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/er.4844 SHORT COMMUNICATION

1**A facile noncatalytic methyl ester production from waste chicken tallow using single step subcritical methanol: Optimization study**

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Summary In this modern era, an increase in urbanization causes the escalating trend of fuel demand as well as environmental pollution problems. Various biofuels research with the respect of climate change and emission reduction recently intensifies, particularly in biodiesel. In Indonesia, diesel oil currently in use contains 20% of biodiesel. Utilizing waste-based resources such as rendered chicken tallow as the feedstock could be the solution to both energy and envi- ronmental challenges. However, chicken tallow contains a significant amount of free fatty acid (FFA) which will obstruct the production yield of biodiesel. In this study, catalyst-free subcritical methanol has been employed to convert waste chicken tallow (WCT) with high FFA into biodiesel. Design of experi- ment was conducted to study the effect of temperature, time,

6and the molar ratio of methanol to fats on the purity and recovery of fatty acid methyl esters (FAMEs). Based on the optimization

study

8performed by response surface methodology (RSM

), all three independent variables

2gave a significant effect on the recovery of

2FAME. From the experimental results, the maximum FAME yield obtained was

98.43 ± 0.22% with the optimum condition as follows: 167°C, 36.8 minutes, and 42.7:1 (methanol/WCT, mol/mol), while the pre- dicted FAME yield obtained using RSM was 97.76%. The methyl ester compo- sition of WCT-based biodiesel ranges from C13 to C24. K E Y W O R D S catalyst-free, facile transesterification, optimization study, renewable energy, subcritical methanol, waste chicken tallow, waste-derived biodiesel 1 | INTRODUCTION Energy sustainability is regarded worldwide as one of the indicators of economic and infrastructure development of a country.1 To date, Indonesia uses fossil energy sources as much as 94%, and only the remaining 6% use the ben- efits of bioresource-based energy to meet the yearly energy requisites. Excessive use of fossil energy increases Abbreviation: WCT, waste chicken tallow; FFA, free fatty acid; FAME(s),

2fatty acid methyl ester(s); RSM, response surface methodology

; SpCM, supercritical methanol; SCM, subcritical methanol; GC-FID, gas chromatography-flame ionization detector; CCC-CCD, circumscribed central composite design

the depletion rate, and the availability of the sources of these fuels in nature will subsequently begin to decline. Another problem that arises from the use of fossil fuels is environmental pollution, caused by the emission of SOx, NOx, and CO2.2 With the depletion of petroleum reserves, various efforts have been taken to seek more sustainable and environmentally friendly alternative energy sources. In the last few decades, studies to find alternative sources of renewable energy have been widely performed with biodiesel as one of the most vastly studied renewable energy. Biodiesel derived from biological sources received extensive interest as it lowers the global reliance on petro- leum products, the energy crisis, and pollution.3,4 Various feedstocks including edible oils,1,5,6 nonedible oils,7,8 raw oils,9 algae,10-12 and animal fats,13,14 as well as sundry of processing techniques,

> 5**have been developed to produce high-quality biodiesel**

. Several routes that have been studied for the conver- sion of lipid to biodiesel are as follows: the base-catalyzed transesterification,1,14

> 4**two steps acidic esterification followed by**

alkaline transesterification9,13,15; enzyme- catalyzed esterification/transesterification3 and

> 4**noncatalytic transesterification using** methanol **under subcritical**,5,12 **and supercritical conditions**

.10,16 The selec- tion of a suitable technique depends on the quality of fats. Currently, Indonesia commercially produces biodiesel from edible oil, primarily from palm oil. However, the use of edible oils leads to food shortages. Thus, they are noneconomic and nonfeasible. Non-edible oil, particu- larly waste-based resources, is one of the best alternatives for biodiesel feedstock due to its lower cost and positive waste utilization. Waste-derived biodiesel also has the additional advantage of avoiding environmental impacts. Indonesia produces more than 2 million tonnes of chicken annually,17 where the fat content is around 13.6%.18 Chicken tallow is usually discarded as waste due to a health hazard. In spite of that, waste chicken tallow (WCT) possesses a substantial amount of free fatty acids (FFA) and triglycerides which is able to be converted into biodiesel. Direct application of WCT

> 14**as raw material for the production of biodiesel**

faces sev- eral problems since it is impossible to directly convert the tallow into biodiesel using transesterification. The presence of water in WCT induces the hydrolysis of triglycerides

> 2**into FFA**, and **high level of FFA content** (> 0.1%) **in the** tallow promotes **the saponification reaction** between FFA **with base catalyst**, leading to **the** lower **conversion of** WCT **into biodiesel**

