THE EFFECTIVE PROCESS OF WASTE COOKING OIL BIODIESEL PRODUCTION BY INTERESTERIFICATION WITH REACTION USING CURCUMIN BIOCATALYST

Biodiesel is a renewable energy source that can replace fossil fuels. Used cooking oil is the most relevant biodiesel feedstock because it is a waste product. The interesterification process for biodiesel production uses a catalyst that requires separation at the end of the reaction, making it less effective. A more effective and economical biodiesel production process using waste raw materials, without the separation of by-products and catalysts, is needed to overcome this problem. Organic aromatic compound biocatalysts are environmentally friendly and relatively inexpensive. Compound biocatalysts eliminate the need for a separation process and any residue left in the biodiesel can be beneficial, as these substances act as antioxidants preventing an increase in oxidation number. In this context, curcumin is an organic aromatic compound with two aromatic groups. Therefore, this research obtains rational conditions for the interesterification reaction of waste cooking oil with curcumin biocatalyst. The operating conditions included 250 grams of waste cooking oil, the mass of curcumin biocatalyst was 0.5, 1, 1.5, 2, and 2.5%, 300 rpm stirring speed, mole ratio of methyl acetate oil was 1:6 and 1:12, reaction temperature 60°C as well as reaction time of 15, 30, 45, 60, 75, 90 and 105 minutes. The results show that the highest crude yield was obtained at 91.74% in the interesterification reaction of waste cooking oil with a curcumin biocatalyst concentration of 2.5% at a reaction time of 105 minutes and the mole ratio of oil:methyl acetate = 1:6. A density value of 0.884 g/ml and an acid number of 0.224 mg KOH/g were obtained which met SNI 7182-2015 under these conditions. In addition, the outcome of the GC-MS analysis shows that the dominant methyl ester component formed was hexadecanoic acid, 1-methylethyl ester

Keywords: interesterification, biodiesel, curcumin biocatalyst, crude yield, methyl ester

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1. Introduction

Biodiesel is presented as a promising alternative to normal diesel both now and in the future as fossil fuel supplies continue to decrease. However, its production still faces challenges due to the limitation of materials used and the long process stages. The transesterification procedure with a base catalyst is a common method for forming biodiesel, where the glycerol and catalyst by-products are separated at the end of the reaction [1]. Following this discussion, the interesterification process is more efficient because the triacetin by-product at a certain concentration can improve the quality of biodiesel [2]. When using acid or base catalyst, the process requires additional steps for neutralization and catalyst separation [3, 4]. Consequently, applying a heterogeneous catalyst comprises a longer procedure, including preparation of the substance before use and separation after the reaction [5]. The interesterification process with heterogeneous catalysts requires a long time to produce high yields [6, 7]. The soybean oil interesterification process with CaO catalyst obtained a low yield at 2 hours [8]. The trioleate interesterification process with iron sulfate catalyst obtained a yield of 75% at a reaction time of 24 hours [9]. In the interesterification re-

action of palm oil and methyl acetate, KOH catalyst needs be neutralized with acid and separated from the reaction products [10].

Research on biodiesel production using organic aromatic compounds as biocatalysts is uncommon. In addition to being environmentally friendly and relatively cheap, the use of aromatic compound biocatalysts does not require complicated as well as lengthy pretreatment such as homogeneous and heterogeneous catalysts. Organic aromatic compounds included in biodiesel remain safe because the substances will act as antioxidants preventing damage to biodiesel [11]. Previous research relied on packaged palm oil as the raw material in the interesterification process of palm oil using cajuput oil-based biocatalyst, which may compete with food industry demands. However, the current research adopted cooking oil that has been used, offering a more sustainable and less commercially competitive alternative [12]. The main content of eucalyptus oil that acts as a biocatalyst is 1, 8-cineol, an aromatic compound with one aromatic group. In this context, used cooking oil is considered a raw material, serving as a waste-based input.

Curcumin is an active compound contained in turmeric. Curcumin is a yellow-orange solid, insoluble in water, has a melting point of around 183°C and a molecular weight

of 368.39 g/mol. Curcumin is a polyphenol that is unstable to pH, temperature, and light. Curcumin acts as an antioxidant and antibacterial. Curcumin with two aromatic groups is used as a biocatalyst to prevent separation at the end of the reaction. Research on the interesterification process using biocatalysts with one aromatic group has not yet obtained satisfactory results in terms of the products and physical properties produced. By using curcumin with two aromatic groups as a biocatalyst, it is expected to obtain better results. The aim of this research is to obtain optimum conditions for the interesterification process of used cooking oil using curcumin biocatalyst. If the stages in the biodiesel production process can be reduced, an effective and efficient process will be achieved, so that the price of biodiesel can compete with diesel fuel. Therefore, studies that are devoted the production of biodiesel from used cooking oil using curcumin biocatalyst are scientific relevance.

2. Literature review and problem statement

Curcumin ($C_{21}H_{20}O_6$) found in turmeric and temulawak rhizomes serves as a yellow or orange colorants. It is an aromatic compound containing two aromatic groups with delocalized molecular orbitals. This electron delocalization generates a magnetic field that affects the stability of polar compounds [13]. As the number of aromatic groups in a compound increases, the resulting magnetic field becomes stronger. This field can attract paramagnetic oxygen (O) atom, leading to the compound to become unstable [14]. When a substance becomes unstable, it tends react more readily with other compounds.

