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Research Paper

Droplet Combustion and Thermogravimetric Analysis of Pure Coconut Oil, Clove Oil, and Their Mixture

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Abstract
The droplet combustion and thermal behavior of pure coconut oil (PCO), clove oil, and their mixture were experimentally investigated. The mixture fuels were PCO and clove oil at the percentage of 10% based on volume (PCO-CO10). The experimental method uses droplet combustion and thermogravimetric analysis. The fuel droplet was suspended in the junction of k-type thermocouple and ignited by a coil heater. The ignition and combustion processes of droplets were recorded using a digital single-lens reflex camera at 25 fps. Thermogravimetric analysis with alumina crucible was prepared to investigate the thermal behavior of fuel. The result showed that the sequence of ovoid flame for PCO and PCO-CO10 take place until 0.4 second and 0.44 second, respectively. Complete combustion was explained in that sequence. The ovoid flame was formed when eugenol, terpene, and lauric acid were evaporated first in both PCO and PCO-CO10. Minimum ovoid flame takes place in clove oil due to soot tendency in the burning process that marked flame as the open tip. PCO-CO10 has the highest peak temperature due to the presence both of double carbon chains in fatty acid and aromatic ring structures, which were easy to decompose in high energy input. Clove oil was the lowest onset temperature, which indicates more volatile matter in this fuel and PCO has the highest thermal stability due to the fatty acid component in their fuel.

Keywords
Coconut Oil, Clove Oil, Droplet Combustion, Fatty Acid, Volatile Oil

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1. INTRODUCTION

The important energy resource in the world today, which is limited and non-renewable, is a petroleum product derived from crude oil. In order to meet the growing global energy requirement and the long-term availability of energy resources, many researchers study renewable energy to address this problem. Vegetable oil is kind a source of renewable energy, which is non-toxic and does not contribute to global warming. Vegetable oils can be harvested from non-edible and edible oil. Pure coconut oil (PCO) is an edible oil extracted from dried copra processed from fresh mature nuts by an effective expeller extraction procedure and an average yield of 68-70% was obtained at each trial (Chinnamma et al., 2015). The industrial-scale of PCO production for food, therapeutic, and nutrition indicates the availability of raw material. However, the higher viscosity and low volatility features of the PCO have limited its usage in the engine for the long term.

Some methods to reduce viscosity in PCO such as preheating the oil, transesterification of the oil to become biodiesel, and blending the oil with liquid in high volatility. Preheated PCO until 100°C was tested in the four strokes diesel engine D240 and reduced viscosity from 28.1 to 5.4 mm^2/s. The result suggested installing a heating system for using PCO (Hoang and Van Le, 2017). In the two-step transesterification process, coconut oil's viscosity was reduced from 28.05 to 2.937 mm^2/s (Nakpong and Wootthikanokkhan, 2010). Another test of transesterification based on SNI 04-7182-2006 exhibits that the viscosity of coconut oil was reduced from 21.61 to 2.62 mm^2/s (Minaria and Mohadi, 2016). However, biodiesel production needs a high energy amount and high processing cost (Sanjeeva et al., 2014). The blending method could be the easiest way to reduce viscosity. Another researcher reduced the viscosity of PCO with the composition of a blend of 87% PCO-10% ethanol-3% butanol in volume ratio from 40.09 cst to 19.27 cst (Singh et al., 2010). The test engine with PCO and PCO–diesel fuel blends in every engine load condition was shown to be successful even without modification. Increasing the percentage of coconut oil in the fuel blend resulted in lower smoke and NO_x emissions (Machacon et al., 2001). The multi-cylinder diesel engine was operated with diesel grade D2 and blended with 10%, 30%, and 50% volume of coconut oil (How et al., 2012). The emission was reduced as a result of the use of coconut oil blend, except in CO_2 emission due to fluctuations result.

