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Characterization and Physicochemical Properties of Biofuel from Mixed Crude Jatropha Oil and Clove Oil

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ABSTRACT

The modern development reveals that the world is facing energy crisis as a result of the dwindling sources of fossil fuels. Biofulel is renewable clean bioenergy which can be made from vegetable oils, micro-algal oil and animal fats. The feasibility of Biofuel production from clove oil and jatropha oil feedstock was studied in the present investigation. In addition, the studies were evaluating the physicochemical properties and thermal stability of fuel blend between clove oil and jatropha oil (JO). In this study, JO-clove oil blend with initial JOC5, JOC10, and JOC15 in percentage 5, 10 and 15% on the basis volume, have been used. Density, viscosity, heating value, flash point, and thermogravimetric analysis were tested by international standard method. The result indicates that the presence of clove oil affect in density and heating value slightly increase while viscosity and flash point decrease. Thermal stability in JO was modified due to presence of clove oil that marked by percentage of mass loss increase and onset temperature decrease. The improvement physicochemical properties and thermal stability caused by molecular interaction between eugenol and triglyceride that makes molecule active in motion. The volatility of clove oil has big contribution in thermal stability of JO beside the percentage of unsaturated fatty acid.

Keywords: Jatropha Oil, Clove Oil, Biofuel, Physicochemical Properties, Thermal Stability.

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INTRODUCTION

The demand for substitute fuels has increased in the several years ago. Many alternatives have come into being in the recent years and several more are on their way to get established as a sustainable fuel substitute. Jatropha oil (JO) is nonedible oil that made up of triglycerides, molecules which consist of three fatty acids joined by a glycerol group. That molecules due to JO has high viscosity and low evaporation rate. The demerit leads to poor fuel atomization, ring sticking, injector cocking, injector deposits, injector pump failure and lubricating oil dilution by crank-case polymerization [1]. Preheating, oil transesterification become biodiesel, and blending are some of the methods used to reduce it. Preheated JO was tested in diesel engine at 1500 rpm and show result that it can be directly used as straight jatropha oil without any modification in the engine [2]. Biodiesel is a transesterification product, which refer to the fatty acid alkyl esters (FAAEs), are derived from lipid substances originated from vegetable oil, animal fats, waste greases, recycled cooking oils etc.[3]. Biodiesel fuel can effectively reduce engine-out emissions of particulate matter, carbon monoxide (CO), and unburned hydrocarbons in modern four-stroke compression–ignition engines [4]. Unfortunately, conversion vegetable oil to biodiesel require a high energy amount and extra cost [5]. The simplest method to reduce viscosity is blending vegetable oil with another fuel that have lower viscosity. Blending 87% crude coconut oil, 10% ethanol, and 3% 1-butanol causes viscosity reduced from 40.09 cst to 19.27 cst [6]. Rapeseed oil blend with propanol and butanol can reduce viscosity until 20% [7]. Neem methyl ester blending with 5% ethanol can decrease viscosity and flash point to 6% and 7%, respectively [8].

Alcohol like Ethanol is a volatile liquid, less toxic, can easily produce from renewable source [9]. Ethanol and methanol (simple alcohol) are completely miscible with water but show very poor miscibility with gasoline containing traces of water. So, blending gasoline with ethanol or methanol in the presence of water may lead to a phase separation problem [10]. JO has limit solubility when blended with methanol and ethanol. Their mixture creates phase separation due to JO is non-polar and simple alcohol is polar. Stability test of ethanol-castor oil blends concluded that only 14 out of 36 samples were considered miscible after 24 h settling [11]. Water contain in alcohol and vegetable oil blend is another obstacle in fuel blend. Emulsifier for alcohol used to enhance phase stability, improve ignition delay and cetane number [7]. Ethanol can be mixed in diesel fuel because of aromatic compounds in diesel fuel that serves as a bridge between ethanol and diesel alkanes [12] [13]. Kadarohman (2012) stated that terpene, which is major contain of aromatic compound, acted as mediator agent between bio-additive and base fuel resulting a perfect solution. Clove oil is a volatile aromatic compounds with the largest content is eugenol and terpene [14]. Their viscosity similar with diesel but its density higher because of bulky and rigid structure [15] [16]. Clove oil could be soluble when blended with JO due to the aromatic compound. This blend can reduce viscosity and flash point to overcome the disadvantage of use JO. The aim of this study on Characterization Biofuel from Mixed Crude Jatropha Oil and Clove Oil. In addition, this study will evaluate the Physicochemical Properties and thermal stability of the new biofuel mixture.

