

Alternative Liquid Fuel from Catalytic Cracking of Candlenut Oil Biodiesel (*Aleurites Moluccana*) and PP/PS Plastic Waste

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Abstract: In this research, synthesis and characterization of alternative liquid fuel obtained from candlenut oil biodiesel (*Aleurites moluccana*) and polypropylene (PP) combined with polystyrene (PS) plastic waste were carried out. This research aims to obtain biodiesel that is more environmentally friendly when compared to conventional diesel fuel today. The synthesis and characterization process was performed through catalytic cracking using a combination of Al-MCM-41 catalyst and ceramics, with a ratio of 7:3, respectively. This catalytic cracking method has been known to produce biodiesel because it can break long-chain carbon bonds to be shorter and simpler. The morphology and acidity of the catalysts were analyzed using XRD and FTIR-Pyridine techniques. The fuel liquid obtained from synthesis was analyzed using Gas Chromatography-Mass Spectroscopy (GC-MS) to obtain its constituent hydrocarbon components fraction. The highest yield was obtained at a feedstock of PS:COB (candlenut oil biodiesel) variation of 68.2%, with hydrocarbon fraction composition C₇, C₇-C₁₂, > C₁₂ each respectively of 14.66%, 85.32%, and 13.05%. The viscosity and the calorific value of the liquid fuel were initially known. In this research, the effects of using other plastic waste towards the yield percentage and the characteristics of the synthesized fuel were also investigated. The results from the analysis show that the properties of the synthesized liquid fuel mixture variations being researched have met the Indonesian National Standard SNI 06-3506-1994, which refers to the quality standards of gasoline-type liquid fuel.

Keywords: alternative fuel, candlenut oil biodiesel, polypropylene waste, polystyrene waste, catalytic cracking.

來自小燭樹油生物柴油 (油桐) 和聚丙烯/聚苯乙烯塑料廢料催化裂化的替代液體燃料

摘要: 在這項研究中, 對由石蠟油生物柴油 (油桐) 和聚丙烯與聚苯乙烯塑料廢料結合獲得的替代液體燃料進行了合成和表徵。這項研究旨在獲得與當今傳統柴油燃料相比更環保的生物柴油。合成和表徵過程是通過催化裂化使用一個 1-物質的流動組成-41 催化劑和陶瓷的組合進行的, 比例分別為 7:3。眾所周知, 這種催化裂化方法可以生產生物柴油, 因為它可以將長鏈碳鍵破壞得更短更簡單。使用 X 射線衍射分析和傅里葉變換紅外光譜法-吡啶技術分析催化劑的形態和酸度。使用氣相色譜-質譜對合成得到的燃料液體進行分析, 以獲得其組成烴組分。在聚苯乙烯: 核桃油生物柴油原料變化為 68.2%, 烴組分組成 C₇、C₇-C₁₂、> C₁₂ 分別為 14.66%、

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85.32% 和 13.05% 時獲得最高產率。液體燃料的粘度和熱值最初是已知的。在這項研究中，還研究了使用其他塑料廢物對產量百分比和合成燃料特性的影響。分析結果表明，所研究的合成液體燃料混合物的特性符合印度尼西亞國家標準 06-3506-1994，該標準是指汽油型液體燃料的質量標準。

关键词：替代燃料、燭油生物柴油、聚丙烯廢料、聚苯乙烯廢料、催化裂化。

1. Introduction

The economic development and the increase of population in Indonesia caused the demand for transport to increase significantly annually, causing the additional demand for liquid fuel, particularly gasoline. On the other hand, this condition is not being balanced with the availability of crude oil in Indonesia, which only remains at 3.6 billion barrels, which is predicted to finish in the next 13 years [1]. The huge demand for liquid fuel also affects the surrounding environment due to the overexploitation of fossil fuel. On this basis, an alternative fuel that is environmentally friendly and sustainable needs to be developed to overcome these problems. A method of doing so is producing gasoline and bio-based material, more commonly known as biofuel, as a replacement for conventional fuels. One source of material that can be used for biofuel is candlenut oil [2].

