FABA Waste Pretreatment Process

The SiO_2 content in the fly ash and bottom ash samples of the Teluk Sirih coal-fired power plant was 25.85% and 40.44%, which are classified as low-calorie coal (Goembira et al., 2023). Based on Belviso's (2018) research results, low-calorie coal has a low SiO_2 and a high impurity content (Belviso., 2018). The presence of impurities in the raw material can interfere with the crystallization process and the stability of the zeolite (Ren et al., 2020). In order to remove impurities and increase the SiO_2 content, pretreatment must be carried out first. The pretreatment procedure was adapted from Goembira et al. (2023). FABA was mixed with HCl solution (10%) with the ratio v/w HCl: FABA 25:1 (L/S) in a beaker. Then the beaker was put in a water bath at 80ºC for 1 hour. FABA was filtered, washed, repeatedly using distilled water, and dried at 100ºC (Goembira et al., 2023).

2.3. Zeolite Synthesis Process

Synthesis of Fly Ash Zeolite

The zeolite synthesis and fly ash composition used a molar ratio of $SiO_2/Al_2O_3 \simeq 2.4$, $Na_2O/SiO_2 \simeq 1.7$, and $H_2O/Na_2O \simeq 55$. The zeolite synthesis method was adapted from Manique et al. (2017) and Milton (1959). A total of 9.63 grams of NaOH was dissolved in 166 ml of H_2O. The NaOH solution forms solution A. The solution was then divided into solution B and solution C. To solution B, 7.87 grams of $NaAlO_2$ was added, while 8.55 grams of SiO_2 was added to solution C. Solution C was poured into solution B, and the moving process was completed for 15 minutes at a rotational speed of 300 rpm. After the solution was homogeneous, the aging process occurred for 90 hours. Crystallization at 100ºC lasted for 6 hours. The washing process was running until the zeolite pH was 9. Finally, a drying process followed at 105ºC for 4 hours.

Synthesis of Bottom Ash Zeolite

The composition for synthetic bottom ash zeolite used a molar ratio of $SiO_2/Al_2O_3 \simeq 2.0$, $Na_2O/SiO_2 \simeq 1.7$, and a ratio of $H_2O/Na_2O \simeq 55$. The zeolite synthesis method was adapted from Manique et al. (2017) and Milton (1959), where 8.78 grams of NaOH dissolved in 166 ml of H_2O to form solution A. Then the solution was divided into solution B and solution C. Solution B was added by 9.44 grams of $NaAlO_2$ while 8.93 grams of SiO_2 (bottom ash) was added to solution C. The next step is the same as the zeolite synthesis process from fly ash but the crystallization time lasted for 8 hours.

2.4. Biodiesel Transesterification Process

FFA Analysis

FFA has an essential role in the transesterification reaction. The content of FFA in the feedstock plays a role in determining the process of making biodiesel. The FFA feedstock is >2%, and an esterification reaction must be carried out first to reduce the FFA content, whereas if the FFA is <2%, the transesterification reaction can be carried out immediately (Andrifar et al, 2022). The analysis procedure and calculation of the FFA content were adapted from Daramola et al. (2015). As much as 1 ml WCO was diluted using 10 ml isopropyl alcohol 99% titrated with 0.1 M NaOH solution drop by drop to the endpoint of the titration using phenolphthalein solution as a pH indicator (Daramola et al., 2015). Calculating the FFA content in the feedstock can be seen in Equation (1) adapted from (Daramola et al., 2015).

$$\%\mathrm{FFA} = (\mathrm{V}-\mathrm{b})\mathrm{x}\ \mathrm{N}\frac{28.2}{w} \quad (1)$$

Where %FFA is the free fatty acid ($C_{18}H_{34}O_2$) content in the feedstock; V is the titrant volume (25 ml); b is the blank volume (24.7 ml); N is the concentration of titration solution (0.1 mg/l); and *w* is the weight of sample WCO.