. Therefore, it requires at least one-step pretreatment to remove the water content and two-step esterification/ transesterification process to achieve the commercial biodiesel conversion and yield. Alptekin and Canakci (2010) investigated the two-step method to transform chicken oil with 15% FFA to biodie- sel, with 90% of said FFA is esterified into methyl esters in the first step using 20% (wt) sulfuric acid (acting as

an acidic catalyst). Subsequently, the transesterification between chicken oil, methanol, and water occurred in the second step, in the presence of caustic soda (NaOH) as a basic catalyst. The yield of biodiesel after the transesterification step is 87.4%.19 Gürü et al (2010) reported a similar two-step esterification/ transesterification route

> 6**for the conversion of** chicken oil **to biodiesel** and **the optimum fatty acid**

methyl ester (FAME) recovery obtained is 89%.20 However, the use of the two-step process will certainly escalate the processing and maintenance costs, thus lead- ing to economic inability to use chicken tallow as raw material for biodiesel. Correct modification of reaction design and operation will be able to reduce the steps of the downstream process and significantly increase savings in the cost of separation. It is important to single out a suitable, environmentally and economically friendly technique to transform this resource. Methanol under subcritical and supercritical condition has attracted much attention since their degree of hydro- gen bonding, cluster formation, ion solvation, and ion association make them widely known as novel reaction medium.21,22 Ong et al (2013) reported the biodiesel pro- duction from

> 8**Ceiba pentandra (kapok) oil via** catalyst- free **supercritical methanol** (SpCM) **transesterification**

with the optimum FAME yield of 95.5% at 322°C, 16.7 MPa, 476 seconds

> 15**of reaction time and** 30:**1 of methanol to oil** molar **ratio**.16 **Meanwhile**

, Gunawan

> 13**et al (2014**) evaluated the **transesterification of** vegetable oils **waste**

- water sludge to biodiesel using methanol under subcriti- cal condition, with the highest yield of FAME (

> 12**92.67** ± 2.**23%) obtained at 215°C, 6.5 MPa, and** 5:1 of **methanol to lipid mass ratio**

.8 Huynh et al (2012) also reported that the noncatalytic subcritical methanol (SCM) method has successfully converted 90% of activated sludge to FAME.23 In this study, WCT was converted into biodiesel using methanol under subcritical condition. Compared with the conventional technique, SCM is a strategic choice in bio- diesel production since it promotes the simultaneous reaction of esterification and transesterification, it does not require a catalyst, and the time needed to convert raw material to biodiesel is shorter. SCM also operates in moderate temperature and pressure, indicating higher process security and lower capital costs as compared with the SpCM technique. According to Ju et al (2013), the SCM method is tolerant of high water and high FFA con- tent, where it still produces high yield biodiesel regardless of the raw material quality.5 To date, there is no research conducted on the conver- sion of WCT to biodiesel using the noncatalytic SCM technique as well as its optimization approach, eventhough a large amount of WCT is produced annu- ally. This study focuses on the utilization of WCT as raw material to produce biodiesel with high purity and recovery under SCM condition. The optimum point of processing variables (

8temperature, reaction time, and alcohol to fats molar ratio

) was determined using response surface methodology (RSM) optimization approach.

52 | MATERIALS AND METHODS 2.1 | Materials WCT was obtained from a local market in Surabaya, Indonesia. Methanol (99.9%) and

n-hexane (90%) were of technical grade and obtained from Merck, Germany, while reagents used in the

1gas chromatography-flame ionization detector (GC-FID) were

of either HPLC grade or analytical grade. The standard of FAME mixture (37 Component FAME mix, 47885 U) was purchased

4from Supelco (Bellefonte, PA, USA). Ultra-high purity grade nitrogen and helium gases (99

.9%) were supplied

1by Aneka Gas Industry Pty. Ltd., Surabaya. 2.2

| Experimental design and optimization The process optimization using the design of experiments (DOE) was statistically employed using RSM, coupled with the standard design tool known as Circumscribed (α = 1.682) Central Composite Design (CCC-CCD). Three optimized variables were reaction temperature (°C), time (min), and the molar ratio of methanol to WCT (mol/ mol). The coded variables and their correlative values were presented in Table 1. The independent variables are encoded

1into three levels: low (−1), high (+1), and center point

(0), whereas the axial values of this CCC- CCD are coded as ± α (± 1.682). The choice of reaction

7TABLE 1 The coded parameters and their corresponding values in the experimental design Reaction Parameter Encoded Factor Factor

Level −1.682 −1 0 1 1.682 Temperature (°C) X1 83 100 125 150 167 Time (min) X2 3.2 10 20 30 36.8 Molar ratio of methanol to WCT (mol/mol) X3 13.7:1 28:1 49:1 70:1 84.3:1 parameters, as well as its factor level used in the experi- ments, was based on the direct relevance of these param- eters to the process efficiency, safety concern, and its economic feasibility to be scaled up to an industrial scale. The matrix of CCC-CCD in regards to the actual and encoded independent variable is listed along with the results in Table 2. To obtain a good reproducibility, all experiments were conducted in triplicates with their aver- age values regarded as the final result. Six replicates of

8central data point (0,0,0

) were carried out and expressed as individual data for every run. The randomized order of experiments was performed with all the responses fitted into a quadratic regression model, developed using three-way

6analysis of variance (ANOVA), generated by Minitab (version 18.1) with

3 95% confidence level. The goodness-of-fit for the mathemati- cal regression model was statistically assessed by the coef- ficient of

determination (R2) and the

3lack-of-fit sum of squares

. Response surface plots were generated from the regression analysis of

3experimental results by holding one parameter constant while changing the

other two parameters.

