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RESEARCH ARTICLE

Optimization of sustainable biodiesel production from waste cooking oil using heterogeneous alkali catalyst

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OBJECTIVES The increasing world population, rapid industrialization, urbanization, and economic growth have led to a continuous increase in the consumption of fossil fuels to meet the ever-increasing demand for energy. Continuous emissions from burning fossil fuels will create a need to find appropriate and sustainable substitutes for fossil fuels. Biodiesel is the right alternative solution for diesel engines because it is renewable, non-toxic, and environmentally friendly. Waste cooking oil (WCO) from the food, non-food, restaurant, and household sectors is produced on a large scale in every country and can contribute to environmental pollution if proper disposal systems are not applied. Instead of throwing it, landfills Environmental pollution can be minimized by recycling WCO. METHODS This study evaluates the potential of using WCO to produce biodiesel using zeolite synthesized from fly ash as a heterogeneous alkali catalyst through a transesterification reaction. The reactor in this study used a 1,000 ml three-necked boiling flask equipped with a condenser, cooling tank, and pump. Stirring and heating during the process of biodiesel production using a magnetic stirrer and a hot plate. The thermometer was used to measure the reaction temperature. Optimization of biodiesel production from zeolite catalyst synthesized from fly ash based on variations in the ratio of methanol:oil (8:1; 10:1; 12:1; and 14:1), catalyst loading (1, 2, 3, and 4% weight), and temperature (45 °C, 55 °C, and 65 °C). RESULTS Zeolite from fly ash produces biodiesel with a yield of 91.67% with optimum operating conditions reaction time of 60 minutes, methanol oil ratio of 8:1, operating temperature 55 °C, and the amount of catalyst 1% by weight. CONCLUSIONS This experiment confirmed the possibility of utilizing fly ash waste for the application of catalysts in biodiesel production.

KEYWORDS alkali; biodiesel; heterogeneous alkali catalyst; transesterification; waste cooking oil

1. INTRODUCTION

The depletion of fossil fuel reserves globally and environmental pollution from combustion are the main driving factors for researching alternative fuels derived from biomass (Malani et al. 2019). Sustainbale energy such as biodiesel is alternative energy to overcome the limitations of non-renewable energy sources (Fadhil et al. 2016). Biodiesel is one of the most environmentally friendly energy fuels because it is a non-toxic fuel, does not pollute the environment, and has fewer gas emission effects (Ullah et al. 2015).

Biodiesel is produced from the transesterification of triglycerides in vegetable oils or animal fats (Fadhil et al. 2017). Transesterification is a chemical reaction between triglycerides and alcohol in the presence of a catalyst to produce monoesters (Katabathini et al. 2007). Aspects that affect the transesterification reaction are the type of catalyst (base or acid), alcohol/vegetable oil molar ratio, temperature, reactant purity (especially water content), and free fatty acid content (Noureddini and Zhu 1997). The content of biodiesel is an insignificant aromatic compound and has more than 10% of the inherent oxygen fraction, which supports complete combustion (Parida et al. 2016). The selection of the right raw material for biodiesel production is one of the important factors to reduce the cost of biodiesel production. Recently, biodiesel is produced from edible oil, such as soybean oil, palm oil, corn oil, etc. (Chuah et al. 2017). However, the use of edible oil for biodiesel production is less profitable because of its competitiveness with the food sector (Naylor and Higgins 2017). The cost of raw materials using edible oil, on the other hand, accounts for about 80% of the final cost of biodiesel produced.

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The use of low-priced oils such as waste cooking oil (WCO) will significantly reduce the cost of the final product (Fadhil et al. 2017).

Cooking oil consumption is high in Indonesia. Technically, the potential for WCO in Indonesia reaches 3,072,280 kL/year. This potential is obtained from restaurants, hotels, schools, urban hospitals, and urban households (Kharina et al. 2018). Similarly, a study conducted by Goembira and Ihsan (2018) in Padang City, West Sumatera, approximately 26,060.79 liters of WCO are produced per week. Most of the WCO comes from households, hotels, and street vendors, respectively. Environmental pollution is minimized by recycling WCO rather than throwing it into landfills (Milano et al. 2018). Therefore, researchers are trying to utilize waste such as used cooking oil in the manufacture of biodiesel. However, the high content of free fatty acids (FFA) in WCO is a challenge for scientists (Putra et al. 2018).