Transestterification Process

The procedure for the transesterification reaction was adapted from Zhang et al. (2020). Firstly, the waste cooking oil with the volume measured (30 ml) was transferred into a three-neck boiling flask and heated to 65^{o}C with continuous stirring (300 rpm). In the next step, 50 ml of methanol and 2% wt of catalyst were added to wco initializing the time of reaction. It took 45 minutes for the whole reaction process. At the end of the reaction, the mixture was transferred to a separatory funnel and let sit overnight. The obtained biodiesel was purified with water.

$$\%\ \mathrm{Yield} = \frac{\text{product volume}}{\text{feedstock valume}} \times\ 100 \quad (2)$$

Catalyst Characteristic Analysis

The resulting catalyst was analyzed using X-ray diffraction (XRD) X'PERT PRO PANalytical to determine the crystal structure formed. The analysis was carried out over 2Å and angles from 5-75° with CuK irradiation. The surface morphology of the zeolite was analyzed by a Scanning Electron Microscope (SEM) Hitachi S-3400N, equipped with an energy-dispersive X-ray analyzer (EDX) with a Back Scattered Electron (BSE) detector. The pore size of the zeolite was analyzed by Brueneur-Emmet-Teller (BET) and was performed using Quantachrome Nova 4200e equipment.

Biodiesel Analysis

The resulting biodiesel was analyzed based on SNI 7182: 2015. Parameters analyzed included acid number, density, free glycerol, total glycerol, water and sediment, methyl ester content, kinematic viscosity, and colour.

3. Results and Discussion

3.1. Characteristics of The Synthesized Zeolite

Zeolite synthesized from FABA CFPP Teluk Sirih was denoted by zeolite fly ash (ZFA) and bottom ash (ZBA). Figure 1 shows the comparison of the raw FABA color with the synthesized zeolite. Zeolite fly ash has a grey color and fine powder.

Figure 1. Physical characteristics of raw FABA and zeolite, (a) fly ash, (b) bottom ash, (c) ZFA, (d) ZBA

The changes in the properties and color of zeolite were caused by the operating conditions and methods used in the zeolite synthesis process (Kusumawardani et al., 2023). The results produced greyish-white zeolite whose colour is similar to the one obtained by Hidayat et al. (2020). Meanwhile, bottom ash zeolite has a dark brown color with a coarser particle shape than fly ash zeolite.

Then, ZFA and ZBA analyzed the characteristics of the crystals formed using the XRD instrument. Based on Figure 2, the results of the characteristics of zeolites ZFA and ZBA using XRD show that the zeolite peaks that appears are zeolite A, mullite, and quartz. The peaks of zeolite A in ZFA appear at 2θ = 12.33°; 16.01°; 21.66°; 23.89°; 30.71; 33.14. Meanwhile, the peak of zeolite A in ZBA based on the XRD results appears at 2θ= 13.86°; 16.04°; 24.36°; 27.86°; 29.85. According to Indira & Abhitha (2022), the peak of zeolite A appears at 2θ = 7.2°; 10.3°; 12.6°; 16.2°; 21.8°; 24°; 26.2°; 27.2°; 30°; 30.9°; 31.1°; 32.6°; 33.4°; and 34.3°.

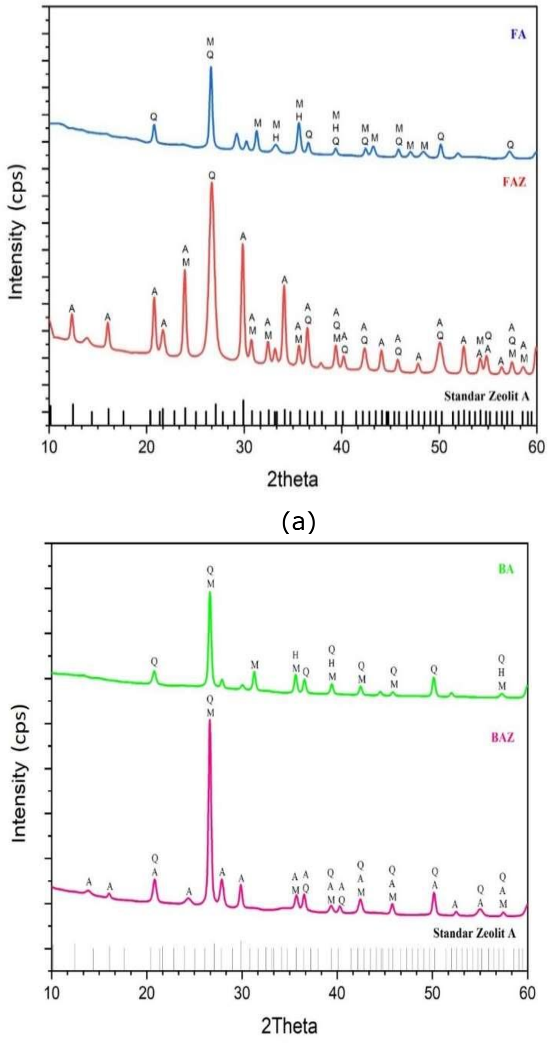
The purity level of the resulting zeolite is calculated from the % crystallinity using the formula adapted from Ren et al. (2020), which can be seen in Equation (3).