In the context of this findings, several researches use curcumin as a catalyst. According to [15], the manufacture of biohydrogen using a combination of activated carbon biocatalyst and curcumin 1:1 with a biomass pyrolysis process at a temperature of 500°C obtained 25.6% hydrogen gas. Aromatic rings in curcumin form phi-phi bond strengthening the electromagnetic force generated by the delocalized electrons to trigger hydrogen and menthane formation. The research conducted by [16] show that the electrocatalytic process of water and leftover water from boiling instant noodles at a reaction time of 7,201 seconds with a turmeric (curcumin) + carbon catalyst, hydrogen was obtained with a concentration of around 18,000 ppm. The phenolic compound of curcumin turmeric inhibits the enzymatic reaction of protein in instant noodles. Several studies above prove that curcumin biocatalyst plays a very important role in the production of biohydrogen, so it is hoped that it can also be used as a biocatalyst in the production of biodiesel. Similarly, the process of forming biodiesel by using the magnetic properties of heterogeneous catalysts produces satisfactory results. In the research carried out by [17] on the transesterification of used cooking oil by applying the magnetic properties of the $CuFe_2O_4$ catalyst, a yield of 93% was found at a reaction time of 90 minutes. The catalyst preparation stage is quite long, namely co-precipitation and combined co-precipitation and hydrothermal. The catalysts were characterized by TGA, SEM, EDX, HR-TEM, FT-IR, Raman spectrometry, nitrogen adsorption-desorption and VSM. In the transesterification process, the by-product produced is glycerol which must be separated from the main product methyl ester so that the process stages become longer and ineffective. Simultaneous research [18] on transesterification and esterification using the Fe_3O_4/SiO_2 magnetic catalyst achieved a conversion of 93% at a reaction time of six hours. The catalyst preparation stages include hydrothermal, coating, grafting, and immobilization. Catalyst characterization includes Bronsted acidity determination, XRD, FT-IR, FESEM-EDS, TEM, VSM, and N_2 adsorption-desorption. In addition to the long catalyst preparation stages, long reaction times are also a limitation in the use of heterogeneous catalysts in biodiesel production, making the process less effective. Furthermore, biodiesel production from the transesterification of Pistacia chinensis seed oil with a cellulose magnetic catalyst achieved a yield of 93.1% at 60°C and an 80 minutes reaction time [19]. The catalyst preparation includes regeneration, modification and immobilization. Catalyst characterization includes Fourier transform infrared, X-ray diffraction and scanning electron microscopy. At the end of the reaction, the catalyst must be removed by filtration. The resulting glycerol byproduct must be removed because it is an impurity that will interfere with the biodiesel's performance. Based on the research performed by [20], a yield of 65.73% was found in the transesterification of sunflower oil with a $K_2CO_3/\gamma\text{-}Al_2O_3$ magnetic catalyst at a 240 minutes reaction time. The catalyst preparation includes coprecipitation, mixing, milling, extrusion, drying and calcination. Catalyst characterization includes mechanical strength, magnetization measurements, thermogravimetric analysis, N_2 adsorption-desorption, X-ray diffraction and scanning electron microscopy. The resulting yield is relatively low for a 4-hour reaction time. Additional reaction time is required to achieve a higher yield, which also increases production costs. The use of heterogeneous magnetic catalysts in biodiesel production requires multiple stages, including preparation, production, activation, and separation at the end of the reaction. These long stages are not effective in the biodiesel production process. Making biodiesel using the interesterification process is more effective because the triacetin by-product does not need to be separated because it functions as a bioadditive in biodiesel.

Several findings have used organic aromatic compounds as biocatalysts and bio-additives to obtain satisfactory results. In the work [21], the addition of eucalyptus oil with a concentration of 5% to CPO will produce a flash point that meets quality. The aromatic compounds in eucalyptus oil will weaken the van der Walls power so that the flash point in CPO will decrease. In the work [22], the addition of 250 ppm of *Areca* nut extract will reduce the viscosity of vegetable oil. The aromatic ring containing the magnetic field in epicatechin, which is an aromatic compound in *Arecha* extract, weakens the van der Walls forces in the oil so that the viscosity will decrease. In the work [23], adding eucalyptus oil to coconut oil will produce a brighter blue flame color. The magnetic field produced by the eucalyptus oil component, namely 1,8-cineole, will cause the oil to be polar, making it easier to burn and evaporate. As reported by [24], the interesterification reaction of palm oil and methyl acetate with eugenol as the catalyst produced 83.16% of crude oil under 60°C reaction temperature, within 15 minutes reaction time and a mole ratio of oil:methyl acetate = 1:6, and a 300 rpm stirring speed. In this study, the maximum crude yield was only 83.16%, which was likely due to the lack of catalyst strength in converting the reactants. In this study, the highest crude yield was obtained at the shortest reaction time, 15 minutes. There was no crude yield data available for reaction times shorter than 15 minutes. The technical eugenol used as a biocatalyst likely contains many impurities

that would affect the interesterification process. According to [12], the interesterification process of palm oil with cajuput oil biocatalyst, a crude yield of 65.88% and an acid number of 0.426 mg KOH/g sample are obtained, which meets Indonesian National Standard (SNI) 7182-2015. The highest crude yield was achieved at the longest reaction time, 75 minutes, so the rational conditions for the study had not been reached. In this study, the biocatalyst concentration was set at 0.75%, so the effect of varying biocatalyst concentration on the interesterification process is unknown. The commercial cajuput oil used as the biocatalyst naturally contains many impurities that will affect the resulting product concentration. In the research of palm oil interesterification process with eugenol and cajuput oil biocatalysts, satisfactory results have not been obtained in terms of crude yield and density of the resulting product. This is because the magnetic properties of the two catalysts are small because they only contain one aromatic group. Previous research on biodiesel production has not yet explored the used of curcumin as a biocatalyst.