3TABLE 2 The experimental design matrix based on CCC-CCD

Input Parameters Response (FAME Yield, %) Run X1 X2 X3 Experimentala Predictiona 1 0 0 0 84.21 84.80 2 0 0 0 84.74 84.80 3 −1 1 −1 66.92 66.59 4 − 1 1 1 77.53 77.41 5 0 0 0 85.85 84.80 6 0 1.682 0 87.31 88.98 7 0 0 0 84.86 84.80 8 1 −1 1 80.99 81.08 9 0 0 −1.682 66.14 66.13 10 1.682 0 0 87.42 88.82 11 0 0 0 86.23 84.80 12 −1.682 0 0 66.09 65.03 13 0 0 0 82.98 84.80 14 0 −1.682 0 79.14 77.82 15 −1 −1 1 75.48 76.23 16 1 1 − 1 91.04 90.04 17 1 −1 −1 78.07 77.95 18 1 1 1 88.74 87.20 19 −1 −1 −1 58.16 59.45 20 0 0 1.682 77.49 77.86 aThe average

1standard error of estimate (SEE) between the experimental result and its corresponding predicted response was

1.06%. The predicted optimization results between the yield of FAME (%) as the response parameter and the indepen- dent

3variables are represented by Equation (1), where Y is the predicted yield

of FAME (%); k0, ki, kii, and kij are the coefficients obtained from the regression for the inter- cept,

3linear, quadratic, and interactions of the indepen- dent variables, respectively; Xi and Xj are the

encoded design parameters. Y ¼ k0 þ ∑ kiXi þ ∑ kiiX2i þ ∑ ∑ kijXiXj 3 3 3 3 (1) i¼1 i¼1 i¼1 j¼1 2.3 | Biodiesel production from WCT using SCM method The biodiesel production from WCT was conducted in a 150-cm3 cylindrical

2reactor, made of stainless steel type 316

and is completed with

2a pressure gauge (0-70 kg

/cm2 scale), thermocouple, and heater (Figure 1). Various amounts of WCT (26.0-70.0 g) and methanol (35.1-108.0 mL) were added into the reactor

1vessel to attain the desired molar ratio of methanol to

WCT. The average

1molecular weight of WCT was calculated using the following equation

: ? Molecular weight of WCT MWCT; g ? 3 mol ¼

1656:1 x 1000 x ðSV − AV Þ (2) where SV is the saponification value

of WCT (mKOH ; moil mg=gÞ;

1and AV is the acid value of

WCT (mKOH ; mg=gÞ:24-26 After the sample was put in the chamber, moil FIGURE 1 Subcritical reactor system. (a) Nitrogen cylinder; (b) safety valve; (c) magnetic-stirring bar; (d) safety valve; (e) reactor; (f) pressure gauge; (g) agitation controller; (h) thermocouple the

2reactor and its cap were then properly tightened using M8 screws

. The reactor was heated with steady heat flow (the rate of temperature increment is 20°

13C/min) until it reached the desired temperature

. To remove air and increase the pressure,

> 1**nitrogen gas at the rate of 3 mL/min was** purged **into the**

system until it reached 45 bar. The reaction begins once

> 10**the desired temperature and pressure** are **achieved. The** mixture **was**

stirred at the constant agita- tion speed (

> 2**500 rpm) to keep the system** homogenous. Throughout **the process, the**

isobaric and isothermal con- dition was maintained by controlling the heating rate and nitrogen gas injection.

> 2**After the** reaction **had completed, the system was** immediately **cooled down to room temperature. The**

sep- aration of FAME was carried out using liquid-liquid extraction. The liquid mixture and 100 mL of n-hexane as the extracting solvent were introduced into a separatory funnel for the extraction process. The mixture was then allowed to settle, and subsequently,

> 11**two layers** were **formed in the separatory funnel. The upper layer**

> 3**contains n-hexane and FAMEs, while the** bottom layer **was glycerol**, unreacted **methanol, and other** byproducts. **The bottom** phase **was**

rewashed for two times using the same amount of

> 3**n-hexane to** confirm **that all** FAMEs had **been extracted**. Afterward, **the FAME was**

obtained by evaporating n-hexane using a rotary evaporator (IKA RV 10B). 2.4 | FAME analysis using GC-FID analysis The