$$\% \text{ Cristalinity} = \frac{\sum \text{intensity of XRD peak (product)}}{\sum \text{intensity of XRD peak (standard)}} \times 100 \quad (3)$$

Based on the results of the calculation using Equation 3, it was found that the % crystallinity in the fly ash zeolite was 65.63%. These results indicate that the zeolite is not pure and contains other impurities such as quartz which has not been fully converted to zeolite, and the presence of mullite. The components have not been wholly converted due to the low solubility of these minerals in NaOH (Manique et al., 2017). The bottom ash zeolite obtained a crystallinity of 36.22%. As with zeolite fly ash, the crystallinity and quartz and mullite compounds are still low.

The factors that influence the formation of zeolite A are the concentration of NaOH and the time of zeolite crystallization. Research by Liu et al. (2019) states that the concentration of NaOH affects the amount of zeolite A produced; the higher the concentration of NaOH used, the greater the amount of zeolite A produced (Liu et al., 2019). Murayama et al. (2002) in their study obtain that the concentration of Na^{+} ions in a basic solution is the main factor that determines the total reaction rate of zeolite synthesis. Another factor that drives the formation of zeolite A is the crystallization time.

Based on the experiments conducted by Murukutti and Jena (2021), it was discovered that the best crystallization time in the synthesis of zeolite A is 6 hours at a temperature of 100 ºC. Research by Liu et al. (2019) shows that a lot of zeolite A was formed at 6 hours of crystallization. ZFA and ZBA also carried out the results of surface analysis of the catalyst using a Hitachi S-3400N SEM. Based on Figure 3, it can be seen that ZFA and ZBA are type A zeolites. The crystals of zeolite A are cube-shaped (Iqbal et al., 2019; Liu et al., 2019).



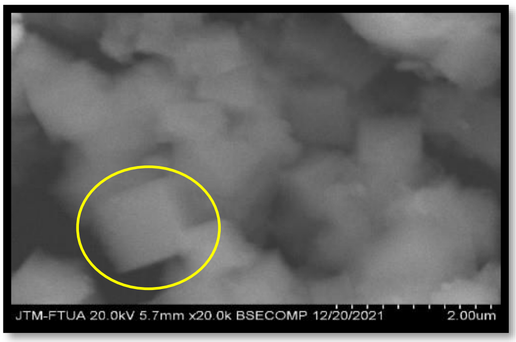
(a)

(b)

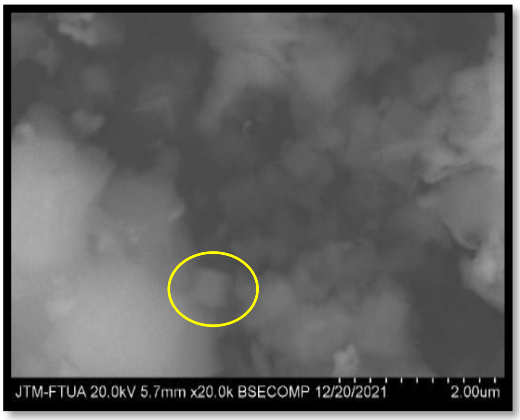
Figure 2. XRD pattern of (a) ZFA, (b) ZFB.