Single droplet combustion is an attractive analysis, low utilization, and cost-effective method for combustion characteristics based on fuel properties (Gamayel et al., 2020a). Pure fuel or single component droplets exhibit steady combustion, while multicomponent reported staged combustion behavior (Hoxie et al., 2014). Many researchers have studied multi-component droplet combustion involving hydrocarbon fuels. Droplet combustion in the methanol/alkane droplet shows that a higher methanol volume fraction leads to the lower ignition delay time (Wang et al., 2005). The average burning rate is not changed by the variation of temperature in the crude jatropha oil droplet (Wardana, 2010). The high differential volatility in the multi-component droplet led to micro-explosion phenomena. The researcher analyzed the burning process with micro-explosion phenomena butanol blend with soybean oil in initial concentrations ranging from 25–75% (Hoxie et al., 2014). Their result showed that Bu40 gets more micro-explosion than all mixture studied. Another researcher studied the suspended droplet of isopropyl nitrate (IPN) and its blend (n-heptane, desensitizer dibutyl sebacate) (Ambekar et al., 2014). They found that simultaneous gasification of IPN-n-heptane leads to absent of micro-explosion.

Clove oil is an essential oil that eugenol is the largest compound with two oxygen atoms and ring benzene. The performance and emission of a 4-cylinder direct injection diesel engine operated using the clove stem oil (CSO)-diesel fuel blend were investigated (Mbarawa, 2010). The CSO-diesel blend has lower HC emissions than pure diesel. The mixture of clove oil fuel blend and air promotes rapid combustion and lower ignition delay (Kadarohman et al., 2012). The oxidation stability of cotton seed biodiesel is increased by the addition of clove oil as an antioxidant (Jeyakumar and Narayanasamy, 2019). No previous study has investigated PCO-clove oil blend in droplet combustion and analysis in thermal. This experiment address to study of combustion behavior and thermal analysis of PCO, clove oil, and their mixture.

2. EXPERIMENTAL SECTION

2.1 Materials

PCO is an edible vegetable oil extracted using an expeller machine from the fruit of a coconut tree. Coconut is a perennial crop that means oil production continuously and uninterrupted throughout the year. The plant is grown up to 30 m tall in tropical countries in Southeast Asia. The grain yield of this plan is 100-150 fruits/tree/year, with 65-67% oil content (Hoang and Van Le, 2017). The molecular structure of PCO is a triglyceride, which is common with other vegetable oil that contains a 3-ester functional group. Glycerol and fatty acids are produced by the hydrolysis reaction of triglyceride (Wardana, 2010). Literature reviews in gas chromatography are performed to determine the fatty acid composition of PCO shown in Table 1.

Table 1. Fatty Acid Composition of PCO (Nakpong and Wootthikanokkhan, 2010)

Fatty Acid Composition	Number of C	PCO
Caprylic Acid	(C8:0)	3.35
Capric Acid	(C10:0)	3.21
Lauric Acid	(C12:0)	32.72
Myristic Acid	(C14:0)	18.38
Palmitic Acid	(C16:0)	13.13
Stearic Acid	(C18:0)	3.6
Oleic Acid	(C18:1)	12.88
Linoleic Acid	(C18:2)	4.35
Linolenic Acid	(C18:3)	n.d.
% Unsaturation		17.23

Table 2 present the composition of clove oil consisting of eugenol, *cis*-caryophyllene, caryophyllene, a-Humulene, and Figure 1 shows the molecular structure of clove oil composition. Eugenol is a phenolic compound composed of two-atom oxygen and benzene ring. Phenol is a primary antioxidant due to the conversion capability of high-energy lipid radicals into thermodynamically more stable compounds (Jeyakumar and Narayanasamy, 2019). The hydroxyl group in eugenol burns at low temperature and easier oxidation in combustion (Botero et al., 2012; Mwangi et al., 2015), while aromatic structures are produced at high flame temperature (Han, 2013).

Table 2. Composition of Clove Oil

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

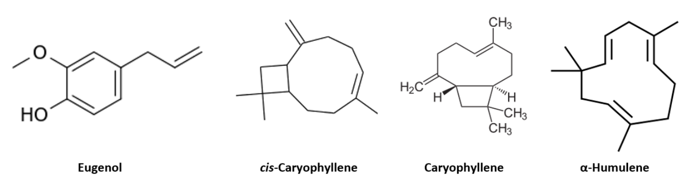

Figure 1. Molecule Structure of Clove Oil

White crystalline solid is the physical appearance of PCO at a temperature under 20°C at room temperature and PCO is a clear liquid when blended with clove oil. Regarding the PCO-clove oil blend, the PCO is a complex mixture of different chemical compounds containing ester groups from highly non-polar to moderately polar aromatic hydrocarbon. It is due to the hydrogen atom of the hydroxyl group at eugenol being a hydrogen donor, while the oxygen atom of the ester group acting as a hydrogen acceptor. The presence of polar aromatic hydrocarbon in clove oil creates the fuel blend stable and dissolves the PCO.

