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Synthesis of Pyrogallol Derivative as a Soluble Antioxidant for Biodiesel Additive

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Abstract. Biodiesel as a promising alternative fuel has a major technical drawback which is its instability during storage. One of the solutions to this problem is the addition of antioxidants to improve storage stability. However, the commonly used antioxidant, pyrogallol, has poor solubility in biodiesel due to its polarity difference with biodiesel. Hence this research is focused on modification of pyrogallol with methyl linoleate through a free radical mechanism using benzoyl peroxide as the radical initiator to increase pyrogallol solubility. The presence of the proposed product (10E,12Z)-9-(2,3,4-trihydroxyphenyl) octadeca-10,12-dienoate and its isomer methyl (9Z,11E)-13-(2,3,4-trihydroxyphenyl) octadeca-9,11-dienoate was confirmed using TLC, HPLC and LC-MS/MS which yielded 0.2%. UV-Vis Spectrophotometer was used for the solubility test and the result showed that the solubility limit of the product 2500ppm which was higher than pyrogallol. For the antioxidant activity, the Rancimat test was performed to see the product's performance in accelerated oxidation as well as acid value determination during four weeks of storage. The Rancimat test result shows the product has higher induction period compared to pyrogallol and fulfilled the oxidation test standard. However, the product showed lower acid value stability compared to pyrogallol and biodiesel.

1. Introduction

Biodiesel, also called Fatty acid methyl ester (FAME) is a renewable fuel derived from biological sources through transesterification reaction that has lower smoke emission [1]. However, apart from the advantages biodiesel has, it has a major technical drawback that affects biodiesel's quality during storage [2]. The high content of unsaturated fatty acid chains and the presence of carbon double bonds in the biodiesel leads to high reactivity with O_2 [3]. Biodiesel oxidative degradation leads to acid sediments formation which causes blockage in engine fuel line and filters. Therefore, antioxidants are required to improve the oxidation stability of biodiesel. The most used Phenolic antioxidants are Pyrogallol, Propyl gallate, TBHQ, BHA and BHT[4]. Due to its long hydrocarbon chain, biodiesel is relatively non-polar. In contrast, phenolic antioxidants which are hydroxyl groups are polar compounds. The difference in polarity between biodiesel and phenolic compounds creates an

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electronegativity gap which results in low solubility of phenolic antioxidants in biodiesel [5]. In recent years, there has been a rise in research regarding the modification of phenolic compounds to increase the solubility of phenolic antioxidant. One of them is phenolic compound modification using a free radical mechanism [6]. One of the commonly used initiators for free radical mechanism is benzoyl peroxide. Upon heating, benzoyloxy radicals were formed from the dissociation of benzoyl peroxide which will initiate the reaction [7], [8]. In this research, pyrogallol will be modified using methyl linoleate using free radical mechanism with benzoyl peroxide as the initiator. The new product is theoretically expected to have a higher solubility than pyrogallol.

2. Research Methods

2.1. Preliminary Reaction

The optimization of the coupling reaction between pyrogallol and methyl linoleate was performed by variating the temperature and the sequential addition of methyl linoleate. The reaction was conducted using a 100 ml boiling flask connected to reflux and equipped with a magnetic stirrer. The result of the reaction was qualitatively observed with thin layer chromatography (TLC).

2.2. Coupling Reaction

The synthesis reaction was prepared by weighing 0.24 g of Benzoyl Peroxide into a boiling flask and diluting it in 10 ml of Ethyl Acetate. Afterwards, 0.33ml of methyl linoleate was added into the flask. The air inside was purged by injecting nitrogen into the flask to remove all the oxygen inside. The mixture was then heated until 90°C in a water bath and stirred continuously for 2 hours. Next, in a 50 ml beaker, 0.126 g of Pyrogallol was diluted in 1ml of ethyl acetate. The pyrogallol solution was then added to the reaction mixture and was stirred for another 15 minutes.