The presence of quartz and mullite was also observed in the SEM results which were shown to be chunks (Liu et al., 2020). From the SEM results of ZFA and ZBA, it can be seen that the formed zeolite A is not clear. This is caused by crystal growth which may be hindered by the presence of an uneven surface (smaller crystals), which can be justified by the presence of impurities in the ash which causes rapid nucleation (Manique et al., 2017).

Heterogeneous catalyst performance is determined by selectivity and activity. Both of these are related to the catalyst's physical properties, such as the catalyst's surface area and porosity, size, and distribution, as well as the morphology and structure of the catalyst (Haryono et al., 2018). BET analysis was performed to determine the surface area, pore diameter, and pore volume of the zeolite.



(a)



(b)

Figure 3. SEM image of (a) ZFA, (b) ZBA.

Based on the test results for zeolite A using N_2 adsorption-desorption, the surface area of the zeolite and the pore volume of the zeolite can be seen in Table 1. The synthesized catalyst's characteristics show that the surface area of ZFA is larger (12.87 m^2/g) than ZBA (5.13 m^2/g). ZFA also has a larger pore volume than ZBA.

The pore characteristics of the catalyst material are determined based on the isotherm curve, which results from a plot of the volume of N_2 gas adsorption-desorption in the pore material under Standard Temperature and Pressure (STP) conditions against the relative pressure of the gas at constant temperature (P/P_0). The formation of pores on the catalyst is indicated by the presence of a "bottled shape" on the curve (Kunecki et al, 2017). The BET isotherm curve consists of Type I, II, III, IV, V, and VI curves. The results of ZFA and ZBA characterization are depicted in Figure 4.

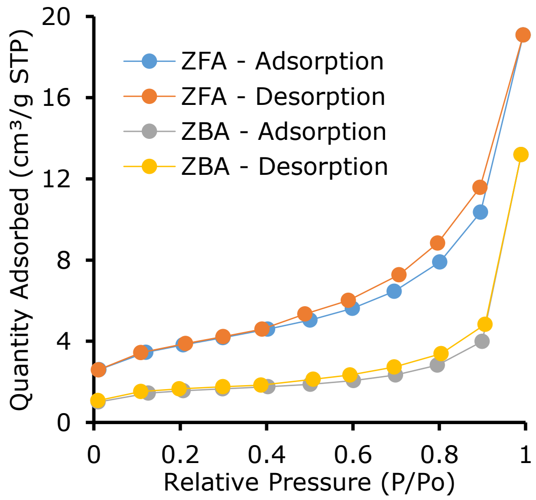


Figure 4. N_2 adsorption-desorption isotherms for ZFA and ZBA

The figure shows that ZFA and ZBA are categorized into type IV isotherm curves. The type IV isotherm curve is characterized by the formation of multilayer hysteresis, which is indicated by the loss of the saturation point. From the curve results, the zeolite pore size is classified as mesoporous with a pore size of 2-50 nm (Haul., 1969). The formation of mesopores is characterized by hysteresis loops at P/Po 0.4 in both fly ash and bottom ash zeolite. Other similar studies' results also showed P/Po in the range of 0.4-1 to indicate a mesoporous type of zeolite (Sholeha., 2017; Zhang et al., 2020).

3.2. Catalyst Performance Test

The catalyst performance test was completed using waste cooking oil. The FFA of waste cooking oil is 0.7%. The operating conditions of the transesterification reaction were accomplished at a temperature of 65 °C, a reaction time of 45 minutes, a 12:1 methanol: oil ratio, and 2% wt catalyst. Yield of biodiesel produced can be seen in Table 2. The yield indicates that ZFA is higher than ZBA. The high yield on biodiesel with ZFA catalyst is related to the characteristics of the catalyst used such as the surface area and pore volume. In the transesterification reaction, the reactions occur in the pores and on the surface of the catalyst so that the surface area and pore volume affect the biodiesel yield.