In this study, PCO was observed as pure fuel and mixture with clove oil at a percentage of 10% in basis volume. Both fuels with initial PCO and PCO-CO10 were tested and compared with clove oil in droplet combustion apparatus and thermogravimetric analysis. Properties in the international standard measurement of PCO and clove oil are given in Table 3.

Table 3. Properties of PCO and Clove Oil

Properties	ASTM	PCO	PCO-CO10	Clove Oil***
Density	D1298	0.915	0.927	1034.1
Viscosity	D445	30.12	25.7	4.1
Heating Value	D240	37.1*	35.8	33.6
Flash Point	D93	249**	218	114

*(Hoang and Van Le, 2017); **(Wahyudi et al., 2018); ***(Mbarawa, 2010)

2.2 Methods

2.2.1 Droplet Combustion

The experimental set-up is similar to a previous study Gamayel et al. (2020b), where the junction of a k-type thermocouple has a diameter of 0.2 mm as a place of the tested droplet. A coil heater made from Ni-Cr wire has a diameter of 0.7 mm and is used to ignite the droplet with a DC power supply of 18V. DLAB Scientific Inc-micropipette was used to create the droplet in the volume range 0.5~1 microliter. The plugged thermocouple in the DATAQ Acquisition system was connected to a computer used to measure the temperature of droplet combustion. The flame evolution was recorded by a digital single-lens reflex camera at 25 fps (Canon EOS 4000D) with set up Exposure Value (EV) in 1600 to minimize disruptive light. Figure 2 shows the experimental setup of this study.

2.2.2 Thermogravimetric Analysis

Mass change with increasing temperature in the boiling phase or evaporation is measured by TGA (thermogravimetric analyzer) (Laza and Bereczky, 2011). TGA investigated fuel samples in thermodynamics and kinetics of reaction & transitions (Jain and Sharma, 2011). Labs TGA-DTG with alumina crucible was the equipment to investigate the thermal properties or thermal behavior of PCO-clove oil. The equipment continuously monitors the loss of sample mass while the sample is heated in dynamic conditions. Therefore, an evaporation temperature range of PCO-clove oil was determined. The heating up to 600°C in argon atmosphere was programmed in the chamber and increased every 10°C/min. The temperature

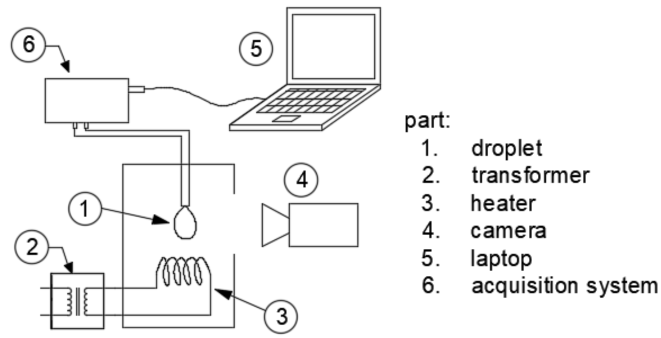

Figure 2. Experimental Set-Up

was held at 600°C for 5 minutes and reheated every 10°C/min in an oxygen atmosphere until it reached 900°C.

3. RESULTS AND DISCUSSION

3.1 Droplet Combustion

Flame evolution was recorded by video recorder with a capability of 25 frames per second. It means that converting a video become a picture in every frame per sequence is 0.04 seconds. The flame evolution of PCO and its blend is shown in Figure 3.

