Biodiesel Production from Waste Cooking Oil with Immobilized Lipase Catalyst Using Activated Carbon as Matrix

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Abstract. Lipase is a biocatalyst that hydrolyses triglycerides (fats) into their component fatty acid and glycerol molecules to produce biodiesel. Due to the benign working conditions, enzymatic techniques enable straightforward biodiesel purification processes with significantly lower energy requirements and high-rate conversions. The effect of enzyme-immobilized concentration on biodiesel production was investigated in this study. The quality of biodiesel was evaluated based on SNI 7182:2015 standard. The result indicated that 1.9% (w/w) lipase concentration was the optimum condition for producing biodiesel. In conclusion, the immobilized lipase using activated carbon can be used for biodiesel production from waste cooking oil for sustainable green chemistry.

Keywords: Biodiesel · Immobilized Lipase · Waste Cooking Oil · Enzymatic Reaction

1 Introduction

The need for energy will keep rising as technology and population growth accelerate. If the output is estimated to be 852 thousand barrels per day, Indonesia's oil reserves will only last 12 years at their current level at the end of 2021 [1]. One of the potential energy issues that can be used to replace diesel/diesel fuel is biodiesel. An alternative fuel derived from renewable natural resources, such as plant and animal oils, is called biodiesel oil [2]. Alternative fuels that can be used in place of diesel fuel are sourced from renewable resources. The most common type of oil consumed by Indonesians is diesel fuel.

Biodiesel is more environmentally friendly because biodiesel produces exhaust gas emissions that are significantly better than those of diesel or diesel, including the absence of sulphur, low smoke number, and cetane number ranging from 57 to 62 so that combustion efficiency is better burned entirely [3]. Cooking oil is manufactured on a big

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scale from palm oil and is derived from plant or animal fats that are purified in liquid form at room temperature and are typically used for frying food [4]. Because cooking oil contains many unsaturated fatty acids, it is easily destroyed during the deep-frying process since the oil is heated continually at high temperatures. The unsaturated layer is typically removed twice during the filtering process of palm oil, increasing the amount of unsaturated fatty acids.

One of the fundamental necessities met by the Indonesian people is cooking oil, with annual consumption levels exceeding 2.5 million tons or more than 12 kg per person. In Indonesia, palm oil is used to make more than 70% of the most common cooking oil [5]. Because palm cooking oil has undergone numerous treatments to remove contaminants, fatty acids, and solid fats, using it as biodiesel is technically more profitable. One alternative to the usage of wasted cooking oil has to be researched, specifically the process of turning used cooking oil into biodiesel [6]. Because cooking oil is utilized at every socioeconomic level, from the lowest to the highest, and in both homes and hotels, it is one of the potential raw materials. Cooking oil is readily degraded during frying because it contains many unsaturated fatty acids and is heated continually at high temperatures [7]. The nature or composition of the biodiesel produced from leftover cooking oil is the issue. Because leftover cooking oil has a high iodine number, viscosity, and flash point than diesel fuel, it must have its two qualities decreased to make biodiesel suitable for use as fuel [8].

Using triglycerides and alcohol as starting materials, lipase, a hydrolytic enzyme with esterase abilities can be employed to generate alkyl esters [9]. On the other hand, the lipase enzyme is frequently utilized as a catalyst in biodiesel production. Because lipase possesses a heterogeneous catalyst, it may selectively focus the reaction on the product, making separation simple. Even though they have benefits, enzymes are expensive and cannot be used repeatedly since they are soluble in the liquid medium, which is a drawback of enzyme catalysts [10]. However, the enzyme immobilization method can solve this issue. Enzyme immobilization (support) is the process of combining an enzyme with a solid so that it can be utilized repeatedly and continuously [11].

An amorphous substance known as "activated charcoal" is created from carbon or charcoal containing materials that have undergone particular processing to increase their adsorption capacity. Depending on the pore capacity and surface area, activated charcoal has the ability to adsorb some gases and chemicals selectively [12]. Activated charcoal absorbs 25 to 100% of its weight in various substances. All carbon-containing substances, whether from plants, animals, or mining resources, can be converted into activated charcoal [13]. Various types of wood, sawdust, rice husks, coconut shells, animal bones and shells, coal, and other materials are among them [14].

2 Material and Methods

2.1 Materials

Waste cooking oil was palm oil that was used for cooking. FFA content has been analyzed through this experiment. Eversa® Transform 2.0 with the lipase activity, 100 LCLU-SL/g, was purchased from Nanobio Laboratory (Indonesia). All chemical reagents were purchased from Sumber Kimia (Indonesia).