Candlenut oil is an example of vegetable oil that can be obtained from a candleberry tree and could be used as a source of bio-oil with some advantages. Amongst these advantages are they are known to have a non-edible characteristic, a massive amount of seeds production, flammable, and oil content in the seeds, which reaches 55% to 66% of the total weight of candlenut seeds. Furthermore, candlenut oil that has been converted into its methyl ester form (biodiesel) has a low crystallization temperature of -11.08°C and is flammable [3].

The fuel obtained from biomass cracking (crude bio-oil) is environmentally friendly and renewable. However, crude bio-oil could not be used directly on vehicle engines due to its high level of water and oxygen, large viscosity, corrosive, and lower calorific value than fossil fuels. In addition, biomass has a low H/C_{eff} ratio, resulting in a large amount of coke during its production process. Hence, the catalytic co-pyrolysis biomass method combined with material rich in hydrogen such as polypropylene and polystyrene plastic waste is recommended to solve such a problem [4].

Polypropylene (PP) is a straight-chain olefin polymer where in terms of its annual production, it reaches 55 million tonnes. PP plastic waste is generally found in

urban wastes with 24.3% of the total plastics. The conversion of PP wastes is used as a source of hydrogen reactant, where the process of catalytic cracking could bring some benefits. Examples of these benefits include being available at a massive amount, cheap, and producing a high amount of energy, 46.4 MJ/kg [5]. In [6], a mixture of lignocellulosic biomass (LB) with PP in catalytic cracking reaction gave a maximum yield of monoaromatics and olefins of 73%. It gave an increased (H/C) ratio of the final liquid products to increase the calorific value of the oil formed, minimizing the production of coke [7].

Polystyrene (PS) is a type of plastic that comprises of polymerized styrene monomer and is commonly found in a final processing facility in the form of Styrofoam. PS waste can be converted into an alternative fuel through pyrolysis. Previous research has performed a lignocellulose biomass co-pyrolysis process using polystyrene. The addition of PS waste as a co-reactant in Karanja seeds pyrolysis positively affects the viscosity and the calorific value of the catalytic cracking product.

A type of catalyst that is most commonly used in catalytic cracking is Al-MCM-41, a material modified from MCM-41 by inserting aluminum metal into its structure to increase the acidity of the material. The catalyst acidity is pivotal for the process of catalytic cracking, in particular when involving Bronsted acid, due to the yield percentage and the selectivity of the catalytic cracking product [8]. In the previous research conducted by Juwono et al. [9], Al-MCM-41 was modified with ceramics. The results showed that it successfully increased the catalyst selectivity on the gasoline fraction hydrocarbon ($\text{C}_7\text{-C}_{12}$) that took place during the formation of the hydrocarbon fraction product, which is similar to gasoline ($\text{C}_7\text{-C}_{12}$). From the other researches Juwono et al. [9], the addition of a ceramic spark plug on the catalyst has successfully increased the surface area of the catalyst by 14.42%, in addition to increasing its pore volume.

Based on these explanations, this research was conducted by synthesizing and characterization liquid fuel obtained from catalytic cracking using candlenut oil (*Aleurites moluccana*), PP/PS plastic waste, and Al-

MCM-41 catalyst at a ratio of 7:3. This research aims to study the effects of the different types of plastic wastes being used as a source of hydrogen throughout catalytic cracking. The yield percentage and characterization of the end product were also analyzed based on the liquid fuel being produced. As an outcome of this research, it is expected that the amount of PP/PS plastic waste can be reduced annually as the amount of plastic waste keeps on rising and increases the usage values of plastic waste.