On a commercial scale, biodiesel is still produced using homogeneous base catalysts such as NaOH and KOH (Ali and Fadhil 2013). The purification step using a catalyst is expensive considering the complexity of the process of separating the catalyst from biodiesel (Wang et al. 2017). The catalyst cannot be recycled and is generally disposed of in a sewage system and have the potential to cause environmental problems (Chen et al. 2015). This can be overcome by using heterogeneous alkali catalysts in the transesterification reaction of biodiesel production which is currently being developed by many researchers (Chen et al. 2015; Nata et al. 2017; Wang et al. 2017). Torres-Rodríguez et al. (2016) have demonstrated the transesterification of mixed soybean oil using sodium cesium zirconate impregnation. A High yield of biodiesel is achieved. However, the developed catalysts still use valuable commercial raw materials. Therefore, waste materials are deemed necessary to be studied further In this study, the production of biodiesel through the transesterification process from WCO was carried out using a heterogeneous alkali catalyst developed from fly ash that synthesized by the hydrothermal method. The effect of methanol:oil ratio, catalyst loading, and reaction temperature on biodiesel yield was further observed.

2. RESEARCH-METHODOLOGY

2.1 Materials

The WCO in this study was obtained from Sahabat Alam Waste Bank, Pariaman, Indonesia. Most of the WCO collected by the Sahabat Alam Waste Bank comes from household frying waste. A heterogeneous alkali catalyst was synthesized from fly ash with hydrothermal method. The methanol, sodium hydroxide, and ethanol used are analytical grade that supplied from CV. Vahana Scientific.

2.2 Procedures

2.2.1 FFA test

Before the transesterification process, free fatty acids (FFA) were determined. This can determine the suitability of using heterogeneous catalysts to catalyze oil stocks with high free fatty acid content in biodiesel production, and their tolerance level (Daramola et al. 2015). The FFA content in WCO was assessed and determined using Equation (1) as described by Daramola et al. (2015).

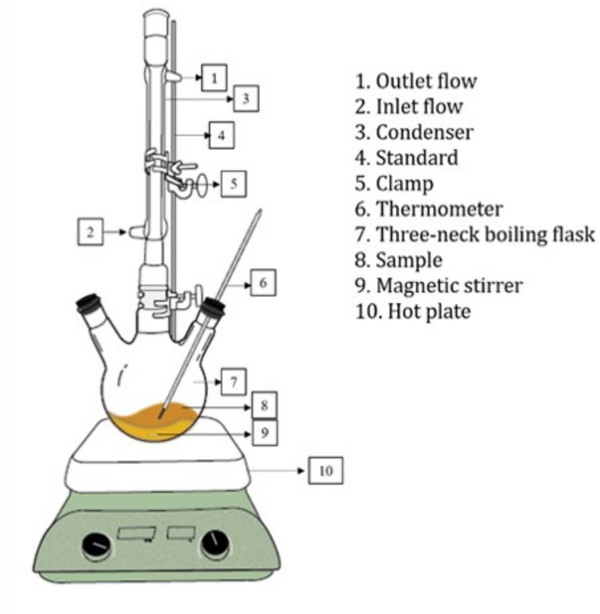

FIGURE 1. An experimental set of biodiesel production.

$$\%FFA = \frac{[(V-b) \times 28.2N]}{W} \quad (1)$$

If FFA > 2%, then the esterification process is carried out until the raw material has FFA < 2%. If FFA < 2% it can carry out the transesterification process.

2.2.2 Experimental setup

The 1,000 ml three-neck boiling flask equipped with a vertical condenser and placed on a hot plate with a magnetic stirrer was used for biodiesel conversion. The cooking oil is filtered to remove suspended food particles. 30 mL of WCO was placed in a three-neck boiling flask. An experimental setup is shown in Figure 1. The methanol is poured into the flask with a methanol:oil ratio (8:1; 10:1; 12:1; and 14:1, respectively). Heterogeneous alkali catalysts (1%; 2%; 3% and 4% w/w) were added to the batch reactor. The transesterification process was carried out for 60 minutes at various temperatures (45 °C, 55 °C, and 65 °C). The experiments were carried out in parallel batches. After the reaction, the catalyst was separated from the solution by filtration. The solution filtrate was left in the funnel overnight to separate the crude biodiesel and glycerol mixture. Due to its lighter density than glycerol, the biodiesel is in the top layer and the glycerol is in the bottom layer. Crude biodiesel was washed using warm water (55 °C) to remove impurities. Furthermore, drying was carried out to evaporate the water in the biodiesel at a temperature of 105 °C.

