

Optimization Process of Biodiesel Production From Nyamplung Seed (*Calophyllum inophyllum* L) Using In Situ Process and Ultrasonic Assisted

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Abstract :

Nyamplung seed (*Calophyllum inophyllum*) contain high oil between 40-73%, so it potential as a raw material in biodiesel production. It The objective of this research to optimization of biodiesel production by in situ process and assisted ultrasonic. Optimization process use central composite design methods. Variables were investigated temperature, catalyst concentration (methanol basis), and the weight ratio of methanol by weight nyamplung. The ultrasonic wave was produced by ultrasonic cleaner unit with frequency 40 KHz, degassing time 10 minutes and reaction time 60 minutes. The results of research obtained Optimum conversion of biodiesel production from nyamplung seed with assisted ultrasonic was 64.429%. The optimum condition are 1:5.9116, 2.556% and 46.2°C for ratio weight of nyamplung seed to

$Y = 69.609 - 0.7586X_1 + 1.9051 X_2 + 3.2476 X_3 - 2.08 X_1 X_2 - 3.665 X_1 X_3 - 2.865 X_2 X_3 - 10.6085 X_1^2 - 6.224 X_2^2 - 6.747 X_3^2$
methanol, catalyst concentration and temperature respectively. Mathematic modeling for describe in this process like expressed;

Keywords: biodiesel, conversion, nyamplung (*Calophyllum Inophyllum*), ultrasonic assisted

1. Introduction

Fossil fuel consumption continues to raise nationwide average 7% per year, estimated in 2020 the consumption of fossil fuels reached 34 million kilo liters. Consumption of fossil fuels percentage is the largest and constantly increasing. In 2008 there was a surge in crude oil around \$ 1000/barrel, this is causing a crisis in the world and in Indonesia. World oil prices in 2011 USD 107.46/barrel and in 2012 USD 110.27/barrel [1]. The increase in oil prices could encourage the energy crisis is more severe, so that alternative energy is needed to overcome.

Indonesia government efforts to solve energy crisis is with the establishment of policies. Indonesia government policies set out in Presidential Decree no. 5 of 2006 regarding the national energy policy, Presidential Instruction No. 1 of 2006 regarding the procurement and use of biofuels as alternative energy, as well as the Presidential Decree no.10 of 2006 regarding the formation of a national policy for the development of biofuels [2].

Indonesia has great potential to produce alternative energy instead of fossil fuels derived from renewable natural resources in the form of plants. Vegetable oil (vegetable) and animal fats as an alternative energy source can be processed into biodiesel. Biodiesel is a methyl ester compound result from the transesterification reaction of triglycerides derived from vegetable oils or animal fats [3-6]. Biodiesel has several advantages if compared with conventional fuel. The advantages are environmentally friendly, good lubricating power, fewer emissions and a relatively clean burning character. In addition to these advantages, the use of biodiesel also provides benefits to the vehicle engine maintenance [7,8]. Processing of biodiesel from renewable raw materials have been widely applied in various countries including countries of the United States uses soybean oil, coconut oil (coconut oil) in the Philippines and Malaysia using CPO (Crude Palm Oil). Indonesia uses Crude Palm Oil, nyamplung oil and castor oil (*Jatropha*) for raw material in biodiesel production.

Calophyllum inophyllum L (nyamplung) is a medium-sized to large evergreen tree that averages 8–20 m (25–65 ft) in height with a broad spreading crown of irregular branches (like Fig 1). The tree supports a dense canopy of glossy, elliptical leaves, fragrant white flowers, and large round nuts. It grows along coastal areas and adjacent lowland forests, although it occasionally occurs inland at higher elevations. It is native to east Africa, India, Southeast Asia, Australia, and the South Pacific. It has been widely planted throughout the tropics and is naturalized in the main Hawaiian islands. One large brown seed 2–4 cm (0.8–1.6 in) in diameter is found in each

fruit. Seeds are prepared by cleaning off the skin and husk from the shell of the seed; there are 100–200 seeds/kg (45–90 seeds/lb), with shells intact but husks removed [9,10]. Nyamplung seed figure like presented in figure 2.