2. Materials and Methods

2.1. Materials and Apparatuses

We used a set of reflux kit, oven, dessicator, stopwatch, pycnometer, analytical balance, Ostwald viscometer, burette, stative ring clamp, pyrolysis reactor, catalytic reactor, press-pellet, calorimeter bomb (IKA C-200), XRD (PANalytical), SEM-EDX FTIR-Pyridine (Shimadzu), and GC-MS (Shimadzu).

The materials used in this research were candlenut oil (*Aleurites moluccana*) obtained from Nuriana in Sidoarjo, PP and PS plastic waste, Al-MCM-41 catalysts from the previous research performed by Juwono et al. [10], spark plug isolator ceramic, $\text{KOH}_{(s)}$ (Merck, 99%), $\text{H}_2\text{SO}_{4(aq)}$ (Merck, 99%), $\text{CH}_3\text{OH}_{(aq)}$ (Merck, 99%), MTBE (Sigma-Aldrich, 99,8%), N_2 gas, UHP, RON 88 Premium.

The flowchart in Fig. 1 explains the stages performed in this research.

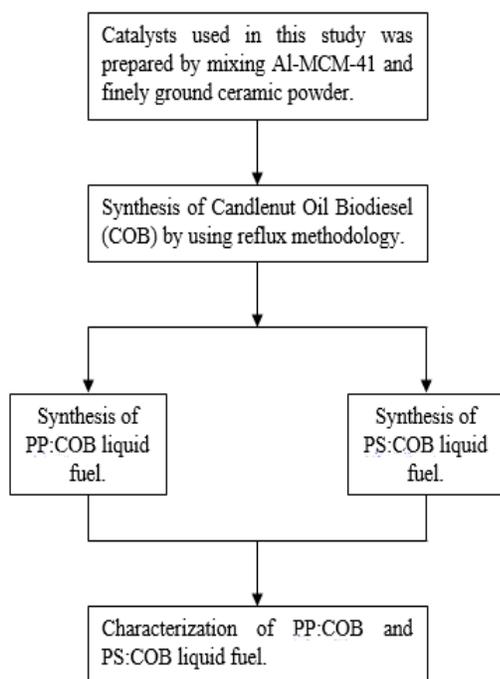


Fig. 1 Flowchart of the experiment

2.2. Preparing the Catalysts

Al-MCM-41 Catalysts and Ceramics with a ratio of 7:3 were made by mixing Al-MCM-41 catalysts from the

previous research performed by Juwono et al. [12] and finely ground ceramic powder. Al-MCM-41 and ceramic powder were both mixed at a ratio of 7:3. Next, the catalyst was activated in an oven at 80°C before cooling in a desiccator. The pellets were produced by using a pellet press. The catalysts were then characterized with XRD, SEM-EDX dan FTIR-Pyridine to study their morphology and acidity.

2.3. Candlenut Oil Biodiesel (COB) Synthesis

Candlenut oil with an FFA (5.24%) was converted into biodiesel through two-stage esterification and transesterification reaction. Both stages were performed using the reflux method. The esterification stage was performed by reacting the candlenut oil with methanol at a ratio of 1:3 (n/n) inside a round bottom flask with the assistance of a concentrated H_2SO_4 acid catalyst (4% w oil). The candlenut oil was heated until 65°C , where a mixture of H_2SO_4 -methanol was subsequently added slowly. The esterification was performed for 90 minutes at 65°C on a stirrer. The oil produced from esterification was poured onto a separatory funnel to separate the esterified oil from the catalyst and the excess methanol.

The esterified oil is then transesterified by reacting the oil with methanol at a ratio of 1:1 (n/n) with the assistance of a KOH alkaline catalyst (1% w oil). The esterified oil is poured into a round bottom flask and heated until 65°C . KOH was dissolved in methanol to form a methoxy solution in the other system. The methoxy solution was slowly inserted into the oil. Transesterification was performed for 90 minutes at 65°C on a stirrer. The transesterified oil was poured onto a separatory funnel to separate the candlenut oil biodiesel (upper phase) from the side product, glycerol (bottom phase). The synthesized biodiesel was washed with warm water until the color of the water became clear before being evaporated to omit the water contents.

