

Biodiesel Production from Tamanu Oil (Callophyllum Inophyllum) with Immobilized Lipase Catalyst Using Activated Carbon as Matrix

Martha Aznury^{1(⋈)}, Ahmad Zikri¹, Siti Chodijah¹, Indiana Damayanti¹, and Nova Rachmadona^{2,3}

Department of Chemical Engineering, Politeknik Negeri Sriwijaya, Palembang 30128, Indonesia

martha_aznury@polsri.ac.id

Abstract. Energy needs in Indonesia until now still depend on fossil energy sources whose availability in the world is running low. Therefore, the search for alternative renewable energy must be developed, one of which is biodiesel. The raw material used in the production of biodiesel is tamanu oil, which is tamanu oil produced from pressed tamanu seeds. Tamanu seeds have an oil content of 71.4% by weight, with this amount of oil content, Tamanu seeds have enormous potential when used as raw material for biodiesel production. The purpose of this study was to determine how to produce biodiesel from tamanu oil using immobilized lipase enzymes with activated charcoal through the transesterification-esterification process. The results of the analysis in the study obtained biodiesel that meets SNI 04-7182-2006, namely the new catalyst with a variation of 4% with a yield percentage of 95.2% with a methyl ester content of 98.25%.

Keywords: Biodiesel · tamanu oil · lipase enzymes · activated charcoal

1 Introduction

Seeing the declining availability of petroleum and the increasing nominal oil consumption level, an alternative renewable energy source is needed so that national energy security can be achieved [1]. The use of biodiesel has the advantage that it can be an environmentally friendly alternative fuel because biodiesel can reduce exhaust gas emissions of CO and carbon dioxide gas CO₂ and is free of sulfur content compared to other petroleum diesel materials [2]. Raw materials that can be used in the production of biodiesel include vegetable oil derived from the seeds of the Tamanu (*Callophyllum inophyllum*) plant. Tamanu oil is a potential renewable energy resource as a biodiesel base material without having to compete with food needs. Because Tamanu oil is nonedible oil. Biodiesel produced from Tamanu are the yield of Tamanu oil are 69% [3], is

Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Padjadjaran, Jatinangor 45363, Indonesia

³ Department of Chemical Science and Engineering, Kobe University, Kobe 658-0032, Japan

relatively high compared to other types of plants jatropha curcas, palm oil,rubber seed, and coconut [4].

Synthesis biodiesel is done by transesterification process, namely by reacting vegetable oil with short chain alcohol such as methanol or ethanol to produce glycerol as a by-product. Transesterification is classified into two categories, namely by using alkaline or acid catalyst chemistry. However, there are many disadvantages of using chemical methods, some of which are high energy consumption and difficulty in transesterification reactions with high free fatty acid content while using enzyme catalysts, energy consumption is low and produces little waste.

According to [5] enzyme reactions are very sensitive to water content in raw materials. Lipase will also be inactive if it is reacted with high concentration of methanol so that in the process the addition of methanol is carried out gradually.

Lipase is one of the enzymes that is often used as a catalyst for making biodiesel, lipase is a hydrolytic enzyme which also has entarase properties so that it can be used for the production of alkyl esters with triglycerides and alcohol as raw materials [6].

The selection of energy sources for the biodiesel production process is very important considering that biodiesel itself is a new energy source so that the manufacturing process must prioritize the effectiveness of energy use. The biodiesel obtained is then compared with the biodiesel quality standard according to the Indonesian National Standard (SNI 04-7182-2006). It is hoped that this research will provide a significant contribution to science, especially in the field of energy, in terms of finding alternative energy sources and can be further developed so that better biodiesel quality can be obtained with an easier process.

2 Material and Methods

2.1 Materials

The materials used in the study included tamanu oil from extraction seed (*Callophyllum inophyllum*), lipase enzymes, activated carbon, methanol, dH₂O. Glassware equipment (Pyrex, USA).

2.2 Methods

The research method carried out was the first stage of Activated Charcoal Activation by pulverizing activated charcoal to powder form, mixing 30 g of activated charcoal powder with 100 ml of NaCl and stirring at 90o C for 2 h, then cooling the activated charcoal, then filtered and washed with distilled water, and drying activated charcoal using an oven at 105 °C for 5 h.