> 5**analysis of** FAME **purity and composition was** per- formed **using GC, completely equipped with a split- splitless injector and**

an FID. The incorporated silica col- umn used was

4DB-WAX capillary column (30 m × 0.25 mm ID × 0.25 μm film thickness, Agilent Technology, CA

). FAME sample (50 mg) was dissolved in 1 mL of 0.01 g/mL internal standard (methyl heptadecanoate, MH) solution; 1 μL of the as-prepared

2sample was injected with a split ratio of 1:50.The

initial column tem- perature was 50°C and held for 15 minutes; then, the temperature was ramped to 220°C at

54°C/min. The final temperature was held constant for another 15 minutes. The

total analysis time was 72.5 minutes. The tempera- ture of

1injector and detector was set constant at 250°C and 260°C, respectively. The velocity of carrier gas (helium, 99.9

%) was adjusted at 30 cm/s.

4External FAME reference (47885 U, containing 37 components FAME standard mix) was used for the

iden- tification of methyl ester peaks in the sample; as well as for the calibration of the instrument along with methyl heptadecanoate solution as internal standard (IS). The purity of FAME in the sample was calculated as follows: FAME purity Fp; % ¼ ∑AFAME − AMH VMHCMH × 100% ? ? ? ? × AMH m (3) where ∑AFAME

6is the area sum of FAME peaks, AMH is the corresponding area of MH peak, VMH is the volume of MH solution (mL), CMH is the actual concentration of MH solution (g/mL), and m is the actual weight of the FAME sample (g). Meanwhile, the

3yield of FAME based on the

lipid weight fraction was determined using the following equation: Yield of FAME ð%Þ ¼ mFAMEx Fp × 100% ? ? (4) mWCT where mFAME is the weight of FAME obtained after the reaction and separation process (g), mWCT is the weight of the initial WCT sample (

1g), and F p is the FAME weight fraction obtained from Equation

(3). 3 | RESULTS AND DISCUSSIONS 3.1 | Characteristics of WCT The characteristics of WCT have been analyzed in accor- dance with the standard method of AOAC 950.46, AOAC 991.36, AOCS Ca 5a-40, and ISO 12966 to determine water content, crude fat content, FFA content, and fatty acid profile, respectively. According to the results, WCT possesses high

> 1**FFA and moisture content, with the** corre- sponding **value of** 0.91% **and**

17.79%. The fatty acid profile in WCT consists of 11.77% tridecanoic acid (C13:0), 5.71% myristoleic acid (C14:1), 5.82% palmitoleic acid (C16:1), 2.19%

> 11**linoleic acid (C18:2**), 33.43% eicosenoic **acid (C20:1**

), 11.15% erucic acid (C22:1), 25.92 % docosadienoic acid (C22:2), 1.23% docosahexaenoic acid (C22:6), 1.37% lignoceric acid (C24:0), and 1.41% nervonic acid (C24:1). WCT also contains quite a significant amount of crude fat, including triglycerides, diglycerides, monoglycerides, phospholipids, and sterols. It covers 72.04% of the total mass of WCT. Canakci and Gerpen (2001) reported that biodiesel production using fat with high FFA content, more or less 1%, through base- catalyzed transesterification leads to the formation of soap due to saponification reaction.27 High water content also interferes the conventional production of biodiesel. Water is able to hydrolyze fats into FFA, which then leads to the saponification reaction. This phenomenon subsequently decreases the

> 2**yield of biodiesel and** causes **difficulty in** the **separation**

. However, in the SCM method, high water content is required to promote the

> 2**simultaneous esterification/transesterification reaction, where FFA is concurrently extracted from the raw** mate- rial **and esterified into fatty esters**.8 **Ju et al** (2013) **also** described **that** under **subcritical**

condition, high water, and FFA content can be tolerated and

> 2**still able to** yield a **high amount of biodiesel**

.5 3.2 | Reaction parameter study The interaction effect between two

> 3**reaction parameters on the yield of FAME** is shown **in**

Figure 2A(i) to (iii). The experimental results revealed that the increase of temperature from the lowest level (83°C) to level 1 (150°C) greatly improve the FAME yield regardless of the processing time. Reaction temperature played a major role in influencing the chemical reaction inside the ves- sel. Both esterification and transesterification reactions are known as reversible and highly endothermic.