2.4. PP:COB and PS:COB Liquid Fuel Synthesis

Liquid fuel synthesis was carried out in a cracking reactor specifically designed with the main component in the form of a feedstock flow and a reactor tube where the cracking took place, a condenser, and an oil container.

Candlenut oil Biodiesel (COB) and oil that have both been obtained from PS/PP plastic waste pyrolysis were initially mixed with a variation composition of 2:1, v/v, which applied to both PP:COB and PS:COB. The next step was to undergo the process of catalytic cracking using a composition of Al-MCM-41 catalyst and ceramic of 7:3 for 2 hours at a maximum temperature of 300°C .

2.5. PP:COB and PS:COB Liquid Fuel Characterization

The liquid fuel obtained from cracking was analyzed using GC-MS to study the composition of the constituent

compound of hydrocarbon fractions. The liquid fuel was mixed with MTBE and Premium RON 88 according to the compositions listed in Table 1 before being characterized for its physical properties that cover its density, viscosity,

and calorific value. The viscosity test was performed based on the ASTM D 445 testing method. Furthermore, to determine the calorific value of the liquid fuel, the bomb calorimeter method was used.

Table 1 Composition of liquid fuel mixture

Liquid Fuel Variation	Liquid Fuel Volume	MTBE Volume	Premium Volume	Liquid Fuel Mixture Code
PP:COB (2:1)	225 mL	18,4 mL	750 mL	A25
PS:COB (2:1)	225 mL	18,4 mL	750 mL	B25
-	-	17 mL	900 mL	Standard

3. Results and Discussion

3.1. XRD Characterisation Results

The characterization results of the catalysts by using the XRD method are shown in Fig. 2. The peak that appeared at 2θ above 22.5° is the natural peak of silica. At $2\theta = 25.5797^\circ$, it shows a characteristic crystal peak of Al which shifted at a range of 2θ above 20° . At the same time, at diffractogram peak of $2\theta = 25.5797^\circ$; 35.0866° ; 43.2584° , shown in Fig. 2, is a characteristic peak of ceramic powder.

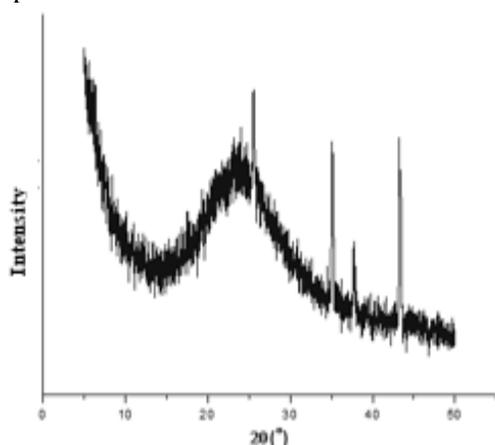


Fig 2 Diffractogram of the Al-MCM-41 catalyst and ceramic with a ratio of 7:3

3.2. SEM-EDX Characterisation Results

The aim of performing characterization on Al-MCM-41 catalyst and ceramics in a ratio of 7:3 with SEM-EDX was to study the surface morphology and the element percentage, in particular, Al and Si elements in the catalyst. The catalyst's surface morphology was analyzed using SEM, as shown in Fig. 3, while the element percentage of Al and Si catalysts are shown in Table 2.