Then proceed with the enzyme immobilization stage, namely by dissolving 10 g of the lipase enzyme into 90 ml of phosphate buffer pH 7, adding 10 g of activated charcoal into the lipase and buffer mixture, incubating for 24 h at 30 °C, and the immobilized lipase enzyme was stored at temperature 4 °C.

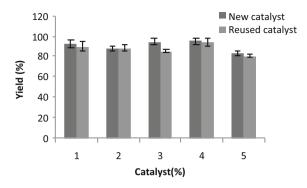


Fig. 1. Effect of percentage of new and reused catalyst on % yield

The synthesis of biodiesel, which is weighing 50 g of tamanu oil, mixing tamanu oil with methanol oil-methanol mole ratio of 1:5 into 5 Erlenmeyer pieces (addition of methanol is done gradually because if it is reacted with high concentration of methanol it will cause the lipase to become inactive), then added lipase enzyme into the erlenmeyer with variations of 1, 2, 3, 4, and 5%, the mixture was stirred at 150 rpm for 24 h at room temperature. After 24 h the resulting product was separated and then separated to obtain enzymes to be reused and the product to be carried out at the analysis stage.

2.3 Analysis Procedure

Determination of percentage of Yield was using SNI 8257:2016 method (SNI, 2016). For density, viscosity and, saponification number analysis with SNI 7182:2015 [7]. Analysis of iodine number with SNI 3961:2015 (SNI, 2015). Cetane number analysis was using ASTM D 613 [8].

Analysis of composition structure chemical in biodiesel were using Gas chromatography with ASTM D6584-13 (Shimadzu GC-2010A Series, Japan) [9].

3 Results and Discussions

3.1 Effect of Mole Percentage of New and Reused Catalysts on % Yield

Research on the effect of mole percentage of new catalyst and repeating catalyst on % yield with oil - methanol ratio 1:5 then the percentage of catalyst variation is 1%, 2%, 3%, 4% and 5%. Then from the catalyst that has been reacted on a new catalyst and a repeating catalyst, a product is obtained which will be used to determine the % biodiesel yield ratio. Based on the research data obtained, the effect of mole percentage of new catalyst and repeating catalyst on % yield can be seen in Fig. 1.

The results of the analysis of the highest % yield was found in the new catalyst with a concentration of 4%, namely 95.2% for the other yield values, which can be seen in Fig. 1. The increasing value of %yield is due to the many triglycerides contained in tamanu oil which will react with methanol which will produce more biodiesel.

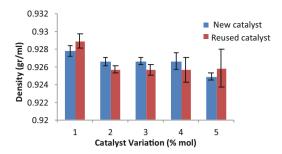


Fig. 2. Effect of percentage of new and reused catalyst on density

3.2 Density Analysis

The results of density analysis using the Triplo method on new and reused catalysts at concentrations of 1%, 2%, 3%, 4%, 5% which have been measured using a pycnometer, the graph above shows differences in density values of various concentrations. It can be seen in Fig. 2 that the highest value is found at a concentration of 1% on the repeating catalyst, the value obtained is 0.929 gr/ml, this value does not meet the SNI (0.850–0.890 gr/ml) density of the oil that has been determined. And the variation of 1, 2, 3, 4, and 5% catalyst is not much different. This is influenced because the mass of each sample is different, but the sample may be mixed with water vapor during the density test process by transferring the sample into the pycnometer because this sample is a material that is affected by humidity and storage. Density can change depending on pressure or temperature.

3.3 Viscosity Analysis

Viscosity testing is carried out using a digital viscometer in the laboratory, which aims to determine the viscosity value of the resulting sample shown in Fig. 3. After analyzing the results, the higher the viscosity value of the oil, the higher the viscosity value obtained, the highest value obtained on the new catalyst, namely at a concentration of 1% with a value of 6.33 Cst. Things that affect the value of viscosity are temperature and solution concentration, the higher the temperature, the viscosity will decrease and vice versa. The highest standard deviation value is at a concentration of 4% in the new catalyst.