All this allows to assert that it is expedient to conduct a study on obtain rational conditions for the interesterification process of used cooking oil related to the reaction time and concentration of curcumin biocatalyst, so that a more efficient biodiesel production process is obtained, namely producing biodiesel with high yield and physical properties that meet standards.

3. The aim and objectives of research

The research aimed to obtain the rational conditions for catalyst concentration and reaction time for interesterification of used cooking oil into methyl ester as well as triacetin by applying curcumin biocatalyst with two aromatic groups. This will make it possible to obtain a more efficient, effective, and economical method of making methyl esters, allowing the price of biodiesel produced to compete with the value of diesel.

To achieve this aim, the following objectives were accomplished:

– to identify the influence of catalyst concentration and reaction time for interesterification of used cooking oil with curcumin biocatalyst;

– to achieve products that meet the standards, namely density and acid number;

– to determine the methyl ester products formed in the interesterification reaction of used cooking oil with curcumin biocatalyst;

– to obtain simulation data related to the molecular behavior of the reacting compounds, namely the C-O or O=C bond length.

4. Materials and methods

4. 1. The object and hypothesis of research

The object of research is the process of interesterification of used cooking oil with curcumin biocatalysts. The main hypothesis of research is that increasing the reaction time and catalyst concentration increase the crude yield, reducing the density and acid value of the product. Assumptions made in the study are the reaction time and biocatalyst concentration increase, more reactants will react to produce more products. Simplifications adopted in the study are the used cooking oil used is household waste and the curcumin catalyst used is pro analyst.

4. 2. Materials

The materials applied in the analysis were distilled water (H_2O), ethanol p.a (Merck, 99.9%), potassium hydroxide (Merck, 90%), methyl acetate (Sigma Aldrich, 99.9%), waste cooking oil, acetone p.a (Merck, 99%), phenolphthalein indicator (pp), activated carbon and curcumin (Merck, 99%).

4. 3. Refining of waste cooking oil

During the process, four liters of waste cooking oil were filtered and heated at 40°C. KOH 0.1 N was added and stirred for 10 minutes until pH = 7. The neutralized oil was heated at 70°C and 700 g of activated carbon was added. Moreover, the oil was heated at 150°C and stirred for 60 minutes. The results were separated by filtration using filter paper and a separating funnel.

4. 4. Preparation of ingredients

Waste cooking oil was examined for % FFA and water content. When % FFA < 3% and water content < 0.6%, it was directly applied for interesterification reaction [25]. Consequently, when % FFA > 3% saponification reaction was conducted and when water content > 0.6%, evaporation was performed in an oven at 110°C until constant mass. The waste cooking oil was filtered, heated to remove its water content and 0.1 N KOH was added until the pH was neutral for the neutralization reaction. The waste cooking oil was then added with 10% activated carbon.

4. 5. Interesterification reaction with curcumin biocatalyst

A total of 250 g of waste cooking oil was placed in a three-neck flask and heated to 60°C. Separately, methyl acetate with moles ratio of methyl acetate to waste cooking oil 1:6 and 1:12 as well as varying concentration of curcumin biocatalyst having 0.5%, 1%, 1.5%, 2%, and 2.5% were placed in Erlenmeyer flasks and heated to 60°C. Once all components reached the desired temperature, methyl acetate and biocatalyst were added to the three-neck flask containing preheated waste cooking oil. The flask, equipped with a reflux condenser was maintained at 60°C and stirred at 300 rpm applying a magnetic stirrer. The reaction time was recorded based on predetermined intervals including 15, 30, 45, 60, 75, 90, and 105 minutes. At the end of the reaction time, a 50 ml sample was collected to separate the remaining methyl acetate from the reaction mixture.

4. 6. Separation of interesterification reaction results with methyl acetate reaction remainder

During this process, the reaction sample was distilled at 70°C to separate any unreacted methyl acetate. Distillation continued until no more distillate was collected. The distillate consisted of methyl acetate ($bp = 57°C$) while the remaining residue included unreacted oil/triglyceride ($bp = 383°C$), methyl ester ($bp = 373.78°C$), triacetin ($bp = 306.74°C$) and curcumin ($mp = 183°C$). After the distillation was complete, the residue, comprising methyl ester, triacetin, and biocatalyst was weighed as the final product of the interesterification reaction. The crude yield of the product was then calculated using equation

$$x = \frac{w}{\rho \cdot v} \cdot 100\%, \qquad (1)$$

where x – the crude yield, %; w – the weight of product, g; ρ represents the density of product, g/ml; and v denotes the volume of sample, ml.

4. 7. Product analysis

The analysis process included density, acid number, and methyl ester composition applying GC-MS (Gas Chromatography-Mass Spectrometry). The analysis results were compared with the Indonesian biodiesel standard SNI 7182-2015.

4. 8. Simulation with ChemDraw Professional 15.0

Molecular behavior (molecular modeling) of compounds included in interesterification reactions was identified by simulation using ChemDraw Professional 15.0 (USA). Moreover, the parameters searched were molecular dynamics, and bond length between C – O or O = C.

5. Results of the interesterification reaction of used cooking oil with curcumin biocatalyst

5. 1. Identifying the influence of catalyst concentration and reaction time for interesterification of used cooking oil with curcumin biocatalyst

Data on the crude yield were obtained through analysis and calculations. Fig. 1 shows the relationship between reaction time and crude yield with the variable mass of curcumin biocatalyst at a mole ratio of oil:methyl acetate = 1:6.