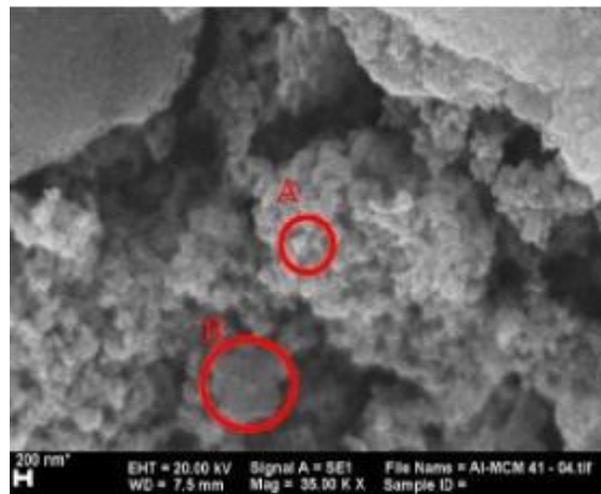


Fig. 3 Catalyst surface morphology

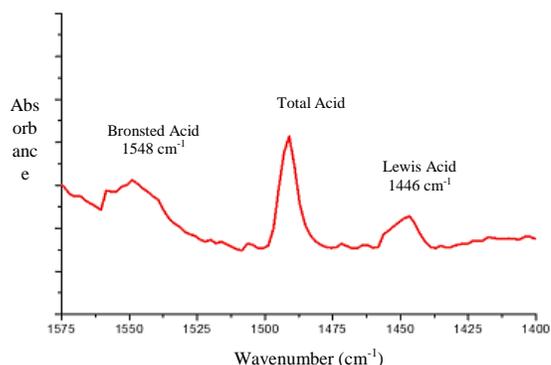


Fig. 4 The FTIR-pyridine spectrum of Al-MCM-41 catalyst mixed with ceramic

Table 2 Al and Si catalyst compositions

Al-MCM-41 Catalyst [12]		Al-MCM-41:Keramik Catalyst	
Atom	Weight, %	Atom	Weight, %
Al	0,27	Al	0,73
Si	2,62	Si	10,16

Based on Fig. 2, area A shows the appearance of the Al-MCM-41 powder, while area B shows the appearance of ceramic powder, which functioned as supporting solids or supporting catalysts.

Based on Table 2, modifying catalysts with ceramic isolated spark plug have successfully increased the

contents of Al in the catalyst. The Si/Al ratio in the catalyst was also increased to 13.92.

3.3. FTIR-Pyridine Characterisation Results

Catalyst acidity characterization using the FTIR-pyridine adsorption method is an essential process as the catalyst acidity, in particular, the Bronsted acidity, has a crucial role in catalytic cracking. The pyridine spectrum from the catalyst acidity analysis is shown in Fig. 4.

Based on the spectrum analysis results obtained, a mixture of Al-MCM-41 catalyst and ceramic with a ratio of 7:3 has Bronsted and Lewis acid contents of each respectively 0.0311 mmol/g and 0.0341 mmol/g.

3.4. Candlenut Oil Biodiesel Synthesis

The synthesis process was performed through 2 stage reactions: esterification and transesterification since the candlenut oil contain a high free fatty acid (FFA) of 5.24%. The esterification process is an oil pretreatment process to reduce the amount of FFA. The feedstock oil with a high FFA content could decrease the yield percentage of transesterified biodiesel. Thus based on this, the FFA content needed to be decreased beforehand [11].

The synthesized biodiesel in this research has a clear yellow color, with both the density value and flash point that met the Indonesian biodiesel standard of SNI 04-7182-2006.

3.5. Synthesis Results of PP:COB and PS:COB Liquid Fuels

Each liquid fuel variation produced a hydrocarbon fuel with a different liquid fuel and coke yield. The cracking data of liquid fuels PP:COB and PS:COB is shown in Fig. 5.