Material and Method

Material

This experiment used clove oil from local market and JO was obtained by mechanical extraction using expeller in Sweetener and Fiber Crops Research Institute, Malang, East Java, Indonesia (Balittas). The ranges of fatty acid composition of each vegetable oil reported in literature are presented in table 1. Clove oil tested by GC-MS to explore about the composition and shown in table 2.

Table 1. Fatty acid composition of JO [17]

Fatty Acid Composition	Number of C	JO
Myristic Acid	(C14:0)	0-0.1
Palmitic Acid	(C16:0)	14.1-15.3
Stearic Acid	(C18:0)	3.7-9.8
Oleic Acid	(C18:1)	34.3-45.8
Linoleic Acid	(C18:2)	29-44.2
Linolenic Acid	(C18:3)	0-0.03
% Unsaturation		63.3-90

Table 2. Composition of clove oil [14,18]

Compound	%
Eugenol	63.74
Humulene ($C_{15}H_{24}$)	26.32
Caryophyllene	6.31
a-Humulene	3.53

The molecular structure of JO is triglycerides that contain glycerol and three functional groups of esters called fatty acid which the mainly are oleic, linoleic, palmitic, and stearic acid that shown in table 1. The fuel thermal instability is directly proportional to the number of double bonds [19]. Generally, the mono and polyunsaturated fatty acids (C18:1 oleic acid; C18:2 linoleic acid; C18:3 linolenic acid) are more susceptible to instable thermal fuel than saturated fatty acid.

Based on table 2, eugenol is a highest compound in clove oil that classified as a phenol and consist of aromatics structures and hydroxyl groups (-OH). Aromatics structure which is ring benzene also known to increase the adiabatic flame temperature [20], and hydroxyl groups which is a relatively electronegative atom, perform as a hydrogen bond donor [21]. Humulene, Caryophyllene, and a-Humulene are classified in sesquiterpenes. Sesquiterpenes can be monocyclic, bicyclic or tricyclic, have strong odor and act as antioxidant.

Density

Density is the relationship between the mass and volume of a liquid and can be expressed in units of grams per liter [22]. The density gives an indication of the ignition delay and specific energy of the fuel in a diesel engine. The densities of JO and its blend were measured in accordance with ISO 4787 standard and determined by means equation (1). Where ρ and m represent density and mass, respectively. Density measurement were conduct three times and average it to minimize error.

$$\rho_{JO\ or\ blend} = \frac{m_{total} - m_{pycnometer}}{m_{water}} \cdot \rho_{water}$$

Viscosity

Viscosity is an indication of fluid on the stickiness and ability to flow. It is of remarkable influence in the mechanism of atomization of fuel spray in the operation of an injection system [23]. The derivation of the relationship between shear stress and shear rate also called viscosity [24]. Kinematic viscosity measured by viscometer apparatus from Leybold Didactic with ASTM D445 standard method and various temperatures: 27, 40, 50, 60 70, 80, 90 and 100°C.

Heating Value

Amount of chemical energy that released by the combustion of a unit mass of fuel called Heating value [25]. The heating value of the CJO and its blend were measured in a bomb calorimeter apparatus from Paar Instrument according to ASTM D240 standart method. A comparison of the energy contents of the JO sample and its blends can predict how an engine would perform on alternative fuels.

Flash Point

Flash point is a flammability property of fuel. It defined as the lowest temperature at which the mixture of vapor and air above the surface of the liquid can be ignited [26]. It was evaluated using the closed cup flashpoint tester from Leybold Didactic according to ASTM D93. The measurements were carried out three times after which the results were averaged.