2.3. Solubility Test

The reaction mixture was added to biodiesel with various concentrations and diluted in *n*-hexane with 1:100 dilution factor. Each diluted sample solution were then scanned using UV/Vis spectrophotometer to find the wavelength of the maximum absorbance. Lastly, the absorbance of each concentration was measured in that wavelength where the absorbance will keep increasing until a certain concentration where it will remain constant or stopped increasing which indicates the antioxidant solubility limit in biodiesel. The solubility limit data will later be compared to the solubility limit data of pyrogallol

2.4. Antioxidant Activity Qualitative Analysis

The antioxidant qualitative analysis uses the method of dot blot assay. First of all, a solution of methanolic DPPH was prepared, 0.1 g of DPPH was diluted in 50 ml methanol in a volumetric flask to make a 0.2% methanolic DPPH solution. Afterward, pyrogallol and product prepared and diluted to ethyl acetate to make a solution with a concentration of 1500 ppm. The pyrogallol and the product were then spotted to TLC paper then sprayed with the methanolic DPPH solution prepared earlier. The color change was then further analyzed.

2.5. Rancimat Test

The Rancimat method is also called the automated Swift test or accelerated oxidation test was conducted to determine the effectiveness of antioxidants to restrict biodiesel to oxidation. The test was performed using Metrohm 873 based on standard test method EN 14112 [2], [9]. The method involves passing airflow at a steady rate of 10l/h and heating with a constant temperature of 110 °C which leads to the oxidation of biodiesel, with volatile, water-soluble short chain carboxylic acids formed as the oxidation products. The oxidation products were transported in the stream of air to a vessel containing distilled water and the conductivity of this vessel was continuously monitored. The time duration of steep increase in the conductivity was measured in rancimat method, it is known as the induction period (IP) and it has been an element of standard for biodiesel oxidative stability; in EN 14214 biodiesel fuels must meet an induction period requirement of 6 h and in ASTM D6751 must meet an induction period requirement of 3 h.

2.6. *Acid Value Analysis*

Both pyrogallol and the product are subjected to acid value analysis, a sample with 1500 ppm concentration mixed with biodiesel were prepared for each antioxidant. A sample of pure biodiesel was also prepared as a comparison with the antioxidant. The samples were labeled: Biodiesel (B), Biodiesel-Pyrogallol (B-PY) and Biodiesel-Product (B-PR). The samples were analyzed weekly during four weeks of storage time. In an Erlenmeyer flask, 10ml of 2-Propanol was prepared. Afterward, 1 ml of sample was added and mixed until homogeneous. A calibrated pH meter was prepared. Then, the solution was titrated with a standard 0.1 N NaOH with increasing volume to obtain the diagram of NaOH volume used to make the pH 7 (V1). After plotting the data, the required NaOH volume to change the pH of the solution into 7 was calculated using the equation of the curve. The acid values method was a modification of Indonesia National Standard for oil and lipid SN1 01-30199. Titration for 2-propanol was done to neutralize the 2-propanol slight acidity. The volume of NaOH used to neutralize 2-propanol was noted as (Vo). At last, the acid values of the samples were calculated using the equation below:

$$Acid\ Value = \frac{V\ x\ N\ x\ 56.1}{m} \tag{1}$$

3. Results and Discussion

3.1. *Preliminary Reaction*

Benzoyl Peroxide as a thermal initiator required heat to decompose, therefore to obtain the optimum condition for the reaction, variating the temperature reaction was performed. According to Odian (2004) benzoyl peroxide half-life ($t_{1/2}$) at 70℃ was 7.3 h, which is why the TLC spots did not show a positive result because the benzoyl peroxides did not encounter enough heat to decompose. The sequence addition of methyl linoleate was also varied because methyl linoleate as a long chain methyl ester took longer time to become radical therefore reaction with addition of methyl linoleate before and after heating was observed. From the variations, the optimum condition of the reaction was observed to be at temperature 90℃ with the addition of methyl linoleate before heating.