Table 1. BET results from FABA zeolites

No	Sample	Surface area (m^2/g)	Pore diameter		Pore volume (cm^3/g)
			nm	Å	
1.	ZFA	12.87	11.45	114.5	0.027
2.	ZFB	5.13	20.84	204.0	0.019

Table 2. Catalyst performance test results

No.	Sample	% Yield
1.	ZFA	89.66
2.	ZBA	88.83

Large surface area can increase catalytic activity in biodiesel production because it allows more contact with reactant molecules to produce high yields. The pore size of ZFA is larger than ZBA; according to Haryono et al. (2018), pore size affects biodiesel yield. The color of the biodiesel produced from the catalytic process of the transesterification reaction can be seen in Figure 5. The biodiesel from the zeolite fly ash produced has a slightly brighter color than the zeolite from bottom ash.

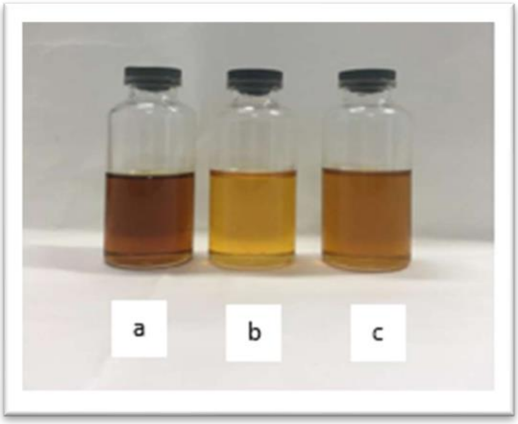


Figure 5. Biodiesel color (a) WCO, (b) biodiesel with ZFA zeolite, (c) biodiesel with ZBA zeolite

3.3. Characterization of The Biodiesel

Zeolite fly ash was tested for performance in producing 2 litres of biodiesel to analyze the characteristics of biodiesel using SNI 7182:2015. The transesterification occurred at a temperature of 65^{o}C, a reaction time of 45 minutes, the ratio of methanol: oil of 12: 1, and the weight of the catalyst 2% wt. The results of biodiesel characterization using SNI 7182: 2015 can be seen in Table 3. The table presents that some of the test parameters of the biodiesel produced have not passed the specified standard limits. These parameters are density (40 ^{o}C), kinematic viscosity, acid number, total glycerol, and alkyl ester content.

The high density of biodiesel produced is due to impurities in biodiesel and the presence of residual methanol in biodiesel (Setiawati & Edwar., 2012). The low concentration of methyl ester is caused by the very short reaction time; thus, not all triglycerides are converted to ester alkyl (Elma et al., 2018). The incomplete separation between biodiesel and glycerol causes a high total glycerol content (Zalfiatri et al., 2019).

Table 3. Biodiesel characterization based on SNI 7182: 2015

Parameters	Unit	Standard	Result
Density (40 ^{o}C)	Kg/m^3	860-900	903
Kinematic viskosity (40 ^{o}C)	cSt	1.9-6.0	47.700
Color	-	3	L 2.500
Acid number	mg KOH/g	0.500	1.440
Free glycerol	% wt	0.020	0.005
Total glycerol	% wt	0.240	0.323
Ester alkil content	% wt	96.500	90.220
Water content	ppm wt	500	999

According to Sanjiwani et al. (2015), the cause of high acid concentration is peroxide compounds from the oxidation of unsaturated fatty acids are oxidized to form aldehyde compounds. Aldehyde will be further oxidized to form carboxylic acid compounds, which increase the amount of acid (Sanjiwani et al., 2015). The high kinematic viscosity is caused by the presence of OH compounds, which tend to form hydrogen bonds with hydrogen atoms (Keera et al., 2018). This research needs to be further refined to obtain biodiesel that complies with SNI 7182: 2015.