Fig. 2 shows the relationship between reaction time and crude yield with the variable mass of curcumin biocatalyst at a mole ratio of oil:methyl acetate = 1:12.
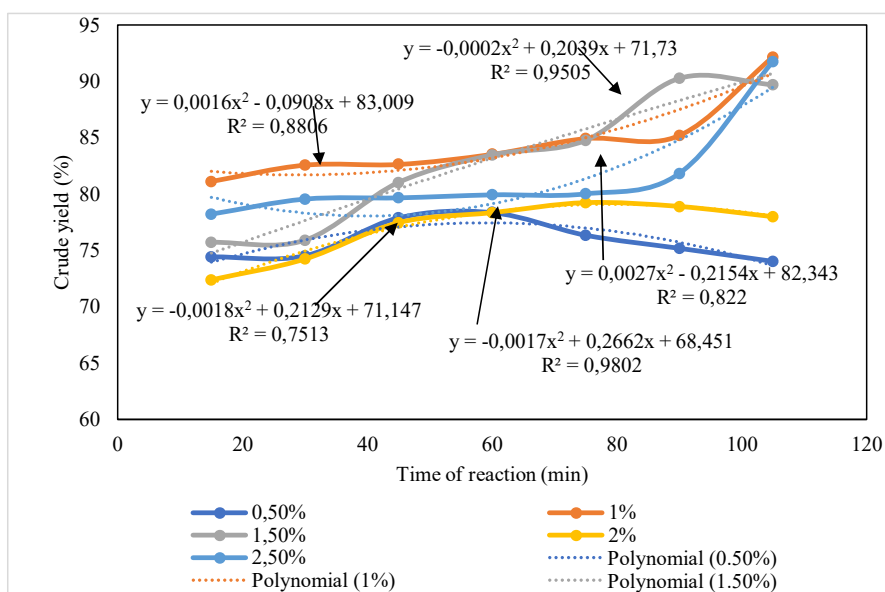

Fig. 1. Relationship between reaction time and crude yield with variations in biocatalyst concentration at a mole ratio of 1:6
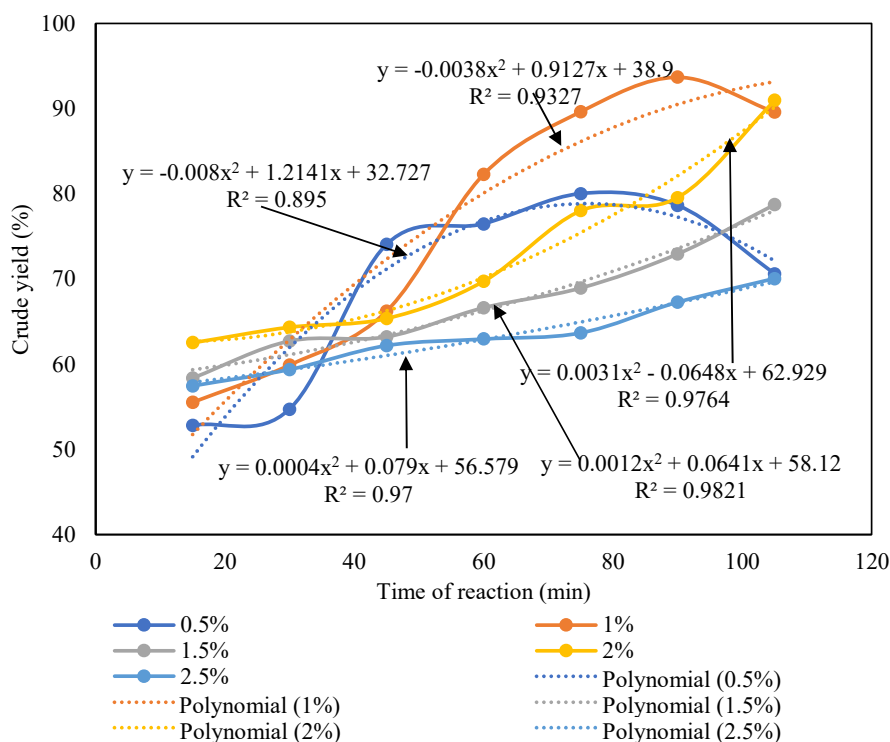

Fig. 2. Relationship between reaction time and crude yield with variations in biocatalyst concentration at a mole ratio of 1:12

Fig. 1, 2 both show that the relationship between crude yield and time and mass of biocatalyst is directly proportional until optimum conditions are obtained.

5. 2. Density and acid number data

Table 1 shows the results of the analysis of the physical properties of the interesterification reaction products of used cooking oil with curcumin biocatalyst at a mole ratio of oil:methyl acetate = 1:6.

Table 1

Density and acid number data of the product at a mole ratio of oil:methyl acetate = 1:6

Concentration of curcumin (%)	Time of reaction (min)	Density (g/ml)	Acid number (mg KOH/g)
0.5	15	0.923	0.897
	30	0.946	0.785
	45	0.93	0.561
	60	0.919	0.561
	75	0.915	0.448
	90	0.907	0.336
	105	0.946	0.330
1	15	0.912	0.785
	30	0.93	0.673.
	45	0.925	0.673
	60	0.919	0.561
	75	0.904	0.448
	90	0.921	0.224
	105	0.91	0.224
1.5	15	0.911	0.336
	30	0.936	0.224
	45	0.919	0.224
	60	0.925	0.224
	75	0.907	0.224
	90	0.913	0.224
	105	0.905	0.224
2	15	0.936	0.336
	30	0.908	0.224
	45	0.909	0.224
	60	0.905	0.224
	75	0.903	0.224
	90	0.89	0.224
	105	0.885	0.224
2.5	15	0.893	0.224
	30	0.88	0.224
	45	0.882	0.224
	60	0.901	0.224
	75	0.89	0.224
	90	0.909	0.224
	105	0.884	0.224

Table 2 shows the results of the analysis of the physical properties of the interesterification reaction products of used cooking oil with curcumin biocatalyst at a mole ratio of oil:methyl acetate = 1:12.