Table 1. Results of Preliminary Reaction

Temperature (°C)	Methyl linoleate addition	New Spots and Intensity
90	After heating	+
	Before heating	++
70	After heating	-
	Before heating	-

3.2. *Coupling Reaction*

Synthesis of pyrogallol with methyl linoleate was executed via coupling reaction by using benzoyl peroxide as the initiator of the reaction. In the synthesis reaction, ethyl acetate was used as a solvent because it is a semi-polar solvent that can dilute all the components for this reaction. Benzoyl Peroxide was heated for 2 hours in a water bath with a temperature of 95°C to make sure that the benzoyl peroxides decomposed into benzoyloxy radicals. During the whole reaction, reflux was used to condense the ethyl acetate because of its boiling point at 78°C. Nitrogen was used because during heating, methyl linoleate radical tends to bond with oxygen. Therefore, oxygen inside the flask was removed to create an optimum condition for the reaction. The reaction mixture between methyl linoleate and benzoyl peroxide produced a transparent non-viscous solution. Pyrogallol was not heated together with benzoyl peroxide and methyl linoleate because pyrogallol as secondary alcohol would decompose into ketone upon heating which would cause pyrogallol to lose its antioxidant ability. When pyrogallol-ethyl acetate solution was added into the benzoyl peroxide and methyl linoleate solution, the color of the product mixture changed into light brown transparent solution. The color change occurs because some of the pyrogallol might have absorbed some oxygen (Graf, 2006). After a few hours, the color of the solution changed to dark red-black color which indicates that some of the pyrogallol might have been oxidized.

3.3. Thin Layer Chromatography (TLC) Identification

As a pre-identification, the reaction product was subjected to a Thin Layer Chromatography (TLC) analysis to observe the retention factors difference. The solvent used was a combination of n-hexane and ethyl acetate with a ratio of 1:5. The combination was chosen because ethyl acetate is categorized as a semi-polar solvent and n-hexane is a polar solvent which results in a fairly non-polar mobile phase. The stationary phase of the TLC paper was the polar silica gel.

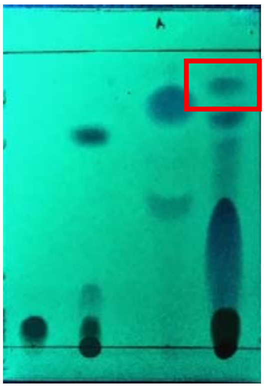

Figure 1. TLC Analysis Result

In the TLC paper, spot (a) was pyrogallol, (b) was benzoyl peroxide, (c) was methyl linoleate, and (d) was the product. The calculated Rf shown on the reaction mixture spot were (a) was 0.1, benzoyl peroxide (b) 0.73, methyl linoleate (c) showed 2 spots one with an Rf value of 0.55 and the other one with an Rf value of 0.83. The product (d) showed 4 spots, each has Rf value from the bottom to the top were 0.13, 0.5, 0.78 and 0.9. Since the predicted product was a combination of pyrogallol and methyl linoleate, the Rf value of the product should be greater than pyrogallol as the product will be more non-polar compared to pyrogallol. Hence, the Rf value of 0.5 or 0.9 is still possible for the predicted product.

To determine which of these Rf values belong to the product, partition coefficient (log P) can be used as an indicator to determine how hydrophilic or hydrophobic a molecule is. A polar compound will have a lower log P and non-polar compounds will have higher log P. Log P of the predicted product was calculated in ChemDraw which was (a) 6.58. Pyrogallol as a polar molecule has a log P value of (b) 0.87 and methyl linoleate has a log P value of (c) 6.23 this confirms that the predicted product was more non-polar than both pyrogallol and methyl linoleate. Therefore, the Rf value of the product taken was 0.9 because the spot was higher or in other words more non-polar than methyl linoleate.

3.4. HPLC Identification

In order to further identify the product, HPLC analysis was performed at a wavelength of 280 nm and 0.5ml/min flow rate. The qualitative analysis was performed by comparing the retention time between the product, pyrogallol, methyl linoleate, and benzoyl peroxide.