4. Conclusion

The synthesis of zeolite A from FABA of Teluk Sirih coal-fired power plant in Padang city to be used as a catalyst in transesterification has the potential as a catalyst in the biodiesel transesterification reaction. The type of synthetic zeolite produced is zeolite A, with a purity level of zeolite A fly ash and zeolite A bottom ash 65.63% and 36.22%, respectively. The surface shape of the catalyst from the SEM results is in the cube form with a surface area of zeolite A fly ash and zeolite A bottom ash 12.87 m^2/g and 5.13 m^2/g, respectively. Testing the performance of zeolite A fly ash and zeolite A bottom ash produces biodiesel yields of 89.66% and 88.83%. The characterization results for biodiesel using zeolite A fly ash based on SNI 7182: 2015 show that several parameters meet the standards, such as the color and free glycerol. From the results obtained, it can be concluded that zeolite A from the synthesis of FABA waste from the Teluk Sirih PLTU has the potential as a catalyst in the biodiesel transesterification reaction for better results. Further research can be carried out by looking for optimum conditions in the biodiesel transesterification reaction.

Author Contributions

Conceptualisation, R.K., F.G.; Formal analysis, M.U., R.K.,; Investigation, R.K., M.A.,; Project administration, R.Y.,; Supervision, F.G.,; Writing – original draft, R.K., R.A.,; Writing – review and editing, R.K.

Acknowledgement

This study was done under financial support from Directorate General for Higher Education, Ministry of Education, Culture, Research and Technology of the Republic of Indonesia, under contract No.104/E4.1/AK.04.PT/2021.

Data Available Statement

Available on request to the corresponding author.

Conflict of Interest

The author declares that there is no conflict of interest regarding the publication of this manuscript.

References

Andarini, N., Haryati, T. & Lutfia, Z. (2018) Sintesis zeolit A dari abu terbang (fly ash) batubara variasi rasio molar Si/Al. *Jurnal ILMU DASAR*, 19(2), 105–110. https://jurnal.unej.ac.id/index.php/JID/article/view/5910

Andrifar, M. Goembira, F., Ulfah, M., Putri, R., Yuliarningsih, R., & Aziz, R. (2022) Optimization of sustainable biodiesel production from waste cooking oil using heterogeneous alkali catalyst, *Jurnal Rekayasa Proses,* 16(2), 4–9. https://doi.org/10.22146/jrekpros.74373

Anya, A.U. Ugwumma, C., & Okoro, A.I.P. (2013) Effect of free fatty acid content on the yield of biodiesel derived from neem oil, *Journal of Basic and Applied Chemistry*, 3(1), 1–4.

Belviso, C. (2018) State-of-the-art applications of fly ash from coal and biomass: A focus on zeolite synthesis processes and issues, *Progress in Energy and Combustion Science*, 65, 109–135. https://doi.org/10.1016/j.pecs.2017.10.004

Bhandari, R., Volli, V. & Purkait, M.K. (2015) Preparation and characterization of fly ash based mesoporous catalyst for transesterification of soybean oil, *Journal of Environmental Chemical Engineering*, 3(2), 906–914. https://doi.org/10.1016/j.jece.2015.04.008

Da Costa, Martins, J., Lopes, D.Q., & Brito, F.P. (2018) Comparison between diesel and biodiesel produced from used cooking oil on Diesel engine performance, *Conference: 3ª Conferência Internacional em Produção do Conhecimento Científico em Timor-LesteAt: Dili, Timor-Lest*e

Đặng, T.H., Chen, B.H. & Lee, D.J. (2017) Optimization of biodiesel production from transesterification of triolein using zeolite LTA catalysts synthesized from kaolin clay, *Journal of the Taiwan Institute of Chemical Engineers*, 79, 14–22. https://doi.org/10.1016/j.jtice.2017.03.009

Daramola, M.O., Nkazi, D. & Mtshali, K. (2015) Synthesis and evaluation of catalytic activity of calcined sodium silicate for transesterification of waste cooking oil to biodiesel, *International Journal of Renewable Energy Research*, 5(2), 517–523. https://doi.org/10.20508/ijrer.v5i2.2164.g6606

De, A. & Boxi, S.S. (2020) Application of Cu impregnated TiO_2 as a heterogeneous nanocatalyst for the production of biodiesel from palm oil, *Fuel*, 265, 117019. https://doi.org/10.1016/j.fuel.2020.117019