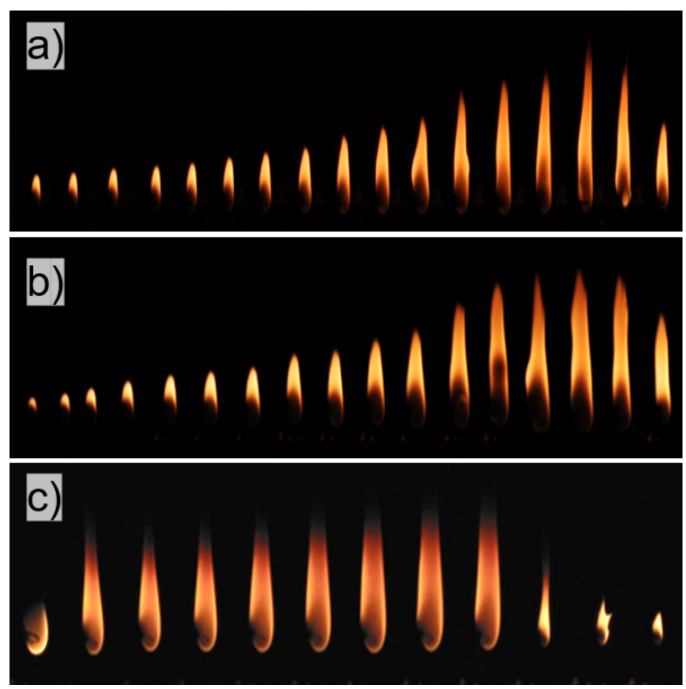

Figure 3. The sequence of Flame Evolution (a) PCO, (b) PCO-CO10, (c) Clove Oil

The ovoid flame defines as a flame with a shape like an egg and has an around-in-tip flame. It explains complete combustion in that sequence. An ovoid flame tends to have stable combustion without micro-explosion phenomena. Figures 3a and 3b show the sequence of ovoid flame for PCO and PCO-CO10 until 0.4 second and 0.44 second, respectively. The presence of lauric acid of more than 30% in PCO and its blend led to stable combustion. An ovoid flame is formed when the lauric acid vaporizes first, then followed by unsaturated fatty acid and glycerol to form a spike flame. The straight carbon chain without a double bond leads lauric acid easier to evaporate and create combustion process completed. In PCO-C10 (Figure 3b), eugenol and terpene evaporate first then, followed by lauric acid, unsaturated fatty acid, and glycerol. Figure 3c shows the minimum ovoid flame in clove oil due to aromatic structure in the combustion process, lead an orange hue in top flame and soot tendency. The soot led to the flame shape of clove oil without tip round or open tip. Rao et al. (2018) state that the orange hue at the top edge of the flame is due to the emission of soot. Micro-explosion is marked as bulge geometry of flame and changes in a short time to become like a spike (Wardana, 2010). In Figure 3, micro-explosion take place in 0.44 second both for PCO and clove oil, thus 0.48 second for PCO-CO10. Except for the clove oil, bulge geometry followed by spike flame and then flame off. The small amount of micro-explosion in clove oil due to the difference in boiling point between eugenol and terpene is smaller than fatty acid-glycerol. Also, the minimum micro-explosion phenomena occur in PCO and its blend due to the more percentage of volatile oil and saturated fatty acid than an unsaturated fatty acid. The detail of temperature and time in the droplet combustion process are shown in Tables 3 and 4.

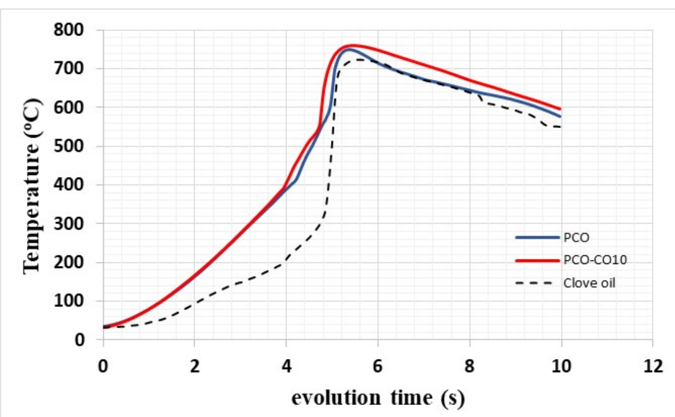

Figure 4. Droplet Combustion Result

The measurement result of temperature droplet combustion is shown in Figure 4, in which the correlation of evolution time and the temperature was described in the graph for clove oil, PCO, and PCO-CO10. A similar trend in the graph in PCO and PCO-CO10 takes place due to the presence of fatty acid as the main component. The combustion process of PCO and PCO10 develop in a similar range of temperature and evolution time. The presence of clove oil in PCO-CO10 led to a slightly reduce in viscosity and flash point. Therefore, evaporation and ignition are slightly faster for PCO-CO10.