> 1**Based on the Arrhenius law, the** escalating reaction **temperature**

affects the

3**rate constant and** stimulates **the reaction to** shift to **the** product (**right**-hand) **side**

. At room temperature,

1**both water and** methanol **have low miscibility with**

WCT. However, their dielectric con- stant was significantly reduced at high temperature, ren- dering the mixture system to be more homogenous. The elevation of temperature caused a weakening of hydrogen bonding between the water molecules as well as that of the hydroxyl group in methanol, magnifying their misci- bility in the lipid phase.28 Chin et al (2009) reported that that higher temperature in the reaction process causes an increase in the intrinsic reaction rate constant.29 Further- more, from the kinetic perspective, enhanced FAME yield at a higher level of temperature was likely attributed to the higher rate of mass transfer and diffusivity between the reactants, generated due to higher miscibility among them. Another reason is the existence of the water inside the system, which hydrolyzed the lipids to form FFA. As the hydrolysis progresses, more FFA formed increased the

1**miscibility between water and lipid** phase, **and** cer- tainly, the **diffusion rate**

of reactants. FFA is also known to be highly reactive with methanol as compared with the acyl glycerides, which lead to a better yield of FAME. High water content in the WCT might as well promote the esterification/transesterification reaction between FFA/triglycerides and methanol to form biodiesel. The

2**rate of dissociation of water into** H_3O^+ **and OH**

− signifi- cantly enhances by increasing the temperature. The pres- ence of hydronium ion

21**acts as an** acid catalyst **for the esterification reaction**

between FFA

2**with methanol, while the** hydroxide ion **acts as the base catalyst for the**

FIGURE 2 The

3**yield of FAME (%) based on** (a) **the experimental results, (b**

) the 3D

2response surface plot, with the interaction between (i) temperature and time, (ii) temperature

and molar ratio of methanol to WCT, and (iii) time and molar

3ratio of methanol to WCT [Colour figure can be viewed at wileyonlinelibrary.com

] transesterification of WCT and methanol.

2Therefore, with a sufficient number of H3O+ and OH− ions in the

system, this simultaneous reaction can occur more intensively to produce a higher

2yield of FAME.8 It was observed from Figure 2A(i) that the

biodiesel yield reached the plateau point near the highest level of temperature. On top of that, in some levels of reaction time (−1.682 and −1), a further escalation in temperature resulted in the slight decline of FAME yield. Wang et al (2018) also reported that a positive effect on the FAME yield was obtained by increasing the temperature from 50°C to 90°C. Further rise of reaction temperature did not increase the FAME yield significantly.30 This phe- nomenon was

1likely due to the thermal decomposition caused by the carbon-chain splitting into shorter ones. The

thermal decomposition product consists of smaller molecular weight of fatty esters in the range of C13 to C14. Marulanda

4et al (2010) and Shin et al (2011) also stated that

increasing temperature might improve the chance of partial degradation to occur during the process, particularly for the unsaturated FAMEs in the mix- ture.31,32 Ortiz-Martinez et al (2019) mentioned that although high temperature generally increases FAME yield, thermal decomposition of the product can occur above certain values.33

5The effect of reaction time was investigated at five dif- ferent levels

of the time period from 3.2 minutes (−1.682) to 36.8 minutes (1.682). Figure 2A(i) and (iii) showed that either in the

1constant temperature or molar ratio of meth- anol to WCT, a

moderate

4**increase of FAME yield** was observed **by** prolonging **the duration of**

1**reaction time from the lowest to the highest level**

. Allowing longer con- tact between the subcritical methanol, water, and lipid phase

5**ensures the conversion of triglycerides and FFA into**

FAME through the simultaneous esterification- transesterification process. However, even though the duration of reaction gave an advantageous effect on the FAME yield, its significance was incomparable to the effect of temperature. The required stoichiometric ratio of methanol to lipid in the production of biodiesel via transesterification method to form

213 **moles of** fatty **esters and 1 mole of**

glyc- erol is 3:1. The transesterification itself is a reversible reaction; thus, it is commonly carried out by using the excess alcohol to purposely shift the chemical equilibrium to the right-hand side to ensure high conversion of FAME within a short time.34,35 In accordance with the experi- mental results, the addition of excess methanol to the sys- tem from the ratio of 13.7:1 (−1.682) to 49:1 (0) greatly increases the FAME yield by 1.5 folds. It is likely due to the

4**more frequent** interaction **between the lipid and methanol, triggering the formation of FAME**. Gunawan **et al** (2014) also mentioned **that excess methanol**

to WCT molar

5**ratio seems to be favorable** toward **the** biodie- sel **yield to a certain extent**

.8 However, as seen in Figure 2 A(ii) and (iii), a remarkable declining trend in FAME yield was monitored by the further addition of excess methanol to the highest level of the molar ratio of meth- anol to WCT. Encinar

14**et al** (2005) **reported that** further **addition of**

excess methanol tends to give

14**a negative** response **on the product yield**

since the presence of excess glycerol reversed the

1transesterification to the reactant side. An increase in the concentration of FAME and

glyc- erol in the system during reaction will lead to the recom- bination of products to monoglycerides, resulting in the lower yield.36