Tables 1, 2 show that the density and acid number values decrease closer to the biodiesel standard with increasing reaction time and increasing mass of curcumin biocatalyst.

Table 2

Density and acid number data of the product at a mole ratio of oil:methyl acetate = 1:12

Concentration of curcumin (%)	Time of reaction (min)	Density (g/ml)	Acid number (mg KOH/g)
0.5	15	0.926	0.336
	30	0923	0.336
	45	0.929	0.336
	60	0.916	0.336
	75	0.911	0.336
	90	0.926	0.336
	105	0.91	0.336
1	15	0.923	0.224
	30	0.915	0.224
	45	0.933	0.224
	60	0.926	0.112
	75	0.905	0.224
	90	0.926	0.112
	105	0.911	0.112
1.5	15	0.907	0.224
	30	0.904	0.224
	45	0.901	0.112
	60	0.906	0.112
	75	0.914	0.112
	90	0.914	0.112
	105	0.892	0.112
2	15	0.901	0.112
	30	0.891	0.112
	45	0.893	0.112
	60	0.889	0.112
	75	0.897	0.112
	90	0.894	0.112
	105	0.897	0.112
2.5	15	0.887	0.112
	30	0.89	0.112
	45	0.888	0.112
	60	0.887	0.112
	75	0.888	0.112
	90	0.885	0.112
	105	0.887	0.112

5. 3. Determination of the methyl ester products formed in the interesterification reaction of used cooking oil with curcumin biocatalyst

GC-MS analysis was performed on samples of the interesterification reaction results of used cooking oil with 2.5% curcumin biocatalyst at a 105 minutes reaction time. This process had a mole ratio of oil: methyl acetate = 1:6 which was the best condition of the research. Table 3 shows the results of GC-MS analysis of methyl ester components in the interesterification process of used cooking oil at a reaction time of 105 minutes. In peak 5, methyl ester compounds were detected with an area of 15%. Fig. 3 shows the chromatogram of methyl ester from the interesterification reaction of used cooking oil at 105 minutes.

Fig. 3 is a chromatogram of the dominant methyl ester formed in the interesterification reaction of used cooking oil with 2.5% curcumin biocatalyst, reaction time 105 minutes and mole ratio of oil:methyl acetate = 1:6, namely Hexadecanoic acid, 1-methylethyl ester

Table 3

GC-MS analysis results at a reaction time of 105 minutes

Peak	Component	Area (%)
1	Propanoic acid, 3-ethoxy-ethyl ester	64.86
2	5,6,7-tris (Decyloxy)-10-methyl-9-phenanthrenecarbaldehyde	8.92
3	2,6-Dimethyl-4-heptanone	8.09
4	3-Methoxy-5-(methoxymethoxy)-7-methyl-6-(3-(trimethylsilyl)	3.13
5	Hexadecanoic acid, 1-methylethyl ester	15

Hit#:3 Entry:209929 Library:WILEY7.LIB
SI:86 Formula:C19 H38 O2 CAS:142-91-6 MolWeight:298 RetIndex:0
CompName:Hexadecanoic acid, 1-methylethyl ester (CAS) Isopal $$ ISOPROPYL ESTER OF PALMITIC ACID $$ Deltyl $$ Propal $$ Isopalm $
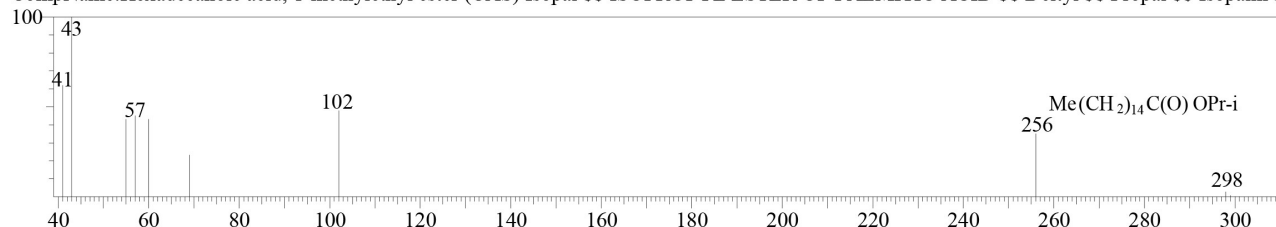

Fig. 3. Hexadecanoic acid, 1-methylethyl ester chromatogram

5. 4. Obtaining simulation data related to the molecular behavior of the reacting compounds, namely the C-O or O=C bond length

Table 4 shows the results concerning simulation of the calculation of C-O and O=C bond length in triglycerides and methyl acetate before and after adding of curcumin biocatalyst. The bond length of O(82)=C(83) in methyl acetate, which was initially 1.396Å, after the addition of curcumin biocatalyst and dynamic molecular settings was performed became 1.43Å, increasing in length by 0.034Å. Moreover, the bond length of C(1)-O(4) in triglycerides, which was initially 1.382Å, after the addition of curcumin biocatalyst and dynamic molecular settings was conducted became 1.445Å, increasing in length by 0.063Å. All C-O and O=C bonds in methyl acetate and triglycerides increased in length after simulation. This occurred because the O atom in methyl acetate and triglycerides had two unpaired electrons leading to paramagnetic. In addition, the paramagnetic O atom was attracted by a compound containing a magnetic field, namely the curcumin biocatalyst which had two aromatic groups, increasing the length C-O and O=C bonds [10, 24].