3.4 Saponification Analysis

The saponification number aims to determine the value of the saponification number in biodiesel, the determination of the saponification number is carried out through acidimetric titration which is then calculated based on the formula for determining the saponification number. Shows the results of the analysis of saponification numbers, the method used in this study is the Triplo method with new and reused catalysts at a concentration of 1, 2, 3, 4%,5% presented in Fig. 4. It is known that the Indonesian National Standard is 168.02 mg KOH/gr. In the analysis that has been carried out, the highest value of the saponification number is found in the 1% repetition catalyst, which is 189.25 mg

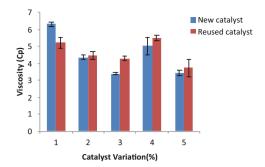


Fig. 3. Effect of percentage of new and reused catalyst on viscosity

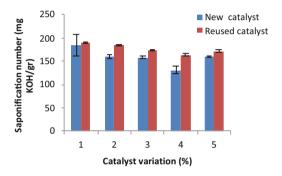


Fig. 4. Effect of percentage of new and reused catalyst on safonification number

KOH/gr. And in the new catalyst variations of 2%,3%,4%,5% and 4% can be seen in the graph above, the repeating catalyst is not included in the SNI (168.02 mg KOH/gr) also temperature.

3.5 Iodine Analysis

The iodine number can express the degree of unsaturation of the oil or fat. The greater the iodine number, the higher the degree of unsaturation. In this iodine titration analysis, the highest iodine number was found at 2% in the repeating catalyst, namely 83.24 mg/g, this value has met the SNI, which is a maximum of 115 mg/g shown in Fig. 5. In the iodine titration process, 100 ml of distilled water is heated, which causes the bonds in the constituent fatty acids in biodiesel to be degraded by temperature so that the iodine number in biodiesel is low. On the other hand, the increase in the iodine number as the catalyst concentration increases because the fatty acid bonds are not completely degraded during the heating process, this causes the iodine number to increase.

3.6 Analysis of Cetane Numbers

The cetane number is the percentage volume of cetane in its mixture with alphamethyl naphthalene, an aromatic hydrocarbon compound that has a large ignition delay, which

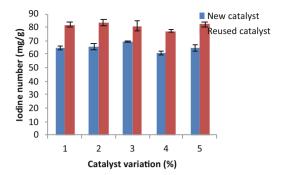


Fig. 5. Effect of percentage of new and reused catalyst on iodine number

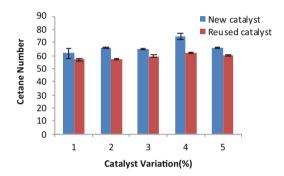


Fig. 6. Effect of percentage of new and reused catalyst on cetane number

has the same quality as diesel fuel [10]. The higher the cetane number, the faster the combustion the better the thermodynamic efficiency. The results of the analysis show that the characteristics of the experimental biodiesel cetane number have met the Quality Standards for SNI biodiesel is 51 shown in Fig. 6. From the results of the analysis of the cetane number, the cetane number value at a variation of 4% in the new catalyst obtained the highest cetane number, which is 67.5 [11], has a significant effect on the short time required between the fuel used. Injected with initiation so as to cause a good start and a smooth sound on the engine [12].

3.7 GC-MS Analisa Analysis

In this study using the Esterification Reaction, esterification is the process of reacting between methanol and free fatty acids (FFA) contained in the oil. This Gas Chromatographic Analysis aims to determine the value of Methyl Ester contained in the oil produced from the Esterification process. The results of biodiesel synthesis were identified qualitatively using Gas Chromatography to determine the composition of methyl esters in biodiesel and FFA. In this analysis, it is stated that biodiesel is a methyl ester. The result of methyl ester obtained is 98.25%, the value is in accordance with SNI, namely 96.50%. The greater the composition of biodiesel in a mixture with petrodiesel, the less gas emissions produced.

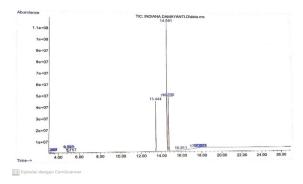


Fig. 7. Result of gas chromatography analysis

Tamanu Oil as raw material which is reacted with alcohol compounds such as methanol. The raw material contains triglyceride chains which can be simplified into monoglyceride Methyl Esters chains with the help of lipase enzyme catalysts. These methyl esters are known as pure biodiesel or commonly known as Fatty Acid Methyl Esters (FAME) [13] The graph of the GC-MS test results can be seen in Fig. 7.