1Thoai et al (2017) also mentioned that high

methanol to oil molar ratio gives a lower mono-, di-, tri- glycerides concentration that makes disadvantages

1for the reaction since both alcohol and oil are needed to promote the reaction rate.37 Moreover

, higher methanol content in the system would also drive the extraction of compounds with higher polarity, namely phenols and proteins which hindered the fatty esters formation.23 As a matter of fact, the enhanced amount of excess methanol above the optimum value will not only decrease the product yield but also escalate the raw material and recti- fication processing costs. 3.3 | Statistical analysis and development of optimization model using response surface methodology (RSM) Statistically, RSM depicts the relationship between sev- eral independent input variables and single or multiple responses. Its primary goal is to employ a series of designed experiments to determine the optimum value of the response variable. SCM technique provides many benefits as compared with the conventional ones, since it reduces the reaction time, eliminates the needs of catalyst as well as the pretreatment and separation cost. However, this method

17is an energy-intensive process which requires the use of

high temperature and pressure. Therefore, the optimization process including the statisti- cal analysis and development of the mathematical model are significant for the implementation of this technique on the industrial scale. For this very reason, RSM was conducted to deter- mine the optimal

1conditions for the production of FAME by integrating three important variables

(

22temperature, reaction time, and methanol to WCT molar ratio

) simul- taneously. Circumscribed type of CCD (CCC-CCD) was used to design the experimental input parameters. Table 2 showed the correlation between the response and the sets of coded input parameters. The response of the experimental design was the yield of FAME (%), and the average

1standard error of estimate (SEE) between the experimental and predicted

results were found to be fairly close to each other, with the value of 1.06% (n = 20). As shown in Equation (1), the second- order polynomial equation, as a function of the indepen- dent variables, was suggested by the RSM using the least square analysis. The result of the statistical ANOVA applied in deter- mining the significance of the independent variable indi- vidually, quadratically as well as their interactions were presented in Table 3. Probability of error value, known as P value, is a parameter to analyze the significance of each regression coefficient. Smaller P value indicated that the term was statistically more significant. Based on the ANOVA results, the mathematical model suggested that all the terms except that of reaction time ((X2)2, P value >0.05) were significant. All linear terms were equally sig- nificant, while the similarly notable quadratic terms were temperature (X1)2 and methanol to WCT molar ratio (X3)2. The two-way interactions were found to be promi- nent between (X1)(X2),(X1)(X3), and(X2)(X3), with the sig- nificance order of temperature/molar ratio of methanol to WCT > time/molar ratio of methanol to WCT > temperature/time. By inserting the coefficient values of the significant parameters to Equation (1), the mathe- matical model can be expressed by the given equation below:

7**TABLE 3 The fitted values of regression coefficients** and their significance **for the calculation of yield** of FAME **Term Coefficient SE Coefficient T**-Value **P** Value **Constant**

84.800 0.581 145.96 <0.0001 X1 7.074 0.385 18.35 <0.0001 X2 3.316 0.385 8.60 <0.0001 X 3 3.487 0.385 9.05 <0.0001 X 1 2 −2.784 0.375 −7.42 <0.0001 X22 −0.495 0.375 −1.32 0.2160 X32 −4.528 0.375 −12.07 <0.0001 X1X2 1.239 0.504 2.46 0.0340 X1X3 −3.414 0.504 −6.78 <0.0001 X2X3 −1.491 0.504 −2.96 0.0140 Yield of FAME ð%Þ; Y ¼ 84:8 þ 7:074ðX1Þ þ 3:316ðX2Þ þ 3:487ðX3Þ − 2:784ðX1Þ2 − 4:528ðX3Þ2 þ 1:239ðX1ÞðX2Þ − 3:414ðX1ÞðX3Þ − 1:491ðX2ÞðX3Þ (5)

3**where Y is the yield of FAME** (%); X1, X2, **and** X3 **are the** coded value **of input variables (−1**.682, −**1**

, 0, 1, 1.682). Equation (5) showed that the coefficients of intercept, linear variables (X1, X2, X3), and interaction variables of temperature and time ((X1)(X2)) indicated the favorable effect to the FAME yield. On the other hand, the coefficient of quadratic variables ((X1)2, (X3)2) and the other two-way interaction variables of either tempera- ture or time with the methanol to WCT molar ratio ((X1)(X3),(X2)(X3)) gave the antagonistic effect to the response. Table 4 showed the analysis of goodness of fit for the mathematical regression model using ANOVA. As pre- sented in Table 4, the R-squared

1**value of the** coded **model (Equation 5**) was **0**.9866, referring **that 98**.66% **of the**

experimental data were able to be reasonably interpreted by the second-order polynomial equation above (Equation 5). The predicted and actual response of yield of FAME was also in a good agreement, pointed by the

3**value of adjusted and predicted R-squared which** were **close to unity (0**.9746 **and 0**.9258, **respectively**