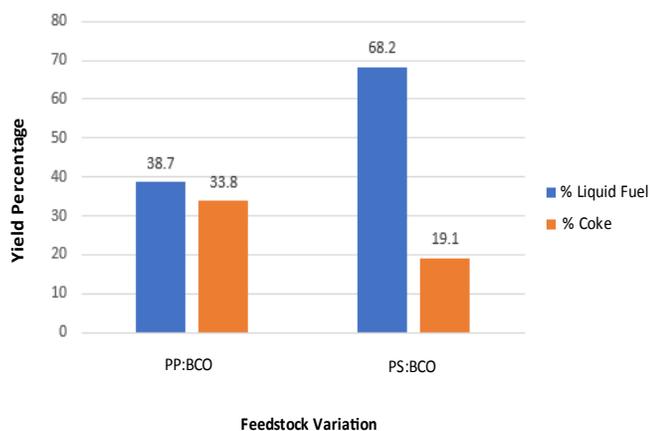


Fig 5. Liquid fuel synthesis results

Based on the graph shown in Fig. 4, the PS:COB variation has a higher liquid fuel yield percentage than the PP:COB and has a lower side product, which is coke. The lower liquid fuel yield of PP:COB is caused by the

catalytic cracking process at temperatures above 250°C that made the polypropylene plastic easier to crack into a short-chain olefin compound ($< C_4$), where it took the form of a gas at room temperature and thus hard condensed [8].

3.6. GC-MS Analysis Results

Based on the GC-MS analysis results on the liquid fuel that underwent catalytic cracking for 60 minutes, it suggested that the end product contained a gasoline fraction (C_7-C_{12}) in addition to both light ($< C_7$) and heavy ($> C_{12}$) hydrocarbon fractions. The yield of each hydrocarbon fraction in fluid catalytic cracking is shown in Table 3.

Table 3 Yield percentage of hydrocarbon fraction

Liquid fuel variation	Yield (%)		
	$< C_7$	C_7-C_{12}	$> C_{12}$
PP:COB	4.09	81.5	14.4
PS:COB	14.66	85.32	13.05

The difference in the plastic waste as a source of hydrogen throughout the cracking process did not show a significant impact on the distribution of hydrocarbon fraction in the liquid fuel compound. Both plastics showed a very similar synergy effect. The only difference is that it increased the yield percentage in the liquid fuel and minimized the side product of cracking, coal. As of this case, PS displayed a better impact when compared to PP.

The liquid fuel variation of PS:COB produced a hydrocarbon fraction similar to gasoline (C_7-C_{12}), which is higher when compared to PP:COB. These results show a slightly better synergy effect of polystyrene on biodiesel liquid fuel in increasing the fraction similar to gasoline compared to PP.

3.7. Density Values of Liquid Fuel Mixtures

Fuel density values are an important aspect to consider because they correlate with fuel combustion quality and catalyst efficiency in the cracking process. In general, the fuel with a lower density tends to be better as the fuel will quickly evaporate and combust easier by the machine. The density values of each liquid fuel mixture from catalytic cracking at each variation are shown in Fig. 6.

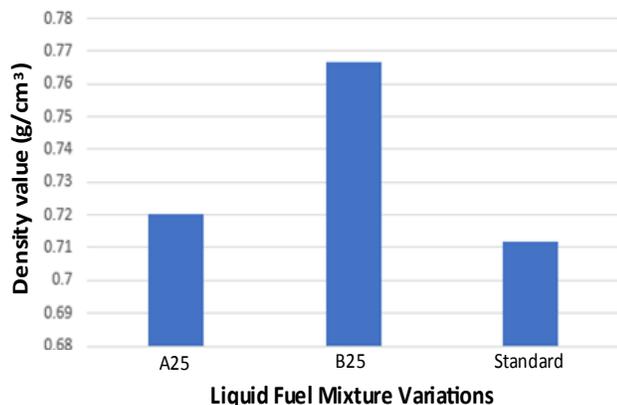


Fig. 6 Liquid fuel mixture density

Based on Fig. 6, the density value of liquid fuel mixture A25 and B25, respectively, are 0.7202 g/cm³ and 0.7668 g/cm³. Both variations of the liquid fuel mixture have a density value above the standard value of 0.7118 g/cm³. However, this density value is still within the range of the allowable density value mentioned in the gasoline quality. According to the Indonesian National Standard of SNI 06-35-06-1994, both A25 and B25 fuel mixtures have an allowable density value of 0.715 g/cm³ to 0.770 g/cm³, respectively.