2.2.3 Characterization of biodiesel products

Biodiesel characterization was analyzed based on its quality and quantity. The biodiesel quality analyzed was alkyl esters content, free glycerol, total glycerol, density (40 °C), color, and acid value. The biodiesel quantity was analyzed based on the percentage of volume yield which was calculated as equation (2) (Singh et al. 2020).

$$\%\text{volume yield} = \frac{\text{Volume of product}}{\text{Volume of feed}} \times 100\% \quad (2)$$

The product volume was the volume of biodiesel produced after the transesterification process and the feed volume was volume of WCO used.

3. RESULT AND DISCUSSION

3.1 FFA sample

The collected WCO has FFA content of 0.7%. FFA content in WCO is lower than 2%. Therefore, the experiment is transesterification.

3.2 effect of temperature

The temperature has a significant effect on biodiesel production. The result of reaction temperature on biodiesel yield (%) using a heterogeneous alkali catalyst was carried out at a reaction time of 60 minutes; methanol:oil ratio 8:1; catalyst weight 1% and stirring speed 300 rpm. The temperature variations performed were 45 °C, 55 °C, and 65 °C. It was found that with increasing temperatures at 45 °C and 55 °C the biodiesel yield (%) increased from 89.00% to 91.67% as shown in Figure 2.

Furthermore, the biodiesel yield decreased to 87.67% at the reaction temperature of 65 °C. In a study conducted by Sahu (2021), the yield of biodiesel produced decreased in the reaction temperature range of 65 °C – 70 °C. Sahu (2021) revealed that reactions at high temperatures can produce unfavorable elements such as free fatty acids which lead to reduced product formation. Similarly, in the study conducted by Erchamo et al. (2021), biodiesel yield decreased in the reaction temperature range of 60 °C to 65 °C. An increase in temperature above 60 °C results in a decrease in biodiesel yield because the methanol evaporates during operation (methanol boiling point 64.96 °C) which causes some of the methanol to be in the gas phase circulating in reflux, resulting decrease in the amount of methanol in the reaction (Yang et al. 2018). Therefore, a temperature 55 °C of would be more efficient for the transesterification reaction in this study.

3.3 Effect of methanol:oil ratio

The effect of methanol on biodiesel yield was taken at a reaction temperature of 55 °C; reaction time 60 minutes; methanol oil ratio 8:1; catalyst weight 1%; and a stirring speed of 300 rpm. The methanol:oil variations performed were 8:1; 10:1; 12:1; and 14:1. The results are presented in Figure 3.

It can be seen in Figure 2 that the yield (%) decreases with increasing the ratio of methanol to oil. When the methanol:oil ratio is 8:1; 10:1; 12:1; and 14:1 yield percentages were 91.67%, 87.67%, 86.33% and 85.33%. In a study conducted by Sahu (2021), biodiesel yields decreased at methanol:oil ratios 18:1 to 21:1. Research conducted by Erchamo et al. (2021) revealed that there was a decrease in biodiesel yield at methanol:oil ratio of 12:1 to 16:1. The decrease in biodiesel yield can be attributed to the high methanol:oil molar ratio which interferes with the separation of glycerin because there is an increase in the solubility of glycerol in excess methanol (Erchamo et al. 2021). Excess methanol is beneficial for conversion of triglycerides to monoglycerides. However, monoglycerides significantly affect the solubility of glycerol in FAME, which is naturally immiscible causes glycerolysis of FAME, and inhibits triglyceride conversion. A similar report was submitted on 89.81% biodiesel production with methanol:oil ratio of 15:1, using Hevea brasiliensis oil and a flamboyant pod derived carbon heterogeneous catalyst (Dhawane et al. 2016), using linseed oil with methanol:oil ratio 9.48:1 achieved biodiesel yield of 98.08% (Hashemzadeh Gargari and Sadrameli 2018) and methanol:oil ratio 9.4:1 achieved biodiesel yield of 98.26% with a catalyst supported by potassium hydroxide (Rabie et al. 2019).

TABLE 1. Comparison of physicochemical properties of biodiesel with SNI 7182:2015 standards.