Simulation using ChemDraw Professional 15.0 software to determine the changes in the length of the C-O and O=C bonds in triglycerides and methyl acetate after the addition of curcumin biocatalyst with two aromatic groups as shown in Fig. 4.

Fig. 4 is a molecular dynamic display of triglycerides and methyl acetate after the addition of curcumin biocatalyst from ChemDraw Professional 15.0.
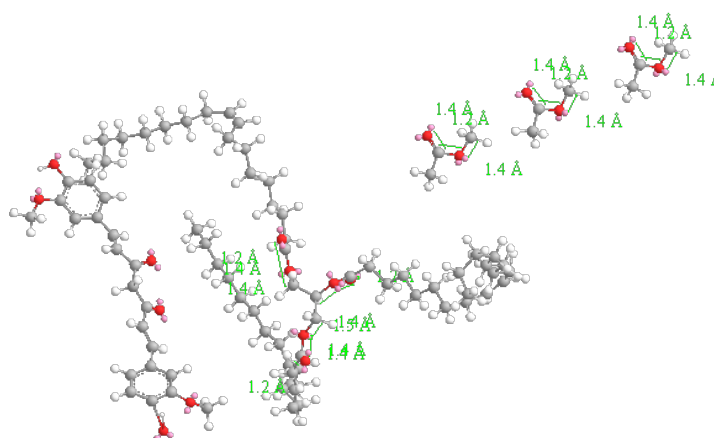

Fig. 4. Molecular dynamic of triglyceride and methyl acetate after adding curcumin biocatalyst

Table 4

C-O/O=C bond length before and after addition of curcumin biocatalyst

C-O bond / O=C bond	Actual final (Å)	Optimal initial (Å)
1	2	3
O(82) = C(83) of methyl acetate	1.430	1.396
C(81) − O(84) of methyl acetate	1.221	1.208
C(81) − O(82) of methyl acetate	1.360	1.338
O(77) = C(78) of methyl acetate	1.432	1.396
C(76) − O(79) of methyl acetate	1.218	1.208
C(76) − O(77) of methyl acetate	1.360	1.338
O(72) = C(73) of methyl acetate	1.428	1.396
C(71) − O(74) of methyl acetate	1.219	1.208

1	2	3
C(71) − O(72) of methyl acetate	1.363	1.338
C(9) − O(15) of triglyceride	1.217	1.208
C(8) − O(13) of triglyceride	1.223	1.208
C(7) − O(11) of triglyceride	1.218	1.208
O(6) = C(9) of triglyceride	1.357	1.338
O(5) = C(8) of triglyceride	1.363	1.338
O(4) = C(7) of triglyceride	1.366	1.338
C(3) − O(6) of triglyceride	1.431	1.389
C(2) − O(5) of triglyceride	1.448	1.389
C(1) − O(4) of triglyceride	1.445	1.382

6. Discussion of results of the interesterification reaction of used cooking oil with curcumin biocatalyst

In Fig. 1, there is an observation that reaction time increased, and the crude yield obtained improved [4]. At a curcumin biocatalyst concentration of 0.5%, the optimum crude yield obtained at a 60 minutes reaction time was 78.37%. Meanwhile, the highest crude yield was at 105 minutes having a curcumin concentration of 1% was 92.15%. At a curcumin biocatalyst concentration of 1.5%, the optimum crude yield obtained at a 90 minutes reaction time was 90.25%. Additionally, the optimum crude yield at 75 minutes having a curcumin concentration of 2% was 79.22%. The highest crude yield at a curcumin biocatalyst concentration of 2.5% was 91.74%, obtained at a 105 minutes reaction time. At a mole ratio of oil: methyl acetate = 1:6, the best conditions for the interesterification reaction of used cooking oil were obtained with a curcumin biocatalyst concentration of 1% and a reaction time of 105 minutes with a crude yield of 92.15%. The results obtained were better when the interesterification process of palm oil was compared with eugenol biocatalyst, which had a crude yield of 83.16% [24].

Fig. 2 shows that as the reaction time increased, the crude yield obtained became higher. The optimum crude yield at a curcumin biocatalyst concentration of 0.5% obtained at a reaction time of 75 minutes was 80.02%. Meanwhile, at a curcumin concentration of 1%, the optimum crude yield at 90 minutes was 93.69%. The highest crude yield at a curcumin biocatalyst concentration of 1.5% obtained at 105 minutes was 78.73%. Additionally, at a curcumin concentration of 2%, the highest crude yield at a reaction time of 105 minutes was 90.99%. The highest crude yield at a curcumin biocatalyst concentration of 2.5% was 70.05%, which was found at a 105 minutes reaction time. At a mole ratio of oil: methyl acetate = 1:12, the optimum conditions for the interesterification reaction of used cooking oil were obtained with a curcumin biocatalyst concentration of 1% and a 9 minute time with a crude yield of 93.69%. Consequently, the results of this research were better when the interesterification process of palm oil was compared with eucalyptus oil biocatalyst, which contained a crude yield of 65.88% [24].