). Based on the

3**lack of fit test** results, **the P value of the** coded **model is 0**

.235. This value indicated that the equation derived by ANOVA was well-fitted to the experimental data. In the lack of fit test, the P value higher than 0.05 is highly desirable since P value lower than 0.05 repre- sents that there might be the interaction of input variable and response that is not considered by the model. From the results of the statistical ANOVA, the model is deemed adequate to represent all the independent variance assumption. The 3D response surface plots of the interactions between the two independent variables were presented by Figure 2B(i) to (iii). As shown in Figure 2B(i), the increase in the level of both temperature and time gave an overall positive

3**influence on the yield of FAME**. At **the** fixed molar **ratio of methanol to**

WCT (49:1), the yield of FAME rose sharply along with the increment of temperature from 83°C (−1.682) to 150°C (1) at constant reaction time; however, further elevation in temperature to the highest level (1.682) leads to a reduc- tion of the yield of FAME. A similar trend was moni- tored for the other level of reaction time at the same profile of temperature increment. The response was monitored to be rapidly escalated before it reaches a stagnant line close to the highest level of temperature. It is also evident from the figure that the temperature gives a more significant effect

3**on the yield of FAME** than **reaction time**

. Figure 2B(ii) depicted the correlation between tem- perature and

16**methanol to** WCT **molar ratio** to **the** FAME **yield**. Based on **this**

response surface plot, it can be seen that the maximum yield of FAME was

19**TABLE 4** The **analysis of variance (ANOVA) for the** fitted **regression model**

Source DF **Sum of Squares Mean Squares Model**

9 1498.30 166.478 X1 1 683.49 683.493 X2 1 150.13 150.130 X3 1 166.07 166.074 X12 1 111.73 111.734 X22 1 3.54 3.535 X32 1 295.52 295.517 X1X2 1 12.28 12.284 X1X3 1 93.27 93.265 X2X3 1 17.79 17.794 Error 10 20.29 2.029 Lack-of-fit 5 13.49 2.698 Pure error 5 6.80 1.361 Total 19 1518.60 R-squared (R2) 0.9866 Adjusted R2 0.9746 F-Value 82.04 336.81 73.98 81.84 55.06 1.74 145.63 6.05 45.96 8.77 1.98 Predicted R2 P-Value <0.0001 <0.0001 <0.0001 <0.0001 <0.0001 0.2160 <0.0001 0.0340 <0.0001 0.0140 0.235 0.9258 obtained at the middle level of both factors. Further

12**increase in the molar ratio of methanol to**

WCT and temperature resulted in a gradual decrease and in signif- icant increase of the FAME yield, respectively. A consis- tent effect of the molar ratio of methanol to WCT on the yield was also monitored in Figure 2B(iii), with the factor gave maximum response at the middle level, while prolonged reaction time caused a steady increase of the yield of FAME. The optimization of biodiesel production was per- formed to

find the optimum levels of combined process variables at which the maximum response is attained. The solution with these independent variables (tempera- ture,

> 22reaction time, and methanol to WCT molar ratio

)

> 1were generated by Minitab software (version 18.1) based on the model obtained and the experimental data input (Table 2). The

optimal variable of the transesterification process of WCT into biodiesel are as shown in Figure 3: temperature of 167°C (1.682), the reaction time of 36.8 min (1.682), and

> 13methanol to WCT molar ratio of 42.7:1

(−0.5266). The predicted yield of FAME under this opti- mum condition was 97.76%

> 3with model desirability of 1.00. Three replicated experiments were conducted using these optimal variables to verify the reliability of the

pre- diction. Based on the experimental results, the optimum yield of FAME was 98.43 ± 0.22% with the purity of 97.17%. The result at the optimum point indicated that the experimental and predicted values were close to each other, with the error of only 0.89%. Thus, it can be con- cluded that the established model is highly reliable and possesses adequate accuracy in predicting the biodiesel

> 1yield using the reaction parameters within the levels

. This optimization results with high temperature (167°C) and low molar ratio of methanol to WCT (42.7:1) are gener- ally highly desirable in the industries since on the process economic viewpoint, the operating expenditures at high temperature are much less important than the cost of raw materials.38

> 13.4 | Chemical composition of WCT-based biodiesel The

resulting biodiesel

> 2obtained at the optimum condi- tion (temperature of 167°C, the reaction time of 36.8 min, and the molar ratio of methanol to

WCT of 42.7:1)

> 1was analyzed by using GC-FID for its purity and chemical composition. The

purity of the FAME obtained from the analysis was 97.17%, higher than that required in ASTM D6751 (96.5% purity). The chemical composition of FAME was

> 4**obtained by comparing the** peaks of **methyl** esters **in the chromatogram with** that of **the external FAME standard (47885 U, containing 37 components FAME standard mix**). There are 11 **identified peaks**

in the chromatogram, namely

> 23**tridecanoic acid methyl ester (C13:0**), myristoleic **acid methyl ester (C14**:1), palmitoleic **acid methyl ester** (C16:**1**), both **cis**

- and trans-linoleic

> 9**acid methyl ester (C16**:2), eicosenoic **acid methyl ester** (C20:**1**), erucic **acid methyl ester** (C22:1), docosadienoic **acid methyl ester** (C22:2), docosahexaenoic **acid methyl ester** (C22:6), lignoceric **acid methyl ester** (C24:0), **and** nervonic **acid methyl ester** (C24:**1**

). 3.