Heating, evaporation, and burning are the three steps in the process of droplet combustion (Wardana, 2010). The heating process of fuel starts from the ignition of a coil heater that affects transient heating and continues heat transfer in steady-state to the droplet and leads to the evaporation phase. In this experiment, the heating process of PCO and PCO-CO10 takes place at around 400°C. Different times to reach 400°C are shown between PCO and PCO-CO10 due to the presence of clove oil that contains eugenol and terpene in PCO-CO10. However, the heating temperature range for clove oil is around 200°C due to eugenol and terpene evaporating first in that range temperature. In the evaporation phase, the ignition droplet starts in a few seconds, and the phase change to the burning process marked as temperature increases very steeply until peak temperature. In the burning process, secondary evaporation occurs in multicomponent fuel due to volatility differences in each compound. In clove oil, secondary evaporation takes place due to oxygen content and a hydroxyl group (-OH) in eugenol, which leads to evaporation at low temperatures.

Table 4. The Time of The Combustion Process

	Time (s)		
	Ignition	2^{nd} Evaporation	Peak
PCO	4.20	4.92	5.36
PCO-CO10	3.96	4.72	5.44
Clove Oil	3.72	4.60	5.60

Table 5. The Temperature of The Combustion Process

	Temperature (°C)		
	Ignition	2^{nd} Evaporation	Peak
PCO	412.81	589.55	749.97
PCO-CO10	398.52	557.02	760.62
Clove Oil	185.04	279.26	722.44

Table 4 shows, in general, that the presence of clove oil in the PCO blend makes the ignition and secondary evaporation process shorter than in PCO. It's due to eugenol with two atoms of oxygen leading to the evaporation process being shorter and easier in that phase. The ignition and evaporation time of clove oil is faster than PCO, indicating that clove oil is a volatile oil, while clove oil needs more time in the combustion process to reach peak temperature due to its highest density with aromatic structure arranged in rigid and bulky form. The density gives an indication of the specific energy of the fuel (Atabani et al., 2013). The density of PCO-CO10, slightly higher than PCO, causes the blends to reach the highest peak temperature (see Table 5). The high flame temperature develops due to the presence of the double bond carbon chain and the aromatic ring structure, which were difficult to decompose in low energy input.

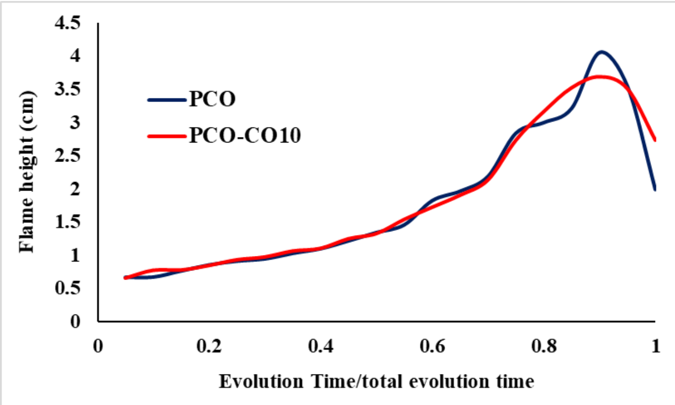

Figure 5. Flame Height Evolution

Figure 5 shows the flame height for PCO and PCO-CO10, which are measured from the flame image in Figures 3a and 3b. PCO and PCO-10 have a similar trend until evolution time in 0.8, then have the different heights in peak flame. PCO-CO10 has a height flame lower than PCO due to the presence of clove oil. Based on Table 3, The viscosity and flash point of PCO-CO10 are lower than PCO. Flashpoints indicate that PCO-CO10 is earlier on ignition than PCO. Lower viscosity in PCO-CO10 develops to easier in the combustion process with the production of low flame height.

The volatility of clove oil that contains a hydroxyl group (-OH) in eugenol is a strong oxidizing agent that leads to perfect combustion. The vapor phase in PCO-CO10 is faster than PCO, which exhibits the burning process that involves oxygen in the atmosphere. The oxygen in the atmosphere grasps the volatility compound rapidly and the formation short of flame height takes place. The buoyancy force that transports the convective heat generated by the coil heater mixes oxygen with the fuel vapor and burns the droplet out.

3.2 Thermogravimetric Analysis

Thermogravimetric analysis has been tested with represent data of the mass percentage in Table 6, Table 7, and thermogram in Figure 6. Mass loss is the consequence of increasing temperature in the boiling phase or evaporation during TGA testing.