Figure 1. Nyamplung tree and seed



Figure 2. Ripe fruit (left), cracked shell showing seed kernel inside (middle), and dry seed (right).

Biodiesel from nyamplung seed (*Calophyllum inophyllum* L) is one of the alternative fuels from vegetable oils other than castor oil, palm oil, sunflower seeds, and used frying oil (UFO). Nyamplung plants (*Calophyllum inophyllum*) is one of the alternative feedstock biodiesel that has considerable potential. Nyamplung can produce oil per year at 20 tons / ha. Nyamplung seed contain high oil between 40-73%. One liter of nyamplung oil can be produced from 2.5 kg of nyamplung seed, while the castor requires 4 kg to produce one liter of oil [10,11].

The main reaction to produce biodiesel is esterification and transesterification reactions [3-8]. In biodiesel production can be done with the conventional method and the method of ultrasonic waves. The process of making the conventional method relatively slow (almost 4-5 hrs), need more catalyst and alcohol and furthermore the reaction is not fully completed [5]. Moreover, the high formation of by-product gives low yield of biodiesel and during the conversion process not all fatty acid chains are turned into biodiesel which consequently reduces the quality and the yield. The current conventional process only can achieve maximum yield of 85% during esterification reaction [4-6]. Therefore, an alternative process is really required to reduce the processing time, to increase the yield, to lower amount un-reacted methanol and also catalyst, and to increase the mass transfer.

Ultrasound (~20 kHz to 10 MHz) is cyclic sound pressure with a frequency greater than the upper limit of human hearing. The important use of ultrasound is to create bubble cavitations which can be used as replacement of mechanical agitation by stirrer, and therefore it is required for enhancing the esterification reaction. This paper describes the potential use of ultrasound for assisting biodiesel production using nyamplung seed In biodiesel production by assisted ultrasonic wave takes up to 1 hour [12-13].

Ultrasonic technology has advantages like minimum energy consumption and can be reduced time operation. So, these technologies promising because it offers the potential for shorter reaction cycles leading to the formation of chemical plants are smaller and cheaper. Excellence performance using ultrasonic solvent extraction method compared to conventional methods has also been reported by several researchers [13-15].

2. Material and Methods

2.1. Apparatus and Materials

Raw materials using in this experiment are nyamplung seed collected from Banyumas, Central Java. Nyamplung seed crushed into powder. Methanol used has industrial grade and H_2SO_4 as catalyst has analytical grade from MERCK. Analytical chemical was used like as; ethanol 95% (technical grade), KOH (analytical grade), HCl (analytical grade), and Dietil Eter (analytical), PP indicator. Chemicals have analytical grade obtained from MERCK. Apparatus using in this experiment are ultrasonic cleaner according to BRANSON, aluminum foil, 250 ml Erlenmeyer PYREX. The experimental set up shown in Figure 3. Ultrasonic cleaner has frequency 40 kHz.



Figure 3. Schematic diagram of the ultrasonic cleaner

2.2. Experimental Procedure

25 grams of nyamplung seed powder mixed with methanol and sulphuric acid as catalyst in erlenmeyer. Then, the Erlenmeyer is inserted into the round pedestal ultrasonic cleaner. Operate the ultrasonic cleaner for 60 minutes at a predetermined variable. Erlenmeyer took from the ultrasonic cleaner and then cooled to room temperature. The mixture is distilled to separate the biodiesel with methanol. Variables used in this experiments like presented in Table 1. Fixed variable was used weight nyamplung seed powder 25 grams, size 200 mesh powder nyamplung, frequency of 40 kHz ultrasonic waves, reaction time of 60 minutes, set Degas 10 minutes.