Table 1 shows that many density values and several acid numbers did not meeting SNI 7182-2015. These values included density 0.85–0.89 g/ml and acid number 0.5 mg KOH/g (12). At a biocatalyst concentration of 0.5%, none of the density values met the required standards, while the acid number values met the criteria at reaction time of 75, 90, and 105 minutes. In similar way at a biocatalyst concentration of 1% none of the density values

met the requirement but the acid number values met the criteria at reaction time variables of 75, 90, and 105 minutes. Using a biocatalyst concentration of 1.5%, all density values were not met, as the acid number values were met at all reaction time variables. Following the discussion, the density values met at reaction times of 90 as well as 105 minutes at a biocatalyst concentration of 2%, and the acid number values met at all reaction time variables. Using a biocatalyst concentration of 2.5%, the density value met the requirements at reaction times of 30, 45, 75, and 105 minutes, while the acid number value also met the prerequisites at all reaction time variables.

Table 2 shows that many density values and some acid numbers did not meet SNI 7182-2015. These values included density 0.85–0.89 g/ml and acid number 0.5 mg KOH/g. Using a biocatalyst concentration of 0.5%, all density values did not fulfill the requirements while the acid number values met the conditions at reaction time variables of 75, 90, and 105 minutes. All density values did not meet the requirements at a biocatalyst concentration of 1%, and acid number values meet the criteria at reaction time variables of 75, 90, and 105 minutes. Using a biocatalyst concentration of 1.5%, all density values did not meet the conditions while the acid number values met the requirements at all reaction time variables. Following this discussion, the density values met the prerequisites at reaction times of 90 and 105 minutes at a biocatalyst concentration of 2% as the acid number values also met the requirements at all reaction time variables. At a biocatalyst concentration of 2.5%, the density value met the requirements at reaction times of 30, 45, 75 as well as 105 minutes and the acid number value met the conditions at all reaction time variables.

The best condition in this study was achieved at a reaction time of 105 minutes and a biocatalyst mass of 2.5% which is the highest value of the research variables.

Further research is needed with a reaction time of more than 105 minutes and a biocatalyst concentration of more than 2.5% to obtain better rational research conditions. In this study, the analysis of physical properties was only density and acid number. A comprehensive analysis of the physical properties of biodiesel is needed according to SNI 7182-2015.

7. Conclusion

1. This research shows that the highest crude yield is found at 91.74% from the analysis and calculations. The outcome is achieved in the interesterification reaction of

waste cooking oil with a curcumin biocatalyst concentration of 2.5% at a 105 minutes reaction time.

2. A density value of 0.884 g/ml and an acid number of 0.224 mg KOH/g are obtained which met SNI 7182-2015 under these conditions.

3. The dominant methyl ester component forms was Hexadecanoic acid, 1-methylethyl ester.

4. The simulation results show an increase in the length of C-O and O=C bonds due to the influence of the magnetic field on the aromatic group of curcumin.

Conflict of interest

The authors declare no conflict of interest concerning this research. This could be through financial, personal, or authorship that could affect the findings and the results presented in the research.

Data availability

The manuscript has no associated data.

References

1. Daryono, E. D., Sinaga, E. J. (2017). Rapid in situ transesterification of papaya seeds to biodiesel with the aid of co-solvent. International Journal of Renewable Energy Research, 7 (1). https://doi.org/10.20508/ijrer.v7i1.5275.g6998

2. Calero, J., Luna, D., Sancho, E. D., Luna, C., Bautista, F. M., Romero, A. A. et al. (2015). An overview on glycerol-free processes for the production of renewable liquid biofuels, applicable in diesel engines. Renewable and Sustainable Energy Reviews, 42, 1437–1452. https://doi.org/10.1016/j.rser.2014.11.007

3. Chuepeng, S., Komintarachat, C. (2018). Interesterification optimization of waste cooking oil and ethyl acetate over homogeneous catalyst for biofuel production with engine validation. Applied Energy, 232, 728–739. https://doi.org/10.1016/j.apenergy.2018.09.085

4. Daryono, E. D., Wardana, I. N. G., Cahyani, C., Hamidi, N. (2021). Biodiesel production process without glycerol by-product with base catalyst: effect of reaction time and type of catalyst on kinetic energy and solubility. IOP Conference Series: Materials Science and Engineering, 1053 (1), 012058. https://doi.org/10.1088/1757-899x/1053/1/012058

5. Lopes, T. I. B., Ribeiro, M. D. M. M., Ming, C. C., Grimaldi, R., Gonçalves, L. A. G., Marsaioli, A. J. (2016). Comparison of the regiospecific distribution from triacylglycerols after chemical and enzymatic interesterification of high oleic sunflower oil and fully hydrogenated high oleic sunflower oil blend by carbon-13 nuclear magnetic resonance. Food Chemistry, 212, 641–647. https://doi.org/10.1016/j.foodchem.2016.06.024

6. Interrante, L., Bensaid, S., Galletti, C., Pirone, R., Schiavo, B., Scialdone, O., Galia, A. (2018). Interesterification of rapeseed oil catalysed by a low surface area tin (II) oxide heterogeneous catalyst. Fuel Processing Technology, 177, 336–344. https://doi.org/10.1016/j.fuproc.2018.05.017

7. Simões, S. S., Ribeiro, J. S., Celante, D., Brondani, L. N., Castilhos, F. (2020). Heterogeneous catalyst screening for fatty acid methyl esters production through interesterification reaction. Renewable Energy, 146, 719–726. https://doi.org/10.1016/j.renene.2019.07.023

8. Nunes, A. L. B., Castilhos, F. (2020). Chemical interesterification of soybean oil and methyl acetate to FAME using CaO as catalyst. Fuel, 267, 117264. https://doi.org/10.1016/j.fuel.2020.117264