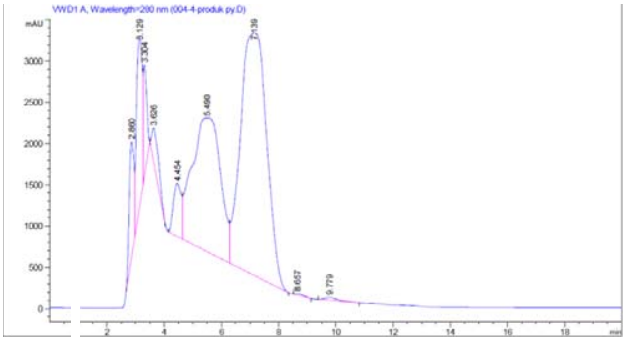

Figure 2. HPLC Spectrum

From the analysis, there were 9 appearing peaks as shown in figure 2. The retention time of pyrogallol was 2.867 min, which means that peak with retention time 2.860 can be eliminated as pyrogallol. Peak with retention time of 7.139 has a similar retention time to benzoyl peroxide retention time which is7.263 min, Methyl linoleate has a retention time of 8.399 min, and as the predicted product has higher Log P value than all methyl linoleate, pyrogallol and benzoyl peroxide, it is expected that the product will have longer retention time. Hence, a peak with a retention time of 9.779 min was taken out as the retention time of the product with a yield of 0.2%.

3.5. LCMS Identification

For further identification of the product, LC-MS/MS analysis was performed to determine the molecular mass of the components in the sample, the product was subjected to LC-MS/MS. The determination of the product was done using mass spectral fragmentations. The results of LC-MS/MS are shown in the figure below:

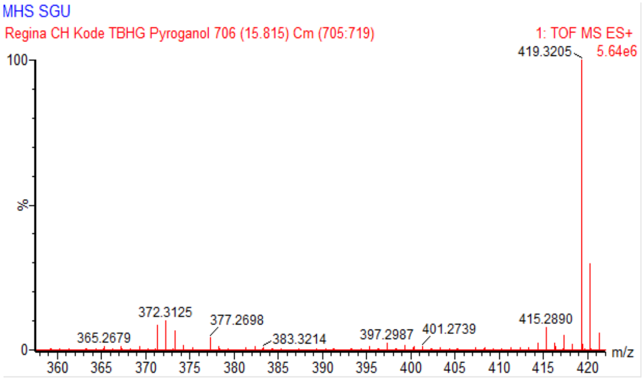

Figure 3. Mass Spectrum of LC-MS/MS

Theoretically, the predicted product ($C_{25}H_{38}O_5$) has a molecular weight of 418.589 g/mol. However, in order to be detected a molecule should be in its charged form. Therefore, metal cation or anions are commonly used to add a charge of the molecule. In this instrument (H^+) was used which results in addition of 1 m/z in order to obtain the real molecular weight of the product. From the result, it could be seen in Figure 3 the spectrum shows the candidate mass of the product ($C_{25}H_{38}O_5$) which was 419.3207 g/mol. The candidate mass number is close to the molecular weight prediction of the product, therefore the presence of the desired product can be confirmed.

3.6. Proposed Mechanism for the coupling reaction

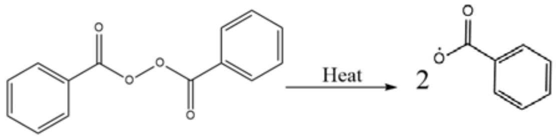

Figure 4. Benzoyloxy radical formation

The first step of radical chain reaction is the radical formation. Upon heating, benzoyl peroxide became unstable and decompose into benzoyloxy radicals. The unpaired benzoyloxy radicals became radical initiators as they are chemically reactive with the presence of other molecules. The next step is initiation, where pyrogallol and methyl linoleate as a non-radical compound became radical by the presence of benzoyloxy radicals. Free-radical molecules are not stable, therefore both pyrogallol and methyl linoleate will go through radical resonance or delocalization to stabilize the molecule.