2.3. Analysis Method

Product was analysis of volume of product, number of acid, viscosity, density and gas chromatography. Volume and density of product was used for yield calculation. Yield of biodiesel was calculated with equation 1. Yield of biodiesel will be processed by the Central Composite Design (CCD) method to evaluate the response and the observation of the influential variables. The analysis process used Statistica 6 software about mathematical model, statistic analysis, analysis of variance and profile of yield versus variable processes. The experiment then compared with conventional processes.

$$\text{Yield (\%)} = \frac{\text{weight of biodiesel}}{\text{weight of nyamplung seed}} \times 100 \quad [1]$$

3. Result and Discussion.

3.1. Experiments Data

The results of data processing are presented in Table 1. Central Composite Design used 2 blocking, 3 variable processes. The experiment 1-9 classified in 1 blocking and 10-16 classified in 2 blocking. X_1 is coding of ration weight of nyamplung seed to methanol volume. X_2 is coding of catalyst concentration (methanol based). X_3 is coding of temperature operation. The value that used for experiment can be calculated with equation 2-4. The real condition of variable can be calculated with equation 2-4.

Table 1. The result and experiments design of CCD methods

| Run | Block | X ₁ | X ₂ | X ₃ | ρ | Y (%) |
|-----|-------|----------------|----------------|----------------|-------|-------|
| 1. | 1 | -1 | -1 | -1 | 0.880 | 29.16 |
| 2. | 1 | -1 | -1 | +1 | 0.881 | 55.28 |
| 3. | 1 | -1 | +1 | -1 | 0.902 | 48.56 |
| 4. | 1 | -1 | +1 | +1 | 0.883 | 42.48 |
| 5. | 1 | +1 | -1 | -1 | 0.891 | 46.92 |
| 6. | 1 | +1 | -1 | +1 | 0.882 | 37.64 |
| 7. | 1 | +1 | +1 | -1 | 0.883 | 37.26 |
| 8. | 1 | +1 | +1 | +1 | 0.884 | 37.26 |
| 9. | 1 | 0 | 0 | 0 | 0.892 | 51.88 |
| 10. | 2 | -1.68 | 0 | 0 | 0.885 | 38.92 |
| 11. | 2 | +1.68 | 0 | 0 | 0.887 | 42.12 |
| 12. | 2 | 0 | -1.68 | 0 | 0.889 | 45.44 |
| 13. | 2 | 0 | +1.68 | 0 | 0.905 | 62.76 |
| 14. | 2 | 0 | 0 | -1.68 | 0.889 | 42.44 |
| 15. | 2 | 0 | 0 | +1.68 | 0.889 | 62.52 |
| 16. | 2 | 0 | 0 | 0 | 0.888 | 86.96 |

Where X1 : (-1) 1:5 (0) 1:6 and (+1) 1:7
 X2 : (-1) 2% (0) 2.5% and (+1) 3%
 X3 : (-1) 40 ° C, (0) 45 ° C, and (+1) 50 ° C.
 (-1) : below value (bv)
 (0) : central value (cv)
 (+) : upper value (uv)

$$X_1 = \frac{[R-6]}{1} \quad [2]$$

$$X_2 = \frac{[K-2.5]}{0.5} \quad [3]$$

$$X_3 = \frac{[T-45]}{5} \quad [4]$$

3.2. Statistics Analysis

The results of statistic analysis include mathematical model, t test, analysis of variance, pareto analysis and validation of mathematical model. Polynomial equations to mathematical model the CCD as follows:

$$Y_u = \beta_0 + \sum \beta_i X_{ui} + \sum \beta_{ii} X_{ui}^2 + \sum \sum \beta_{ij} X_{ui} X_{uj} + \epsilon \dots \dots \dots (5)$$

Where:
 Yu = predicted response to u, u: 1, 2, 3,, n
 β₀ : average of yield
 β_i: linear coefficient, β_{ii}: squared term coefficient, β_{ij}: interactions variable coefficient
 xi: non dimensional number of independent variables

In this experiment the number of independent variables was 3, so the polynomial equation becomes

$$Y_u = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 \quad [6]$$

The results of analysis regression for equation 6 like presented in equation 7. Data that used for this analysis presented in table 1. In the mathematical model shows that the coefficient of linear variable-value range, most of the variables X₃ and X₁ and X₂ variables followed. Variables X₂ and X₃ are positive which means the larger value of this variable, and then the yield of biodiesel obtained the greater. The first variable is the inverse of the variables X₂ and X₃. Increased temperatures will lead to increasing the solubility also increases the reaction rate. Increasing of catalyst concentration can enhance the catalytic activity of direct transesterification reaction, thereby increasing the amount of methyl ester.