No	Properties	Standards	Results	Units
1	Alkyl Esters content	96.5, min	99,64	%
2	Free glycerol	0.020, max	0,012	% wt
3	Total glycerol	0.240, max	0,1521	% wt
4	Density (40 °C)	860-890	902,8	Kg/m^3
5	Color	3, max	L 2,5	-
6	Acid value	0.50, max	0,0094	mg KOH/g

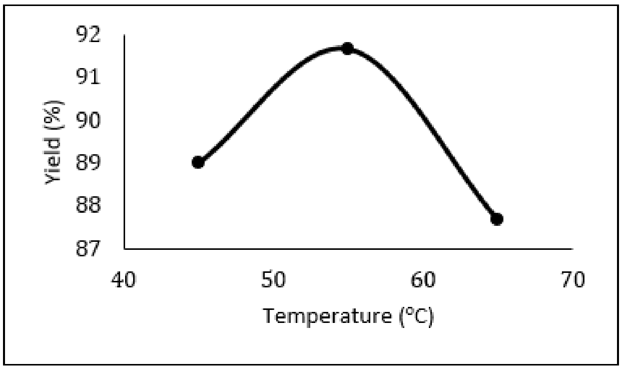

FIGURE 2. Effect of temperature on biodiesel yield.

3.4 Effect of catalyst loading

The amount of catalyst used is one of the key factors in the transesterification reaction. The effect of catalyst weight was carried out at the reaction temperature of 55 °C; reaction time 60 minutes; methanol:oil ratio of 8:1 and stirring speed of 300

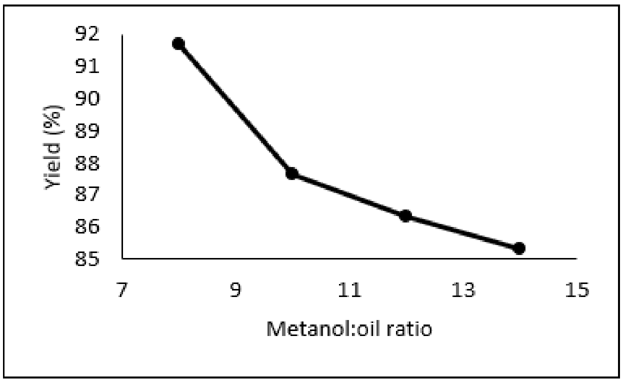

FIGURE 3. Effect of methanol:oil ratio on biodiesel yield.

TABLE 2. Comparative study of biodiesel production from WCO with heterogeneous alkali catalyst.

No	Catalyst used	Methanol:oil (mol:mol)	Catalyst Loading (%)	Reaction Time (min)	Reaction Temperature (°C)	Yield (%)	References
1	Nanocatalyst (CZO)	8:1	1.2	50	55	97.71	Gurunathan and Ravi (2015)
2	Eggshell	12:1	1.5	120	65	96.00	Tan et al. (2015)
3	Zeolite	8:1	1	60	55	91.67	This study
4	CaO/SiO_2	14:1	8	90	60	91.00	Putra et al. (2018)
5	CaO/KOH	10:1	3	30	50	88.00	Joshi et al. (2017)
6	Sulfated Alumina	12:1	1	60	60	86.67	Ulfah et al. (2019)
7	Calcined scallop shell	6:1	5	120	65	86.00	Sirisomboonchai et al. (2015)
8	KBr/CaO	12:1	3	180	65	82.48	Mahesh et al. (2015)
9	Ash palm oil	18:1	5.35	30	60	71.70	Chin et al. (2009)
10	CaO	12:1	0.85	60	60	66.00	Kouzu et al. (2008)

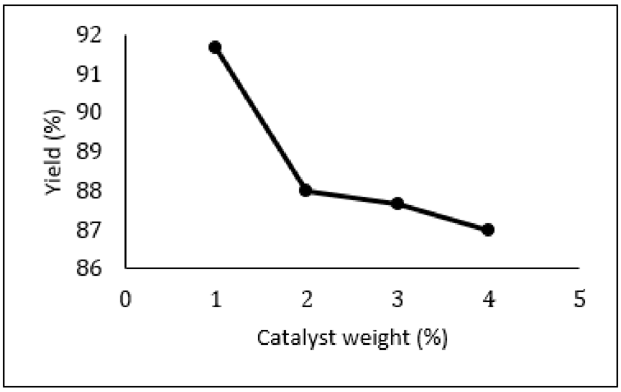

FIGURE 4. Effect of catalyst weight on biodiesel yield.

rpm. The experiments were carried out at different catalyst loading variations were 1%, 2%, 3%, and 4% by weight, which is shown in Figure 4.