Thermogravimetric analysis

Thermogravimetric analyzer (TGA) measures mass changes in materials with regard to temperature such as in boiling or evaporation [7]. Thermogravimetric data were used in characterizing the materials as well as in investigating the thermodynamics and kinetics of the reactions and transitions that resulting from the application of the oil samples [27]. Thermo analytical methods include a group of techniques in which the thermal behavior or thermal properties of a material are determined as a function of temperature. In this study, the equipment continuously monitors the loss of sample mass while the sample is heated in dynamic conditions. TGA was used to determine the evaporation temperature range of the JO and its blends. Samples weighed about 24-26 mg and put into the alumina crucible. Crucible sample is placed in a test chamber with heating program up to 600°C in argon atmosphere and increase every 10°C/min. Temperature is held (isotherm) for 5 minutes at the temperature 600°C and reheating to 900°C in an oxygen atmosphere increase in every 10°C/min.

Result and Discussion

Density

Density of JO in this study is 0.921 gr/ml, there is not much different from previous study where the range density of JO in 0.901-0.940 gr/ml [28,29]. Density directly influences the fuel injection process because the amount of fuel injected into the engine is estimated by its volume [30]. Besides that, density is a parameter that have correlation with viscosity and heating value. JO have high density because of their molecular weight around 600-900 g/mol [31]. Bulky structure in JO caused by percentage of double bond fatty acid in range 63-90% (see table 1).

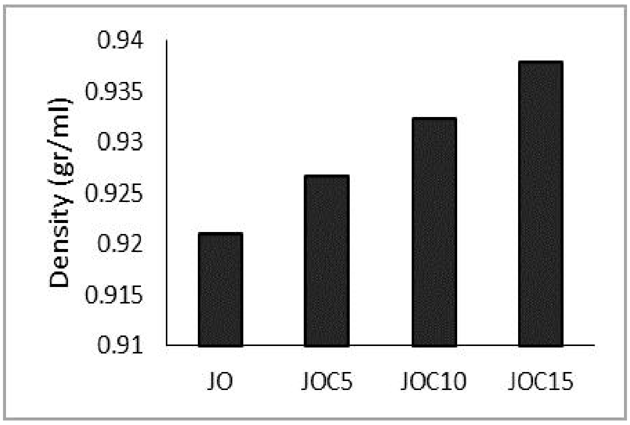

Fig 1. Density of JO and its blend

Based on fig.1, Density slightly increase in more percentage of clove oil. In percentage 5% clove oil, density increase 0.5% become 0.92665 gr/ml. Density increase consistently in 15% clove oil, when its up 1.8% become 0.93795 gr/ml. Clove oil was compound by aromatic structure that usually consist of one aromatic ring with one or several side alkyl chain. The aromatic ring in clove oil was arranged in a dense and rigid, causing the density higher. This result from the fact that density of JO-Clove Oil blend depends on carbon content, double bond, and aromatic structure. A fuel with high density implies that the energy content per unit volume increase, energy delivered per cycle increase, and higher air-fuel ratio inside the combustion chamber. Consequently, it results unburnt fuel and particle emission in the exhaust gas.

Viscosity

As seen on fig.2, the viscosity has been decreased with increase of temperature. JO decrease from 52,419 cst to 9,0189 cst, JOC5 from 48,71 cst to 8,798 cst, JOC10 from 45,074 cst to 8,441 cst, JOC15 from 41,442 cst to 7,758 cst. While temperatur is increase, the molecular interaction has been decreased and molecular bond would be stretchable. Thermal energy transfered to the molecule and because kinetic energy gained then makes oscillation of molecule greater than before. In increasing temperature environment, both molecular vibration and rotation can become considerable [32]. Viscocity decreased due to energy input in molecule lead to active vibration and rotational movement of molecule.