$$Y = 69.609 - 0.7586 X_1 + 1.9051 X_2 + 3.2476 X_3 - 2.08 X_1 X_2 - 3.665 X_1 X_3 - 2.865 X_2 X_3 - 10.6085 X_1^2 - 6.224 X_2^2 - 6.747 X_3^2 \quad [7]$$

Coefficient values in equation 7, further evaluated the t test and its variations. For the t test using an $\alpha = 0.05$. The results of such analysis are presented in Tables 2 and 3. In Table 2 show that the coefficients of variables X_2 and X_3 have a value of t (5) is greater than the value of p, but the value of t (5) is still in the range t test.

Table 2. Results of regression coefficient and t test

| Factor parameter | Regression coef | Standar error | t(5) | p | -95% Cnf. Limnt | +95% Cnf. Limnt |
|------------------|-----------------|---------------|----------|----------|-----------------|-----------------|
| Mean | 69.6093 | 8.557627 | 8.13419 | 0.000456 | 47.6113 | 91.60741 |
| Blocking | 5.7113 | 3.049887 | 1.87263 | 0.120007 | -2.1287 | 13.55130 |
| X_1 | -0.7586 | 3.212522 | -0.23613 | 0.822701 | -9.0166 | 7.49949 |
| X_1^2 | -10.6085 | 3.757510 | -2.82327 | 0.036966 | -20.2674 | -0.94947 |
| X_2 | 1.9051 | 3.212522 | 0.59302 | 0.578971 | -6.3530 | 10.16315 |
| X_2^2 | -6.2244 | 3.757510 | -1.65653 | 0.158516 | -15.8834 | 3.43457 |
| X_3 | 3.2476 | 3.212522 | 1.01093 | 0.358442 | -5.0104 | 11.50568 |
| X_3^2 | -6.7474 | 3.757510 | -1.79571 | 0.132487 | -16.4064 | 2.91158 |
| $X_1 X_2$ | -2.0800 | 4.279292 | -0.48606 | 0.647476 | -13.0803 | 8.92027 |
| $X_1 X_3$ | -3.6650 | 4.279292 | -0.85645 | 0.430872 | -14.6653 | 7.33527 |
| $X_2 X_3$ | -2.8650 | 4.279292 | -0.66950 | 0.532843 | -13.8653 | 8.13527 |

Table 3. Result of analysis of variance

| Factor parameter | SS | Degree of Freedom | MS | F | p |
|------------------|----------|-------------------|----------|----------|----------|
| Blocking | 513.734 | 1 | 513.734 | 3.506746 | 0.120007 |
| X_1 | 8.168 | 1 | 8.168 | 0.055756 | 0.822701 |
| X_1^2 | 1167.718 | 1 | 1167.718 | 7.970840 | 0.036966 |
| X_2 | 51.520 | 1 | 51.520 | 0.351674 | 0.578971 |
| X_2^2 | 402.004 | 1 | 402.004 | 2.744082 | 0.158516 |
| X_3 | 149.718 | 1 | 149.718 | 1.021978 | 0.358442 |
| X_3^2 | 472.397 | 1 | 472.397 | 3.224578 | 0.132487 |
| $X_1 X_2$ | 34.611 | 1 | 34.611 | 0.236256 | 0.647476 |
| $X_1 X_3$ | 107.458 | 1 | 107.458 | 0.733507 | 0.430872 |
| $X_2 X_3$ | 65.666 | 1 | 65.666 | 0.448235 | 0.532843 |
| Error | 732.494 | 5 | 146.499 | | |
| Total SS | 2894.021 | 15 | | | |

Pareto diagram is histogram of the data is sorted based on categories of greatest to smallest. It is based on the principle that there are many factors that affect anything but just a few important factors are taken into account the factors that cause the most significant impact. Thus, pareto diagrams can assist in focusing efforts on the most important thing alone [17], [18]. Pareto diagram of data processing results in this study are presented in Figure 4. Pareto analysis results showed that a linier variable of X_1 has smaller value. So this variable can be neglected in optimization process. Pareto chart show variable of ratio nyamplung to methanol (Q) has a histogram that excess the line $p = 0.05$. The others variable have histogram don't cross the line $p = 0.05$.