The experimental results showed that the biodiesel yield at 1%, 2%, 3%, and 4% catalyst weight was 91.67%, 88.00%, 87.67%, and 87.00% respectively. In a study conducted by Erchamo et al. (2021), the yield of biodiesel produced decreased in experiments with a catalyst weight of 2.5% to 4%. Erchamo et al. (2021) revealed that the decrease in biodiesel yield after reaching the optimum value could be caused by a larger amount of catalyst exceeding the average value making the transesterification reaction product more sticky which usually inhibits the mass transfer process in oil, alcohol, and catalyst. A slight amount of catalyst loading is not sufficient to convert triglycerides into fatty acid esters and an excess amount of catalyst will turn the product separations (Kashyap et al. 2019). Therefore, the optimum amount of 1% by weight of heterogeneous catalyst would be more efficient for the transesterification reaction in this study. Another study showed that the 96.47% biodiesel yield was achieved by using 6 wt% heterogeneous catalysts with CaO/MgO (Rabie et al. 2019); the use of CaO derived from the shell of Chicoreus Brunneus as a catalyst in the amount of 0.5 wt% resulted in biodiesel yield of 93.5% (calcined 1,100 °C) (Mazaheri et al. 2018), 95.23% biodiesel yield with 2 wt% of papaya Carica stems (Gohain et al. 2020), and 99.16% biodiesel yield with 4 wt% of banana peel catalyst (Jitjamnong et al. 2021).

3.5 Physicochemical properties of biodiesel

Biodiesel which has the highest yield in this experiment is analyzed for its characteristics. Table 1 shows the comparison of the physicochemical properties of biodiesel compared to SNI 7182:2015 standards. In general, it is observed that the biodiesel produced with heterogeneous alkali catalyst fulfills the fuel specifications of SNI 7182:2015 standards. However, the density of the biodiesel needs to be improved because it slightly higher than SNI standard limits.

3.6 Comparative study of biodiesel

The comparison of biodiesel production from WCO with heterogeneous catalysts is shown in Table 2. It can be observed that with heterogeneous nanocatalysts the maximum biodiesel conversion is 97.71% with operating conditions methanol:oil ratio 8:1; catalyst loading 1.2%; reaction temperature 55 °C; reaction time 55 minutes (Gurunathan and Ravi 2015). The reaction time is shorter than in this study and the reaction temperature is the same. Another study reported 96.00% biodiesel conversion with the eggshell catalyst at methanol:oil ratio of 12:1; catalyst loading 1.5%; reaction temperature 65 °C; reaction time 120 minutes (Tan et al. 2015). Homogeneous catalysts have disadvantages such as inconvenience for reactants separation; product and water are polluted after the chemical washing process (Hsiao et al. 2020). The current overall analysis of biodiesel production from WCO with zeolite from fly ash as a heterogeneous alkali catalyst was found to be appropriate.

4. CONCLUSIONS

Biodiesel is a promising and more attractive fuel for diesel engines due to its renewable nature and environmental benefits. The main issue to consider is the higher price of biofuels than fossil fuels. Using low-quality raw materials – which do not compete with food supply and land for food cultivation such as non-edible oils and animal fats is considered an effective way to reduce the cost of biodiesel production.

The production of biodiesel from used cooking oil using zeolite from fly ash as a heterogeneous alkali catalyst is an economical opportunity to reduce environmental impacts. For maximum results the optimal operating parameters are the reaction temperature of 55 °C, reaction time of 60 minutes, ratio of methanol to oil 8:1, catalyst used was 1% by weight, and a stirring speed of 300 rpm resulted in a biodiesel yield of 91.67%.

Overall, the development of heterogeneous alkali catalysts synthesized from waste for biodiesel production was found to be suitable and possible to be improved. Utilization of waste as raw material for making biodiesel certainly has economic potential considering that used cooking oil is still not fully utilized. The nation's economic growth depends on the long-term excess of energy from sources that are safe, affordable, and easily available.

5. NOTATION

1. V = the titrant value, ml
2. b = the blank volume, ml
3. N = concentration of titration solution, N
4. W = the weight of the oil used, g

6. ACKNOWLEDGEMENTS

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