Elma, M., Suhendra, S.A., & Wahyuddin, W. (2018) Proses Pembuatan Biodiesel Dari Campuran Minyak Kelapa Dan Minyak Jelantah. *Konversi*, 5(1), 8. https://doi.org/10.31213/k.v5i1.23

Goembira, F. Putri, R., Ulfah, M., Yuliarningsih, R., Andrifar, M., & Aziz., R. (2023) Evaluation of biodiesel catalysts derived from coal-fired Teluk Sirih power plant fly ash and bottom ash, *AIP Conference.* https://doi.org/10.1063/5.0129148

Haruna, I., Fatima, M., & Ndam, V. (2015) Effect of high free fatty acid feedstock on methyl esters yield using bulk calcium oxide catalyst, *International Journal of Scientific & Technology Research*, 4(3), 186–189.

Haryono, Natanael, C.L., Rukiah., & Yulianti, Y.B., (2018) Kalsium oksida mikropartikel dari cangkang telur sebagai katalis pada sintesis biodiesel dari minyak goreng bekas, *Jurnal Material dan Energi Indonesia*, 8(1), 8–15.

Haul, R. (1969) Adsorption, surface area & porosity*, Zeitschrift für PhysikalischeChemie*, 63(1), 220. https://doi.org/10.1524/zpch.1969.63.1_4.220

Indira, V. & Abhitha, K. (2022) A review on recent developments in zeolite a synthesis for improved carbon dioxide capture: Implications for the water-energy nexus, *Energy Nexus*, 100095. https://doi.org/10.1016/j.nexus.2022.100095

Iqbal, A. Sattar, H., Haider, R., & Munir, S. (2019) Synthesis and characterization of pure phase zeolite 4A from coal fly ash, *Journal of Cleaner Production*. 219, 258–267. https://doi.org/10.1016/j.jclepro.2019.02.066

Keera, S.T., El Sabagh, S.M. & Taman, A.R. (2018) Castor oil biodiesel production and optimization, *Egyptian Journal of Petroleum*, 27(4), 979–984. https://doi.org/10.1016/j.ejpe.2018.02.007

Kesserwan, F. Ahmad, M.N., Khalil, M., & Rassy, H.E. (2020) Hybrid CaO/Al_2O_3 aerogel as heterogeneous catalyst for biodiesel production, *Chemical Engineering Journal*, 385, 123834. https://doi.org/10.1016/j.cej.2019.123834

Kunecki, P. panek, R., Wdowin, M., & Franus, W. (2017) Synthesis of faujasite (FAU) and tschernichite (LTA) type zeolites as a potential direction of the development of lime class C fly ash, *International Journal of Mineral Processing*, 166, 69–78. https://doi.org/10.1016/j.minpro.2017.07.007.

Kusumawardani, L.J., Iryani, A., & Sinaga, E.L. (2023) Modification of zeolite made from coal fly ash with TiO_2: Effect of aging time on physical and optical properties, *Makara Journal of Science*, 27(1), 63-72. https://doi.org/10.7454/mss.v27i1.1483

Li, H. Liu, F., Ma, X., Cui, P., Guo, M., Li, Y.,Gao, Y., Zhou, S., & Yu, M. (2020) An efficient basic heterogeneous catalyst synthesis of magnetic mesoporous Fe@C support SrO for transesterification, *Renewable Energy*, 149, 816–827. https://doi.org/10.1016/j.renene.2019.12.118

Liu, F. Ma, S., Ren, K., & Wang, X. (2020) Mineralogical phase separation and leaching characteristics of typical toxic elements in Chinese lignite fly ash, *Science of the Total Environment*, 708(xxxx), 135095. https://doi.org/10.1016/j.scitotenv.2019.135095

Liu, Z. Li, Z., Li, L., Wang, J., Zhou, Y., & Wang, D. (2019) One-step high efficiency crystallization of zeolite A from ultra-fine circulating fluidized bed fly ash by hydrothermal synthesis method, *Fuel*, 257, 116043. https://doi.org/10.1016/j.fuel.2019.116043