4 Conclusion

Based on the research that has been done, it can be concluded that biodiesel based on Tamanu Oil uses an immobilized lipase enzyme with activated charcoal, the quality and quantity results obtained from biodiesel analysis in cetane number analysis and %yield analysis, the highest value of cetane number is in the new catalyst concentration. 4% with the addition of the lipase enzyme as much as 2 g/ml is 74.28 and for the % yield value, the highest value is obtained at the concentration of 4% of the new catalyst, which is 95.2% with the addition of the lipase enzyme 2gr/ml. The methyl ester content obtained in the gas chromatography analysis was 98.25% on a new catalyst with a concentration of 4%. This value has met the Indonesian National Standard SNI 04-7182-2006.

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References

- Aznury M, Zikri A, Syakdani A, Erlinawati (2020) Catalytic cracking process of waste cooking oils using iron (Fe) catalysts to produce biofuel. IOP Conference Series: Materials Science and Engineering 823. https://doi.org/10.1088/1757-899X/823/1/012025
- Zikri A, Aznury M (2020) Green diesel production from Crude Palm Oil (CPO) using catalytic hydrogenation method. IOP Conference Series: Materials Science and Engineering 823. https://doi.org/10.1088/1757-899X/823/1/012026

- 3. Adenuga AA, Oyekunle JAO, Idowu OO (2021) Pathway to reduce free fatty acid formation in Calophyllum inophyllum kernel oil: A renewable feedstock for biodiesel production. Journal of Cleaner Production 316:128222. https://doi.org/10.1016/j.jclepro.2021.128222
- Atabani AE, César ADS (2014) Calophyllum inophyllum L. A prospective non-edible biodiesel feedstock. Study of biodiesel production, properties, fatty acid composition, blending and engine performance. Renewable and Sustainable Energy Reviews 37:644–655. https:// doi.org/10.1016/j.rser.2014.05.037
- 5. Zenevicz MCP, Jacques A, de Oliveira D, et al (2017) A two-step enzymatic strategy to produce ethyl esters using frying oil as substrate. Industrial Crops and Products 108:52–55. https://doi.org/10.1016/j.indcrop.2017.06.018
- Rachmadona N, Harada Y, Amoah J, et al (2022) Integrated bioconversion process for biodiesel production utilizing waste from the palm oil industry. Journal of Environmental Chemical Engineering 10:107550. https://doi.org/10.1016/j.jece.2022.107550
- 7. Standar Nasional Indonesia (SNI) 7182 (2015) Sni Biodiesel 2015. Sni 88
- D613-05 A (2000) Standard Test Method for CetaneNumber of Diesel Fuel Oil. ASTM International 14:1–17
- 9. Provided S, No ISO, Licensee IHS (2013) International Standard Iso. 2010:
- Hadiantoro S, Moentamaria D, Syarwani M (2018) Efektifitas Penggunaan Co immobilized -Lipase pada Reaksi Esterifikasi Asam Lemak Hasil Hidrolisis Minyak Kelapa. Jurnal Teknik Kimia dan Lingkungan 2:23. https://doi.org/10.33795/jtkl.v2i1.60
- Badan Standarisasi Nasional (2015) Standar Nasional Indonesia 7182:2015 Biodiesel. Badan Standarisasi Nasional 1–88
- 12. Rizwanul Fattah IM, Masjuki HH, Kalam MA, et al (2014) Experimental investigation of performance and regulated emissions of a diesel engine with Calophyllum inophyllum biodiesel blends accompanied by oxidation inhibitors. Energy Conversion and Management 83:232–240. https://doi.org/10.1016/j.enconman.2014.03.069
- 13. Amoah J, Quayson E, Hama S, et al (2017) Simultaneous conversion of free fatty acids and triglycerides to biodiesel by immobilized Aspergillus oryzae expressing Fusarium heterosporum lipase. Biotechnology Journal 12. https://doi.org/10.1002/biot.201600400

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