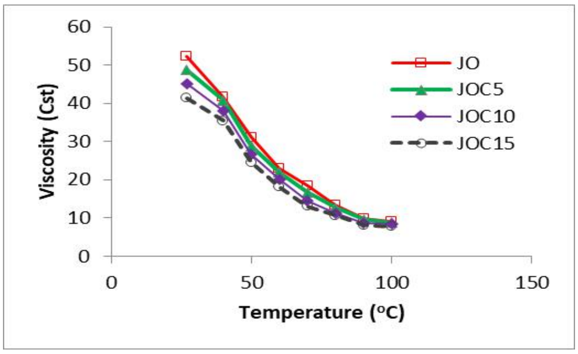

Fig. 2. Viscosity of different percentage of JO-clove oil blend

In JO, the viscosity increases with chain lengths of fatty acids of triglyceride and decreases with increases in the unsaturation. So, viscosity is a function of molecules dimension and orientation [33]. Based on clove oil percentage, viscosity of fuel blend decreases with more percentage of clove oil. The largest composition of clove oil is eugenol that has hydroxyl group (-OH) and ring aromatic. This group create van der waals bond with triglyceride in JO (Fig.3). Although van der waals bond is weak, it makes molecule fuel blend more active and oscillation greater than JO. Triglyceride in JO is nonpolar, where the movement of molecule caused by london force that weaker than van der waals.

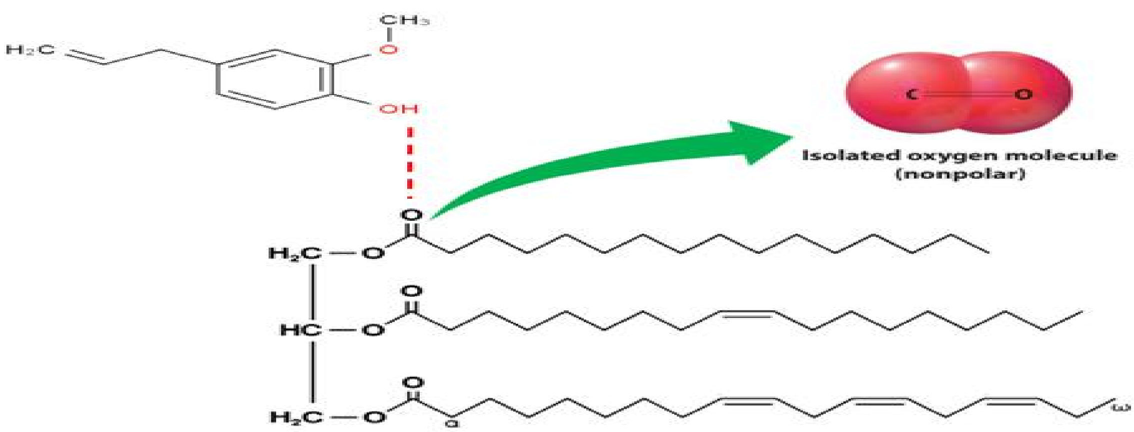

Fig 3. Ilustration of molecular interaction between hidroxyl group and triglyceride

Viscosity of preheating JO and its blend meet the requirement of diesel fuel standard ASTM D396 for grade no.4-D which set limit value of 5.8-26.4 cst. It needs to be ensured that fuel pump robust enough to circulate the oil.

Higher viscosity lead to lower efficiency of fuel pump, shorter life time of pump and increase risk of fuel leakage [34]. Another problem that created by higher viscosity are piston ring sticking, gum formation, and fuel atomization [19].

Heating Value

Based on fig.4, heating value of fuel blend is increase with more percentage of clove oil. JO have heating value 37.46 MJ/kg and slightly increase in 37.98 MJ/kg when blend with clove oil 15%. Rise of heating value was related to rise in density, where density increase slightly as more percentage of clove oil. Heating value increase due to presence of oxygen content, aromatic ring and hydroxyl group in eugenol structure. Oxygen content of the blended fuel improve their burning characteristics, hydroxyl group makes blended fuel burns at lower temperature [35] and aromatic ring produce high flame temperature [20]. Fuel having higher heating value gives higher power output from smaller engine and can provide longer transportation distance with smaller storage fuel tank [25]. Because of their heating value lower than diesel (49.95 MJ/kg), blends will need more fuel consumption and produce lower energy power. Lower heating value lead to reduction temperature in exhaust gas and brake horse power decreased at higher compression [36].