B25 liquid fuel mixture has a higher density value than A25 liquid fuel mixture due to the density property of the liquid fuel produced from catalytic cracking of PS, which has a higher density when compared to PP. This theory also matches the previous research conducted by Assari [13] and Nugroho [14].

3.8. Determining the Viscosity of Liquid Fuel Mixture

The liquid fuels obtained from cracking mixed with Premium RON 88 and MTBE were then tested for their viscosity. Determining the viscosity is essential as it correlates with the fuel injection process. A high viscosity value would likely lead to a problem as this affects the flow of the fuel inside the injector and the degree of fuel atomization inside the combustion chamber [15]. The process of obtaining the viscosity of the liquid fuel in this research was performed using an Ostwald viscometer. The viscosity values of the liquid fuel are shown in Fig. 7.

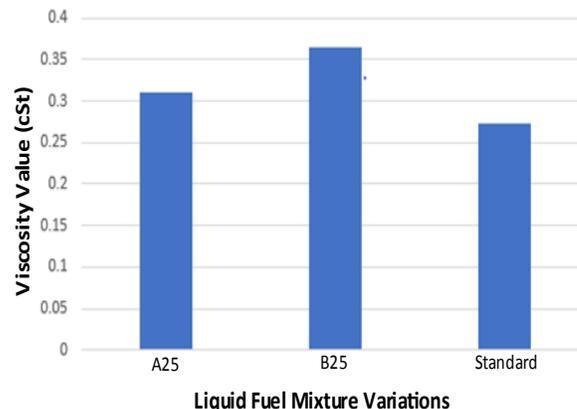


Fig. 7 Viscosity values of different liquid fuel mixture variations during the combustion

From Fig. 7, it is exhibited that the viscosity of the different fuel mixtures of A25 and B25 are 0.3095 cSt and 0.3651 cSt, respectively. Each fuel mixture has a higher viscosity than the standard liquid fuel mixture, 0.2729 cSt. However, the viscosity of A25 and B25 has met the standard viscosity for gasoline fuel when referred to the ASTM D-445 standard, which mentions a maximum allowable viscosity of 1.17. This result correlates to the hydrocarbon fraction composition of liquid fuel constituents, particularly the hydrocarbon fraction > C₁₂. Each liquid fuel has a higher hydrocarbon fraction of > C₁₂ than the 10%, which caused the viscosity value to be higher than the standard viscosity value.

The liquid fuel mixture with the mixture of feedstock PS:COB has a higher viscosity value compared to the liquid fuel mixture with the mixture of feedstock PS:COB. This is an outcome of the cracking of liquid fuel in the PS:COB reactant having a main component in the form of a derivative compound of styrene, which can easily depolymerize. The polymerization reaction formed a complex compound with a higher viscosity.

3.9. Determining the Calorific Value of Liquid Fuel Mixture

The calorific value of liquid fuel was determined using the bomb calorimeter apparatus. The calorific values of each liquid fuel mixture variation are shown in Fig. 8.