2.2 Immobilization of Lipase on Activated Carbon

Crushed activated charcoal is used to make powder. Then, 100 cc of 3 M NaCl and 30 g of activated charcoal were combined and agitated at 90 °C for 2 h. The activated charcoal is then filtered, cleaned with distilled water, and allowed to cool before being dried for 2 h at 105 °C. Additionally, 30 g of the enzyme was immobilized in 270 mL of pH 7 phosphate buffer after being dissolved in activated charcoal. Following thorough mixing, 30 mL of phosphate buffer and 30 g of activated charcoal powder were added. The procedure takes 6 h to complete.

2.3 Biodiesel Production from Waste Cooking Oil Using Immobilized Lipase

Transesterification reaction was carried out in a 250 mL Erlenmeyer. Methanol is combined with 50 g of waste cooking oil, with oil to methanol mole ratio of 1:5. The reaction was then supplemented with immobilized lipase at concentrations of 1%, 2%, 3%, 4%, and 5%, and shaken at 40 °C for 24 h while being swirled at a speed of 300–450 rpm.

2.4 Analytical Method

Various tests and analyses were conducted to describe the biodiesel's physic-chemical characteristics. According to SNI 7182: 2015 standards, the viscosity, density, iodine, saponification, cetane number, and component composition of biodiesel are tested and analyzed.

3 Results and Discussions

3.1 Biodiesel Production

This study used activated charcoal and an immobilized lipase enzyme catalyst to create biodiesel products from wasted cooking oil. Two layers, one containing immobilized enzymes and the other containing biodiesel products, were created following the 24 h production of biodiesel employing an immobilized lipase enzyme catalyst and activated charcoal. Biodiesel was then used to separate the immobilized enzyme created once more after the procedure. The biodiesel's weight after being separated from the catalyst and glycerol is measured. After that, the computation is done to get the yield percentage.

Figure 1 shows that the larger the catalyst percentage, the lower the yield for biodiesel as an enzyme immobilized catalyst and activated charcoal, with 1% immobilized enzyme producing the maximum yield. Similar outcomes were also attained by [15]. They found that excessive substrate concentrations can produce substrate inhibitors and that too much catalyst can make the reactant mixture too viscous and delay churning. Higher levels may increase glycerol production, reducing the amount of biodiesel produced [16]. The average yield generated when using 1% catalyst is 80.65%. This amount exceeds the 65.8% yield of used cooking oil biodiesel produced with an H_2SO_4 catalyst [17].

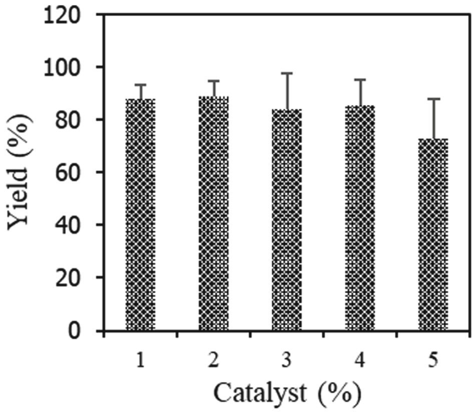

Fig. 1. Biodiesel production by various weight of immobilized lipase

3.2 Characterization of Biodiesel

Biodiesel was characterized according to SNI 7182:2015 standard. Table 1 depicts the fuel properties of optimized produced biodiesel. The result showed that all of the properties were in the acceptable range of the standard. The density of the biodiesel produced was 870–890 kg/m^3 from the range of standard in 850–890 kg/m^3. The high density of biodiesel had an impact on decreasing the combustion ability in the combustion process that occurred. In the terms of viscosity, saponification, iodine value, and cetane number, it showed that using 1% of immobilized enzyme, the parameter characterization was acceptable according to SNI 7182:2015 standard.

The fatty acid methyl ester content of biodiesel was characterized by using GC-MS. This analysis used to determine the content of the chemical compounds. Table 2 shows the synthesis composition of biodiesel in this study. Octadecadienoic acid and 9-octadecanoic acid are the highest composition of methyl ester for this biodiesel with the percentage of content, 29.94% and 20.67%. Busyairi et al. [18], also found that octadecadienoic acid methyl ester with levels of 29.65 and octadecanoic acid methyl ester 28.11%.