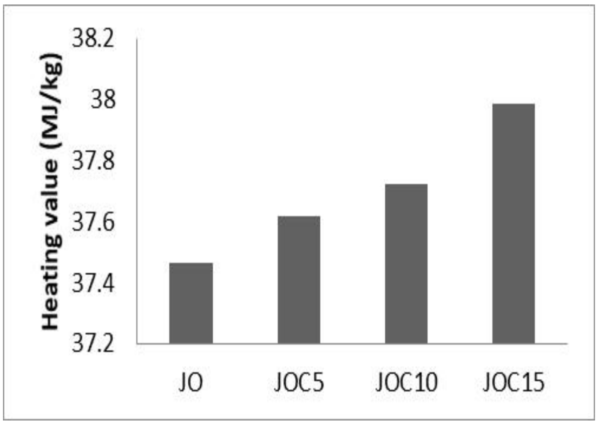

Fig 4. Heating value of JO and its blend

Flash Point

Flash point temperature is the minimum temperature at which volatile fuel ignites momentarily when contact in spark of flame [30]. Flash point decreased with increase clove oil according to fig.5. Flash point of JO is 208 °C decrease until 17% at 172 °C when blend with clove oil 15%. Presence of clove oil cause vapor content of fuel blend increase. Eugenol and terpene possible to become vapor first and promote triglyceride in second. Resonance in aromatic ring structure and presence of terpene lead to clove oil as a volatile oil and produce fragrance. Adding a more volatile liquid can depress the flash point [37].

Flash point also influenced by molecular interaction, molecule size, and orientation of group dipole [38]. Molecule of JO and clove oil are bulky structure lead to high density meanwhile presence clove oil in JO blend lead to molecular interaction more active because of clove oil polar and JO nonpolar. The volatility of clove oil makes eugenol interaction with triglyceride in van der waals become vapor in evaporation phase. Spontaneous ignition in minimum temperature occur when fuel blend in vapor phase mixture with air and contact with spark in surface. More molecular interaction makes flash point of fuel blend decrease. Generally, flash point of JO and its blend are higher than diesel (60-80°C) ensures greater safety in the handling and storage of the fuel.

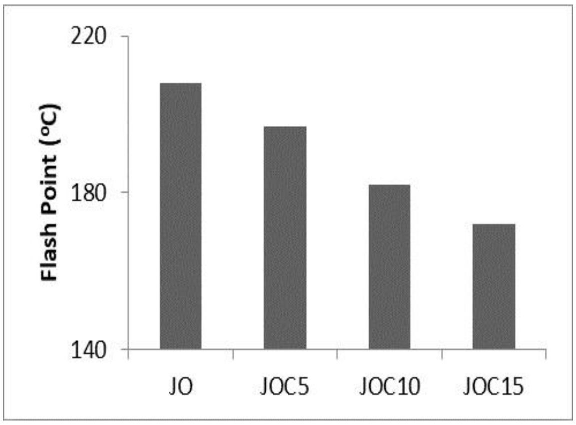

Fig 5. Flash Point Close Cup of fuel blend

Thermo gravimetric analysis

Comparison of TG-DTG curve of JO and its blend are shown in fig. 6. Curve of JO and its blend plot show that the sample take place in more than single continues step due to fatty ester. Chain length, branch of chain, and degree of unsaturation are the factor that influence the thermo-oxidative properties of fatty ester [39]. Based on table 1, JO contain of unsaturated fatty acid more than 50% which is less stable than saturated fatty acid. In range temperature 200-400 °C (fig 6 a), 26 % mass loss which include moisture loss and decomposition unsaturated fatty acid mainly oleic acid and linoleic acid. Rapid mass loss occurs in second step at range temperature 400 -500°C due to decomposition of saturated fatty acid. Start from range temperature between 500 C – 600°C, the mass percentage become stagnant and curve flattering because non-volatile fraction in remaining sample consist of ash and fixed carbon. In this step also shown no further conversion was occurring.