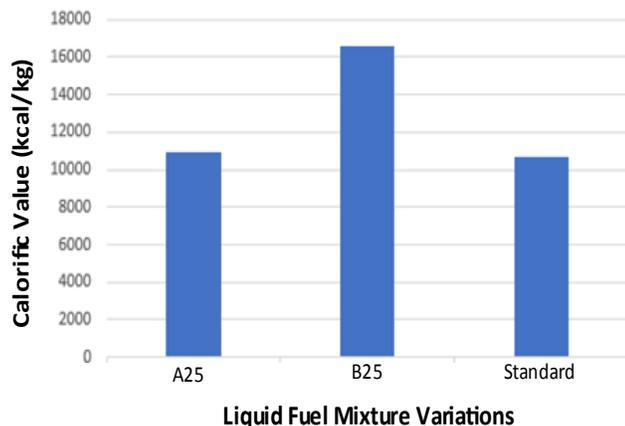


Fig. 8 Calorific values of liquid fuel mixture variations

Based on Fig. 8, the calorific value of liquid fuel mixture variations of A25 and B25, respectively, are 10 960 kcal/kg and 16 590 kcal/kg. These liquid fuel mixtures have a higher calorific value than the standard liquid fuel mixture, which is 10 647 kcal/kg, seeing as there was a difference in the compound component of liquid fuel produced from catalytic cracking. In the liquid fuel, the feedstock variation of PS:COB has a dominant content of aromatic compounds with a high calorific value [16].

The calorific values of both liquid fuel mixtures have met the standard of the minimum calorific value of gasoline set by Pertamina (Indonesian State Mining Company). It states that the standard for calorific value in gasoline is 10 160 kcal/kg.

3.10. Catalytic Cracking Process

In this research, the process of catalytic cracking was performed to crack and convert the long-chain hydrocarbon from biodiesel candlenut oil with PP and polystyrene PS wastes into a light hydrocarbon fraction, in particular forming them into a fraction similar to gasoline (C_7 - C_{12}). The cracking process occurred as a result of thermal and catalytic effects, which took place simultaneously. The presence of thermal effects caused the cracking hydrocarbon molecules to become free radical fragments which tend to undergo an oligomerization reaction. Meanwhile, the role of catalysts is to catalyze the cracking reaction by forming carbonium ions that was achieved by breaking the carbon chain at the beta position from the carbonium ion. During this process, besides breaking the hydrocarbon chain, other reactions include isomerization, cyclization, aromatization, and repolymerization [17]. Using a mixture of Al-MCM-41 catalyst with ceramic at a mixture of 7:3 was also intended to increase the catalyst selectivity during the forming process of hydrocarbon fraction that is similar to gasoline (C_7 - C_{12}).

The yield difference in the liquid fuel, hydrocarbon fraction distribution, and the types of compounds in the liquid fuel due to catalytic cracking was a result of synergy effects between the biodiesel candlenut oil with the different types of plastic waste. The different synergy effects point to the possibility of different cracking effects, which also took place. The different possibilities of the reactions would also result in a product with a different compound component. In the variation of PP:COB, the dominant compound produced from catalytic cracking was an open-chain olefin compound and saturated aliphatic compound. Meanwhile, in the PS:COB variation, the dominant compound found was an aromatic compound and a non-aromatic cyclic.

4. Conclusion

Liquid fuel as a product of catalytic cracking from candlenut oil biodiesel (COB, or *Aleurites moluccana*) along with PP and PS plastic wastes have been successfully synthesized and characterized. The cracking process was assisted by a mixture of Al-MCM-41 catalyst and ceramic at a ratio of 7:3. The outcomes of this research confirmed that the different plastics used (PP or PS) affected the liquid fuel yield, hydrocarbon fraction distribution in the liquid fuel product itself, and other physical properties (density, viscosity, and calorific values). Based on the analysis by using the method of liquid fuel yield and GC-MS data, the liquid fuel variation of PP:COB produced a liquid fuel yield of 68.2%, with each content of hydrocarbon fraction $< C_7$ of 4.09%, C_7 - C_{12} of 81.5%, and $> C_{12}$ of 14.4%. Simultaneously, the liquid fuel variation of PS:COB resulted in a liquid fuel yield of 38.7%, with each hydrocarbon fraction content of $< C_7$ of 14.66%, C_7 - C_{12} of 85.32%, dan $> C_{12}$ of 13.05%. Based on the density, viscosity, and calorific values tests, all of these have revealed that an alternative liquid fuel produced by catalytic cracking has properties that met the Indonesian National Standards of SNI 06-35-06-1994, which refers to the fuel quality of gasoline type.

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