Table 6. Mass Loss in TGA Tested

% Mass	Temperature (°C) PCO	PCO-CO10	Clove Oil
100	30	30	30
80	390.98	378.34	159.32
60	404.09	399.38	177.26
40	413.47	410.89	189.81
20	423.35	421.52	199.72

Based on Table 6, clove oil has the lowest temperature to reach 20% mass percentage. The temperature range 159 to 199.72°C in clove oil decomposition is included in the classification of high volatile oil. The temperature of decomposition in PCO-C10 is slightly lower than in PCO due to the presence of clove oil that supports the fuel blend being easier to evaporate. Overall, PCO and PCO-CO10 have temperature decomposition in the classification of medium volatile oil (200-600°C). The residual inorganic is present in the remaining mass during decomposition at 900°C. It can be seen in Table 7 that the percentage of remaining mass with PCO has the highest at 3.5%.

Table 7. Remain Mass in Maximum Temperature

Oil	Temperature (°C)	Remain Mass (%)
PCO	900.25	3.5
PCO-CO10	906.17	0.63
Clove Oil	902.31	0.45

A comparison of PCO and clove oil volatility are shown in Figure 6. The thermogram plot is more than a single continuous step due to the multi-component contained in the fuel. Chain length, a branch of the chain, and degree of unsaturation are the factor that influences the thermo-oxidative properties of fatty ester (Borugadda and Goud, 2014). Based on Table 1, PCO contains unsaturated fatty acid in 17%, which is more unstable than saturated due to the presence of a double bond. Remain components of PCO, which contain saturated fatty acid, have decomposed and lost in range temperature 400-500°C. The non-volatile fraction in remain sample consists of fixed carbon and ash, which create the curve-flattering, and mass loss becomes stagnant in the range of temperature 600-900°C.

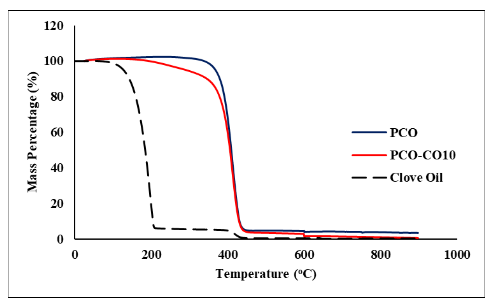

Figure 6. Thermogram of PCO and its Blend

Eugenol and terpene evaporate from PCO-CO10 at an early stage of the healing process in a range temperature of 200-400°C. Eugenol evaporated easier than fatty acid due to the benzene ring with resonance activity. This activity is called the delocalization of electrons, which could increase stability when thermal energy is supplied. In the range temperature 400-900°C, the thermogram exhibited the same trends as those observed in the case of the PCO sample. The presence of clove oil in PCO-CO10 led to decreasing curve in a sloping trend between 200-400°C. The mass loss of clove oil during the heating process in range temperature 100-200°C more than 80% demonstrates that clove oil is a high volatile oil.

The onset temperature is defined as begin temperature of decomposition in the fuel sample (Borugadda and Goud, 2014). It was determined in a thermogravimetric curve as the intersection point of the tangent to two branches and related to thermal stability (Pelegrini et al., 2017). In the experiment result, the onset temperature of PCO, PCO-CO10, and clove oil are 385.04°C, 381.30°C, and 151.40°C, respectively. The thermal stability of PCO is higher than PCO-CO10 due to the presence of unsaturated fatty acids, mainly oleic acid and linoleic acid. The presence of clove oil led PCO to become unstable and need lower heat to decompose. The lowest onset temperature for clove oil indicates more volatile matter in this oil. The more the volatile degradation product creates, the lower the onset temperature (Nik et al., 2005).

4. CONCLUSIONS

The droplet combustion and thermal behavior of pure coconut oil (PCO), clove oil, and their mixture were experimentally investigated in order to understand the fundamental of flame evolution and thermal properties for multicomponent fuel. The ovoid flame of the fuel droplet takes place until 0.40 seconds, attributed to the volatility compound that abstraction in PCO and PCO-CO10. In PCO, lauric acid was evaporated first due to its single carbon chain structure, which is an easier abstraction than a double carbon chain. The sequence of the component which is evaporating in PCO-CO10 combustion, namely eugenol, terpene, lauric acid, and unsaturated fatty acid. Soot tendency in the burning process of clove oil has formed the flame without tip round or open tip. In thermal stability, clove oil was the lowest onset temperature, which indicates more volatile matter, and PCO has the highest thermal stability due to the fatty acid component in their fuel.

5. ACKNOWLEDGMENT

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