Mathematical model in 7 equations was validated with experiments data. The results shown in Figure 5. Mathematical models that is less valid because the experimental data coincide with the results of the calculation very little.

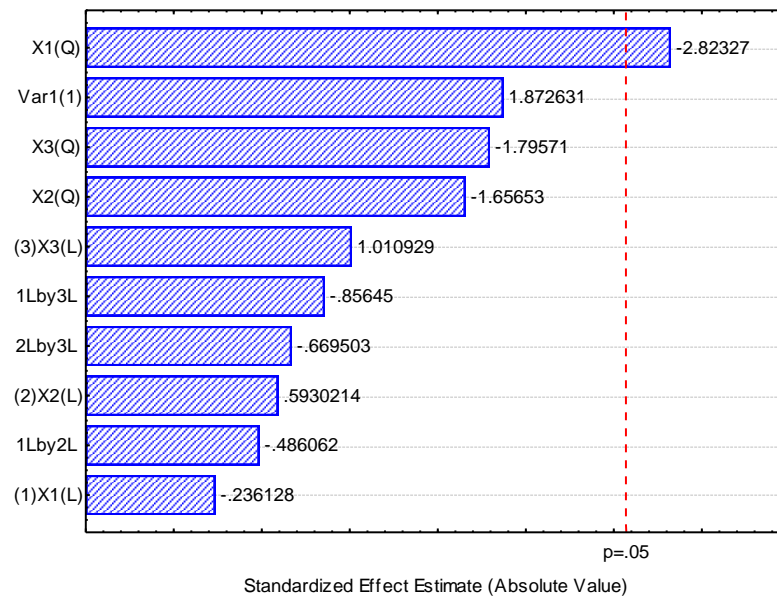


Figure 4. Pareto chart

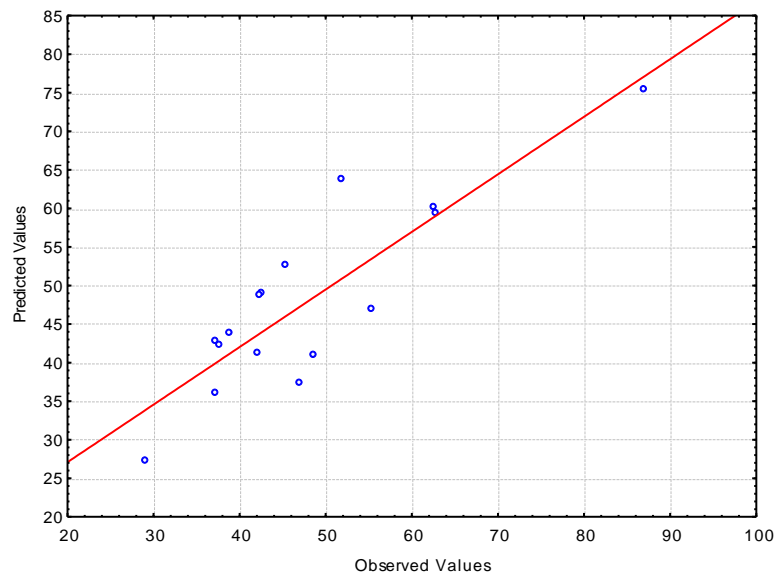


Figure 5. Graph of validation model mathematics

3.3. Optimization Results

Optimum operating conditions sought by looking at optimization 3-dimensional graph and surface contours graph. Optimization of three-dimensional graph consists of axis x, y, and z, where x and y axis is the variable being tested while the z-axis shows the value of the conversion can be achieved from the interaction of two variables tested so from optimization of three-dimensional graphics can be seen the value of the conversion which can be achieved from the interaction of two variables that are tested and as well as optimal conditions. Surface contour graph consists of axis x and y, where x and y axes are the variables tested. In surface contours figuring in color areas, so it can be seen from this graph the points of interaction of two variables is clear, where most interactions are optimal in the red region of the oldest.