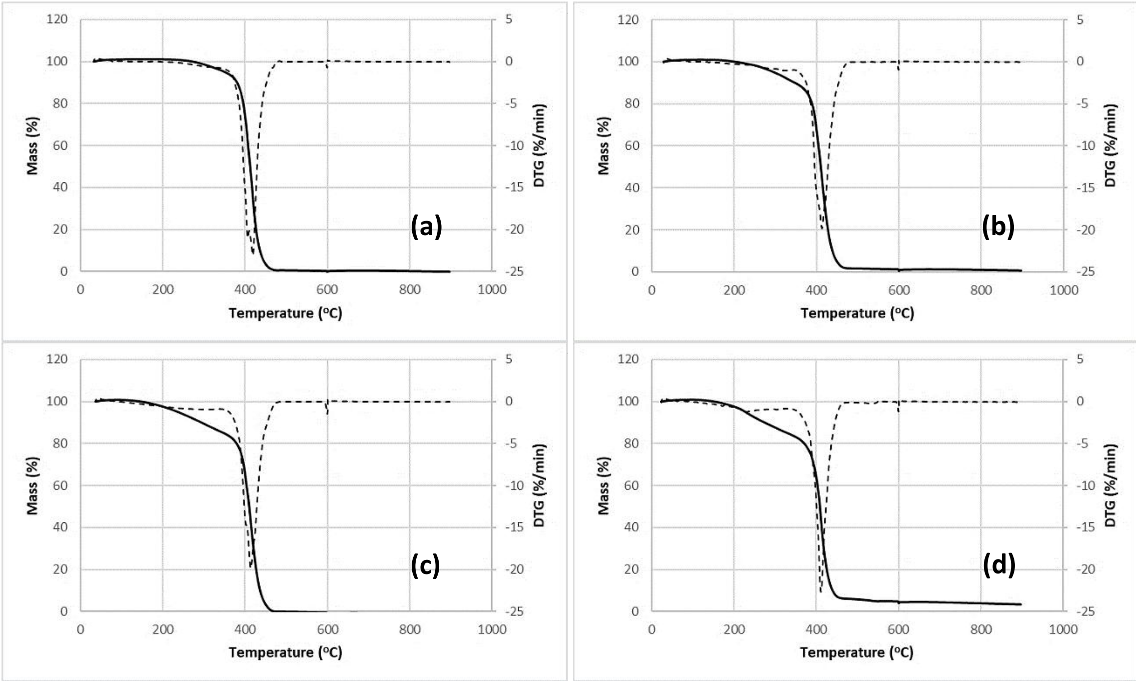

Fig 6. TG-DTG of: (a) JO, (b) JOC5, (c) JOC10 and (d) JOC15

Effect of clove oil in JO blend describe in thermogram that shown in fig 6 (b), (c), (d). Unsaturated fatty acid and the volatile components in clove oil, which are eugenol and terpene evaporate from the blend sample at an early stage of the heating process. Mass loss in 32, 35, and 36% for JOC5, JOC10, JOC15, respectively. After the evaporation of clove oil and unsaturated fatty acid in range between 200-400°C, the TGA and DTG curves exhibited the same trends as those observed in the case of the pure JO sample. Increasing clove oil percentage lead to decreasing curve in sloping trend between 200-400°C, indicate increase the percentage of medium volatile matter that found in JO-clove oil blend.

Thermal stability related to the onset temperature which was determined in thermogravimetric curve as the intersection point of the tangent to two branches [40]. Temperature of decomposition begin in sample can be defined as onset temperature [39]. Onset temperature of JO, JOC5, JOC10, and JOC15 are 408.15°C, 400.22°C, 389.85°C, 380.12°C, respectively. Thermal stability of JO is higher than its blend due to unsaturated fatty acid mainly oleic acid and linoleic acid. Presence of clove oil lead to JO unstable and need lower heat to decomposed. JOC15 get lower onset temperature indicate more volatile matter in this blend. The more of the volatile degradation product, the lower of the onset temperature [41].

Conclusion

A study on Characterization of Biofuel from Mixed Crude Jatropha Oil and Clove Oil has been conducted. The Physicochemical Properties and thermal stability of the new biofuel mixture has been studied also in the present investigation. Based on the archived results, the conclusions can be summarized as follows:

a. Density of fuel blend slightly increase due to bulky structure in triglyceride, eugenol and terpene.
b. The viscosity of fuel blend linearly decrease as the proportion of clove oil in the blend increase. Beside that, viscosity of blend decrease when the temperature increase. Van der waals interaction lead to movement of fuel blends molecules more active.
c. Heating value of the blend increase as the clove oil percentage increase because of presence of oxygen content, aromatic ring and hidroxyl group in eugenol.
d. Flash point of fuel blend decrease as the clove oil concentration increase because clove oil is volatile oil that can depress flash point of fuel blend.
e. Presence of clove oil decrease the onset temperature which is lead to JO unstable and decompose in lower temperature.

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