> 11**5 | Properties of** WCT-based **biodiesel The properties of**

WCT-based methyl esters are presented in Table 5. These properties are required to meet interna- tional standards such as ASTM D6751 to be defined as biodiesel. The results were also compared with the properties of biodiesel prepared from chicken fat using the conventional method19

> 1**and diesel fuel** specifications (**ASTM D975-08**

). As seen in Table 5, the flashpoint of FIGURE 3

> 1**Response optimization plot of the three independent reaction** parameters (**D —composite desirability, y = predicted response, d = desirability**

) [Colour figure can be viewed at wileyonlinelibrary.com] TABLE 5

> 1**Fuel properties of** WCT-**based biodiesel** and its comparison **with ASTM D6751**, diesel fuel (**ASTM D975-08**

), and a similar study by Alptekin and Canakci (2010)19 Properties Density (at 15°C) Kinematic

18**viscosity (at 40°C) Flash point** Acid value **Calorific value Methods ASTM D4052 ASTM D445 ASTM D93 ASTM** D664 ASTM **D240**

Unit g cm−3 mm2 s−1 °C mg KOH/g MJ kg−1 WCT-Based Methyl Ester 0.869 2.13 97.2 0.19 39.752 Chicken Fat Methyl Ester19 0.883 4.94 171.8 0.22 40.173 ASTM D6751 - 1.9 – 6.0 93 min 0.50 max - Diesel Fuel (ASTM D975-08) - 1D: 1.3-2.42D: 1.9-4.1 1D: 38 min2D: 52 min - - WCT-based biodiesel is close to the minimum value and the adjusted coefficient of determination close to required by the ASTM D6751, indicating the low activa- unity (0.9746). The properties of WCT-based biodiesel tion energy needed for combustion and feasibility of this

1**are in accordance with ASTM D6751 and ASTM D975**

- fuel to be used in the diesel engine without extensive 08. This study indicated that SCM is a prospective modification. This goes as well for the kinematic viscos- technique to replace the traditional process in the utili- ity, which is one of the most critical properties in biodie- zation of WCT as

15**low-cost raw** materials **for biodiesel** sel that **is** related **to the**

fluidity performance. According production since it is more sustainable and environmen- to ASTM D975-08 standards, WCT-based biodiesel pos- tally friendly compared with the latter. Further study on sesses kinematic viscosity that is comparable to the diesel the use of the SCM method to convert WCT to FAME is fuel, emphasizing the possibility of the product to be used still being done to determine its feasibility to be scaled widely as a petrodiesel blend. Both density and acid value up to the mass production. are also suitable for the standards. The calorific value of WCT-based biodiesel was found to be comparable to the study conducted by Alptekin and Canakci (2010),19 but slightly lower than the usual petro-diesel (42-46 ACKNOWLEDGEMENTS MJ/kg).39 Based on the summarized fuel properties This research did not receive any specific grant from (Table 5), it can be concluded that the measured proper-

1**funding agencies in the public, commercial, or not-for**

- ties met the requirements, indicating that the biodiesel profit sectors. produced from WCT can be used as an energy replace- ment for diesel fuel. O R C I D

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4 | CONCLUSIONS WCT is an appealing alternative resource to produce REFERENCES biodiesel using the catalyst-free subcritical methanol 1. Ahmad T, Danish M, Kale P, et al. Optimization of process var- (SCM) method. RSM and three-way ANOVA have been iables for biodiesel production by transesterification of flaxseed well employed to design, predict, and optimize the oil and produced biodiesel characterizations. Renew. Energy. experiments, by integrating three independent variables 2019;139:1272-1280. (temperature, reaction time, and methanol to WCT 2. Moser BR. Preparation of fatty acid methyl esters from hazelnut, molar ratio). High biodiesel recovery of 98.43 ± 0.22% high-oleic peanut and walnut oils and evaluation as biodiesel. with high purity (97.17%) was obtained as the maximum Fuel. 2012;92(1):231-238. result in this optimized SCM process at 167°C, 36.8 minutes, and methanol to WCT molar ratio of 42.7:1. 3. Christopher LP, Hemanathan K, Zambare VP. Enzymatic bio- The predicted value of FAME yield in this optimum diesel:

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