The fatty acid methyl ester is one of the important factors that determine the suitability of raw materials for use in fuel production [19]. The results in Table 4.4 show that most ester compounds are monounsaturated fatty compounds, namely methyl octadecanoic. Methyl octadecanoic is a very good compound and tends to be more stable for biodiesel because methyl octadecanoic does not have much affinity for oxygen which can cause polymerization and peroxidation [20].

3.3 Reutilization of the Immobilized Lipase for Biodiesel Synthesis

This study conducted a strategy for reusing the immobilized lipase after transesterification. After the reaction was done, biodiesel and the immobilized lipase was separated and was reused to the next synthesis. The reaction was conducted as the same experimental methods as the new catalyst. The result showed that the optimum yield for the reused

Table 1. Characterization of biodiesel production

Catalyst (% w/w)	Density (kg/m^3)	Viscosity (mm^2/s)	Yield (%)	Saponification value (mg KOH/g oil)	Iodine value (g I_2/100 g lipid)	Cetane number
1	912.53	61.04	88.00	116.89	54.99	813.04
2	913.87	48.18	89.00	108.75	65.14	819.94
3	920.09	38.24	83.73	400.22	64.80	827.86
4	891.67	38.60	85.47	950.05	60.32	885.11
5	922.00	39.12	72.53	701.32	53.21	112.41

Table 2. GC-MS analysis result

No	Peak Area (%)	Component	Molecule
1	0.63	1,1- Dimetil-2-P propenyl Methyl Ester	$C_9{:}_{16}$
2	0.08	Octanoic acid – Methyl Ester	$C_9H_{18}O_2$
3	0.06	Decanoic acid – Methyl Ester	$C_{11}H_{22}$
4	0.80	Undecanoic acid 10 – Methyl Ester	$C_{11}H_{22}$
5	0.63	1,2 Propenyl Methyl Ester	$C_4H_{10}O$
6	0.07	Pentadecanoic Acid, 14 Methyl Ester	$C_{16}{:}_3$
7	0.56	9- Hexadeconic Acid. Methyl Ester	$C_{18}H_{36}O$
8	12.27	Pentadecanoic Acid, 14 Methyl Ester	$C_{16}{:}_3$
9	20.67	9- 0ctadecanoic Acid, Methyl Ester	$C_{19}H_{36}O$
10	0.47	Eicosanoic Acid, Methyl Ester	$C_{20}H_{40}O_2$
11	29.94	Octadecadienoic Acid, Methyl Ester	$C_{18}{:}_3$
12	12.25	Hexadecanoic acid Methyl Ester	$C_{17}{:}_3$
13	0.14	Heptadecanoic Acid, Metil Ester	$C_{17}H_{34}O$
14	0.14	Heksadesimal acid Methyl Ester	$C_{18}H_{36}O$
15	0.14	Hexadecanoic 14 – Methyl Ester	$C_{18}H_{36}O$
16	19.31	Heksadesimal acid, Methyl Ester	$C_{17}{:}_3$
Total	**98.02**		

immobilized lipase was 82.51% with 3% catalysts as presented in Fig. 2. It is different from the immobilized lipase that used for the first time which 1% of catalyst was enough for the reaction. This result indicated that the activity after first batch reaction reduced so that the biodiesel yield decreased after recycling.

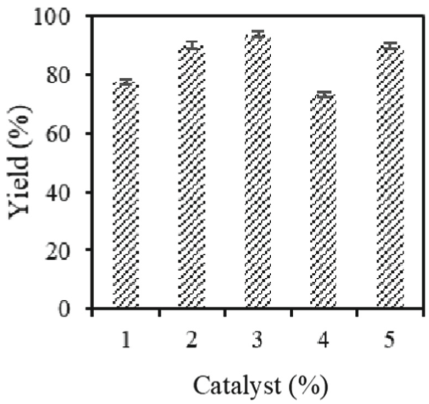

Fig. 2. Reutilization of the immobilized lipase for biodiesel synthesis

4 Conclusion

Immobilized lipase enzyme catalyst variations have an impact on the properties of the resulting biodiesel. Except for the density characteristic of biodiesel with a catalyst variation of 1%, 2%, and 3% because the density is too high, all manufactured biodiesel complies with SNI 7182:2015. This study found that a 5% catalyst variation provided the best biodiesel because all of its attributes complied with SNI 7182:2015, and the highest cetane number was created. Octadecanoic acid methyl ester and octadecanoic acid methyl ester, with concentrations of 29.65 and 28.11%, respectively, make up most of the biodiesel's chemical makeup.

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