9. Tian, Y., Xiang, J., Verni, C. C., Soh, L. (2018). Fatty acid methyl ester production via ferric sulfate catalyzed interesterification. Biomass and Bioenergy, 115, 82–87. https://doi.org/10.1016/j.biombioe.2018.04.013

10. Daryono, E. D., Wardana, I. N. G., Cahyani, C., Hamidi, N. (2022). Interesterification Process of Palm Oil Using Base Catalyst: The Effect of Stirring Speed and Type of Catalyst on Kinetic Energy and Dipole Moment. International Journal on Advanced Science, Engineering and Information Technology, 12 (4), 1580. https://doi.org/10.18517/ijaseit.12.4.12500

11. Daryono, E. D. (2015). Reactive extraction process in isolation of eugenol of clove essential oil (Syzigium aromaticum) based on temperature and time process. International Journal of ChemTech Research, 8 (11), 564–569.

12. Daryono, E. D., Dewi, R. K. (2022). Biodiesel from palm oil with interesterification process using bio-catalyst cajuput oil. Konversi, 11 (2). https://doi.org/10.20527/k.v11i2.13982

13. Purnami, P., Wardana, I. N. G., Hamidi, N., Sasongko, M. N., Darmadi, D. (2019). The Effect of Rhodium (III) Sulfate and Clove Oil Catalysts on The Droplet Combustion Characteristics of Castor Oil. International Journal of Integrated Engineering, 11 (5), 66–71. https://doi.org/10.30880/ijie.2019.11.05.009

14. Purnami, Hamidi, N., Sasongko, M. N., Widhiyanuriyawan, D., Wardana, I. N. G. (2020). Strengthening external magnetic fields with activated carbon graphene for increasing hydrogen production in water electrolysis. International Journal of Hydrogen Energy, 45 (38), 19370–19380. https://doi.org/10.1016/j.ijhydene.2020.05.148

15. Suprianto, T., Winarto, Wijayanti, W., Wardana, I. N. G. (2021). Synergistic effect of curcumin and activated carbon catalyst enhancing hydrogen production from biomass pyrolysis. International Journal of Hydrogen Energy, 46 (10), 7147–7164. https://doi.org/10.1016/j.ijhydene.2020.11.211

16. Willy Satrio, N., Winarto, Sugiono, Wardana, I. N. G. (2020). Hydrogen production from instant noodle wastewater by organic electrocatalyst coated on PVC surface. International Journal of Hydrogen Energy, 45 (23), 12859–12873. https://doi.org/10.1016/j.ijhydene.2020.03.002

17. Ali, R. M., Elkatory, M. R., Hamad, H. A. (2020). Highly active and stable magnetically recyclable CuFe2O4 as a heterogenous catalyst for efficient conversion of waste frying oil to biodiesel. Fuel, 268, 117297. https://doi.org/10.1016/j.fuel.2020.117297

18. Xie, W., Wang, H. (2020). Immobilized polymeric sulfonated ionic liquid on core-shell structured Fe3O4/SiO2 composites: A magnetically recyclable catalyst for simultaneous transesterification and esterifications of low-cost oils to biodiesel. Renewable Energy, 145, 1709–1719. https://doi.org/10.1016/j.renene.2019.07.092

19. Han, Y.-Z., Hong, L., Wang, X.-Q., Liu, J.-Z., Jiao, J., Luo, M., Fu, Y.-J. (2016). Biodiesel production from Pistacia chinensis seed oil via transesterification using recyclable magnetic cellulose-based catalyst. Industrial Crops and Products, 89, 332–338. https://doi.org/10.1016/j.indcrop.2016.05.015

20. Silveira Junior, E. G., Justo, O. R., Perez, V. H., Reyero, I., Serrano-Lotina, A., Campos Ramirez, L., dos Santos Dias, D. F. (2018). Extruded Catalysts with Magnetic Properties for Biodiesel Production. Advances in Materials Science and Engineering, 2018 (1). https://doi.org/10.1155/2018/3980967

21. Marlina, E., Basjir, M., Ichiyanagi, M., Suzuki, T., Gotama, G. J., Anggono, W. (2020). The Role of Eucalyptus Oil in Crude Palm Oil As Biodiesel Fuel. Automotive Experiences, 3 (1), 33–38. https://doi.org/10.31603/ae.v3i1.3257

22. Wardoyo, Widodo, A. S., Wijayanti, W., Wardana, I. N. G. (2021). The Role of Areca catechu Extract on Decreasing Viscosity of Vegetable Oils. The Scientific World Journal, 2021, 1–8. https://doi.org/10.1155/2021/8827427

23. Soebiyakto, G., Wardana, I. N. G., Hamidi, N., Yuliati, L. (2020). Addition of bio-additive as a catalyst of burning vegetable oil influenced by 4 pole magnetic field. Eastern-European Journal of Enterprise Technologies, 2 (6 (104)), 46–55. https://doi.org/10.15587/1729-4061.2020.198308

24. Daryono, E. D., Jimmy, J., Setyawati, H. (2024). Production of Biodiesel Without Catalyst Separation with Palm Oil Interesterification Process Using Essential Oil Biocatalyst. Chemistry & Chemical Technology, 18 (3), 356–362. https://doi.org/10.23939/chcht18.03.356

25. Pullen, J., Saeed, K. (2015). Investigation of the factors affecting the progress of base-catalyzed transesterification of rapeseed oil to biodiesel FAME. Fuel Processing Technology, 130, 127–135. https://doi.org/10.1016/j.fuproc.2014.09.013