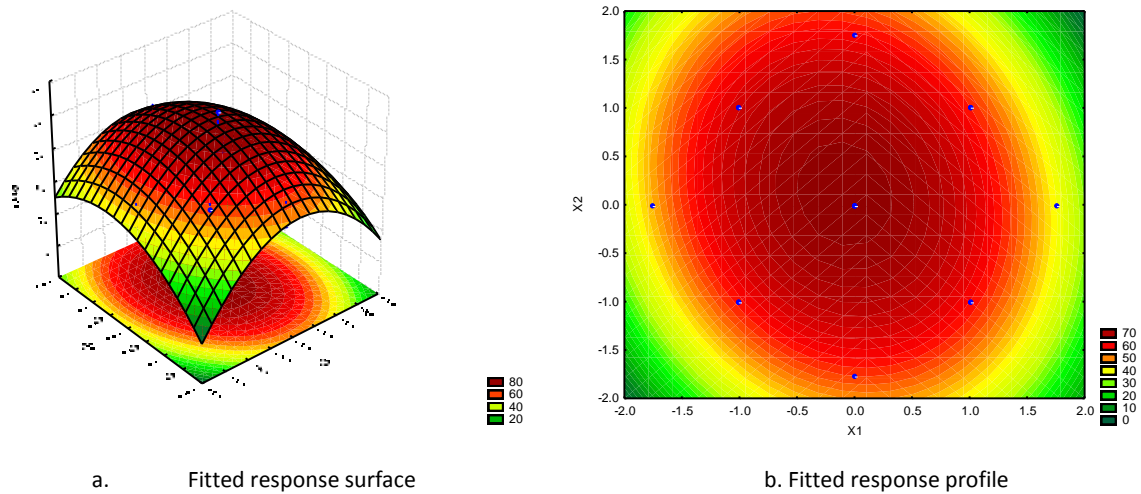


Figure 6. Yield versus X_1 and X_2

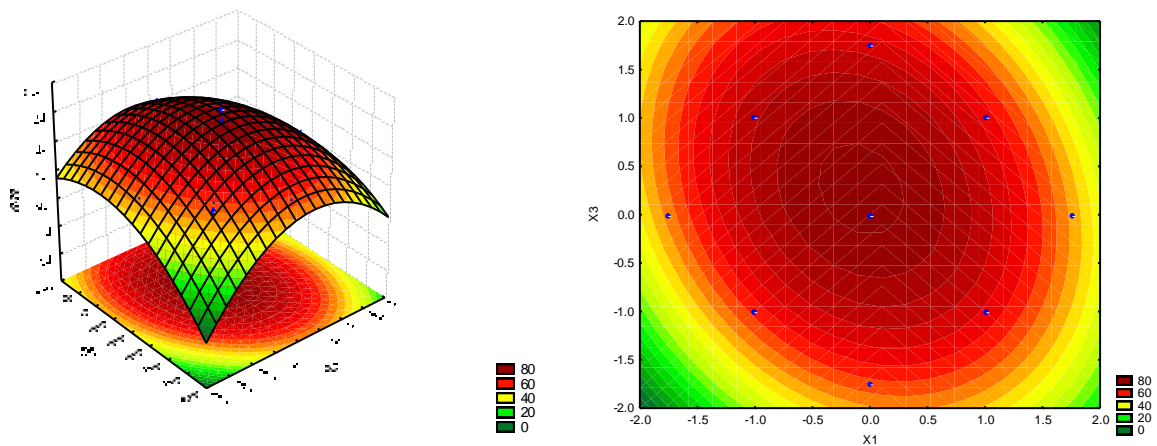


Figure 7. Yield versus X_1 and X_2

The results of the response fitted surface graphs are parabolic and contour plot is shaped oval. This suggests that the type of optimization process is already maximized. The critical value for each variable is shown in the following table:

Table 2. Critical value each variables

| No. | Variable | Observed Minimum | Critical Values | Observed Maximum |
|-----|--------------------------------|------------------|-----------------|------------------|
| 1. | Ratio of Nyamplung to Methanol | -1.76000 | -0.0884 | 1.760000 |
| 2. | Catalyst concentration | -1.76000 | 0.1124 | 1.760000 |
| 3. | Temperature | -1.76000 | 0.2408 | 1.760000 |

In table 2, the critical value of dimensionless numbers for each variable. Critical dimensionless value obtained for X_1 (weight ratio nyamplung to methanol) is -0.0884, X_2 (catalyst concentration) 0.1124 and X_3 (temperature operation) is 0.2408. These values were input in 7 equations and obtained conversion of biodiesel is 64.429%. These values were intake in 2-4 equation ad obtained optimum condition are 1:5.9116, 2.556% and 46.2oC for ratio weight of nyamplung seed to methanol, catalyst concentration and temperature respectively.

3.4. Biodiesel Production with Assisted Ultrasonic versus Conventional Process

Table 4. Characteristics biodiesel with ultrasonic and conventional

| No. | Characteristics | Conventional | Ultrasonic wave |
|-----|---------------------------|--------------|-----------------|
| 1. | Density | 0.932 | 0.888 |
| 2. | Kinematic viscosity (Cst) | 4.88 | 4.5 |
| 3. | Conversion | 54.09 | 86.96 |

Table 4 shows the processed biodiesel that processed with ultrasonic waves have better characteristics than conventionally processed biodiesel. With reaction time and the same reaction process variables biodiesel conversion that resulting from ultrasonic wave is greater than the conventional processes. The rate of transesterification reaction using ultrasonic waves higher than the use of a mechanical stirrer [14,15]. This is because the effects of cavitations and oscillations of the use of ultrasonic sound waves in the transesterification reaction. Oil molecules or FFA that reacted with methanol has a very fast movement and bounced hard. The movement of oil and methanol molecules is random, so that the stirring becomes more homogeneous. The micro bubbles of storing large energy in the form of surface tension, where the magnitude of surface tension is inversely proportional to the radius of the bubble. Small diameter of the bubbles that will save energy in the form of a large surface tension [12,13].

3.5. Biodiesel Characteristics

Table 5. Characteristics experiment biodiesel and SNI standard [16]

| No. | Characteristics | Biodiesel from experiment | SNI standard |
|-----|------------------------|---------------------------|--------------|
| 1. | Density (g/ML) | 0,888 | 0,85-0,89 |
| 2. | Viscosity (cSt) | 4,5 | 2,3-6,0 |
| 3. | Acid number (mg KOH/g) | 0,44 | Max 0,8 |

Table 5 shows that the characteristics of the biodiesel meets the characteristics of the experimental results already established by the SNI. Calophyllum inophyllum-biodiesel viscosity slightly higher but still meet the standards set by SNI. Oil viscosity is expressed by the number of time (seconds) used by a certain volume of oil to flow through small diameter holes with a particular, the lower the second number means that the lower the viscosity. Viscosity is too high can the burden of the pump and cause poor fogging [8].

The density of biodiesel from nyamplung oils meet the requirements of SNI biodiesel is 0.888 g / ml. Although the density of biodiesel is slightly higher, but still included in the SNI standard biodiesel making biodiesel produced viable. Differences related to the density of biodiesel fatty acid composition and degree of purity of the biodiesel [7].

4. Conclusion

Biodiesel derived from the seeds of raw materials Nyamplung (*Calophyllum inophyllum*) is in conformity with the SNI. This is indicated by the physical and chemical properties of biodiesel. Optimum conversion of biodiesel production from nyamplung seed with assisted ultrasonic was 64.429%. The optimum condition are 1:5.9116, 2.556% and 46.2oC for ratio weight of nyamplung seed to methanol, catalyst concentration and temperature respectively. Model equations for the biodiesel formation reaction is:

$$Y = 69.609 - 0.7586X_1 + 1.9051 X_2 + 3.2476 X_3 - 2.08 X_1X_2 - 3.665 X_1X_3 - 2.865 X_2X_3 - 10.6085 X_1^2 - 6.224 X_2^2 - 6.747 X_3^2$$

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