Mahlia, T.M.I*.*, Syazmi, Z.A.H.S., Mofijur, M., Abas, A.E.Pg., Bilad, M.R., Ong, H.C., & Silitonga, M.R. (2020) Patent landscape review on biodiesel production: Technology updates, *Renewable and Sustainable Energy Reviews*, 118, 109526. https://doi.org/10.1016/j.rser.2019.109526

Manique, M.C, Lacerda, L.V., Alves, A.K., & Bergmann, C.P. (2017) Biodiesel production using coal fly ash-derived sodalite as a heterogeneous catalyst, *Fuel*, 190, 268–273. https://doi.org/10.1016/j.fuel.2016.11.016

Milton, R.M, Penemu; assignor to union carbide corporation, a corporation of New York. December 24, (1959) Molecular sieves adsorbents. US patents. Us2882243

Murayama, N., Yamamoto, H. & Shibata, J. (2002) Zeolite synthesis from coal fly ash by hydrothermal reaction using various alkali sources, *Journal of Chemical Technology and Biotechnology*, 77(3), 280–286. https://doi.org/10.1002/jctb.604

Murukutti, M.K. & Jena, H. (2021) Synthesis of nano-crystalline zeolite-A and zeolite-X from Indian coal fly ash, its characterization and performance evaluation for the removal of Cs^{+} and Sr^{2+} from simulated nuclear waste, *Journal of Hazardous Materials*, 423(PA), 127085. https://doi.org/10.1016/j.jhazmat.2021.127085

Putri, R.D., Taufiq, I. & Norokhim (2019) Analisis radionuklida pada Fly Ash dan Bottom Ash PLTU Teluk Sirih menggunakan spektrometer gamma, *Jurnal Fisika Unand*, 8(4), 387–393, https://doi.org/10.25077/jfu.8.4.387-393.2019

Ren, X. Liu, S., Qu, R., Xiao, L., Hu, P., Song, H., Wu, W., Zheng, C., Wu, X., & Gao, X., (2020) Synthesis and characterization of single-phase submicron zeolite Y from coal fly ash and its potential application for acetone adsorption, *Microporous and Mesoporous Materials*, 295, 109940. https://doi.org/10.1016/j.micromeso.2019.109940

Renilaili, (2022) Pemanfaatan minyak jelantah menjadi biodiesel bahan bakar cair alternative dengan metode pengadukan yang konstan, *Jurnal Tekno*, 19(April), 11–19.

Roschat, W. Siritanon, T., Yoosuk, B., Sudyoadsuk, T., & Promarak, V. (2017) Rubber seed oil as potential non-edible feedstock for biodiesel production using heterogeneous catalyst in Thailand, *Renewable Energy*, 101, 937–944. https://doi.org/10.1016/j.renene.2016.09.057

Sanjiwani, N., Suaniti, N. & Rustini, N. (2015). Bilangan peroksida, bilangan asam, dan kadar FFA biodiesel dengan penambahan antioksidan dari kulit buah pisang kepok (*musa paradisiaca linn.*), *Jurnal Kimia*, 9(2), 259–266

Setiawati, E. & Edwar, F. (2012), Teknologi pengolahan biodiesel dari minyak goreng bekas dengan teknik mikrofiltrasi dan transesterifikasi sebagai alternatif bahan bakar mesin diesel, *Riset Industri*, VI(2), 117–127

Sharma, Y.C., Singh, B. & Upadhyay, S.N. (2008) Advancements in development and characterization of biodiesel: A review, *Fuel*, 87(12), 2355–2373. https://doi.org/10.1016/j.fuel.2008.01.014

Zalfiatri, Y., Restuhadi, F. & Zulhardi, R. (2019) Karakteristik biodiesel dari minyak jelantah menggunakan katalis abu gosok dengan variasi penambahan metanol, *Chempublish Journal*, 4(1), 1–8. https://doi.org/10.22437/chp.v3i1.5774

Zhang, P. Chen, X., Leng, Y., Dong, Y., Jiang, P., & Fan, M. (2020) Biodiesel production from palm oil and methanol via zeolite derived catalyst as a phase boundary catalyst: An optimization study by using response surface methodology, *Fuel*, 272, 117680. https://doi.org/10.1016/j.fuel.2020.117680