Pyrogallol as the main as a phenolic compound has three -OH substituents. A substituent on a benzene ring had an effect on the additional substituent placement, which was the substitution of hydrogen with methyl linoleate radical. The substituent of pyrogallol affected the orientation of the new substituent. In general, when the substituent group attached to the benzene ring had at least one pair of the unshared electron, it is *ortho-para* directing. On the contrary, when the substituent group has either a partial positive charge or full positive charge, it is *meta* directing [10]. In this situation, the substituents attached to the ring was a hydroxyl group (-OH) with two pairs of unshared electrons, which was an *ortho-para* director. Since pyrogallol had three -OH substituents, there were several possibilities of the methyl linoleate radical orientation with each -OH substituent directing *ortho-para*.

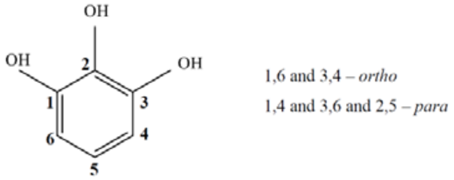

Figure 5. Orientation possibilities of methyl linoleate in pyrogallol

As seen in the figure above, atom C-4 and C-6 had more possibilities to be substituted compared to C-5. Hence, in Figure 6 the radical delocalization mechanism proceeded towards the substitution of atom C-4 (or C-6 which can be countered as C-4 because pyrogallol is a symmetric molecule), which was a *para* position from -OH number 1 and *ortho* position form -OH number 3.

The resonance delocalization of pyrogallol after radical attack by benzoyloxy radical can be seen in the figure below:

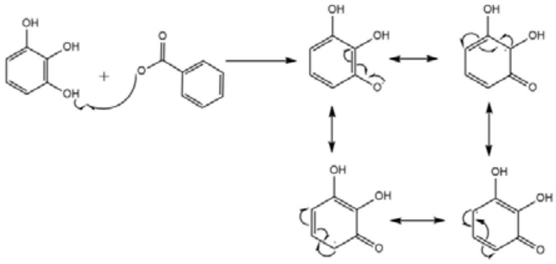

Figure 6. Pyrogallol resonance delocalization after the radical attack

From figure 6, it can be seen that there were two forms of product, in keto and in enol form. For aldehydes and ketones, keto form is considered because of it more stable compared to enol form. However, in phenolic compound, the keto-enol equilibrium lies entirely on the enol side due to thermodynamic stabilization by the aromaticity [11].

Methyl linoleate as a polyunsaturated fatty acid with two double bonds located in C-9 and C-13 atom has methylene hydrogen in C-11 atom, which is prone to radical attack. When C-11 became radical, resonance delocalization will occur to the C-9 and C-13 atom as illustrated in Figure 7.

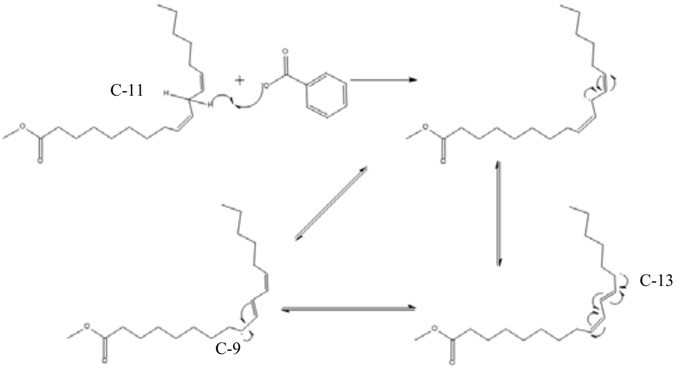

Figure 7. Methyl Linoleate resonance delocalization after the radical attack

Theoretically, with the optimum condition and proper mixing of the reaction, the radical pyrogallol and radical methyl linoleate were expected to encounter the last step of radical polymerization which is the coupling termination, where pyrogallol and methyl linoleate forms a bond as seen in Figure 8 and Figure 9. The product from the reaction will be a pyrogallol molecule attached by a methyl linoleate molecule.

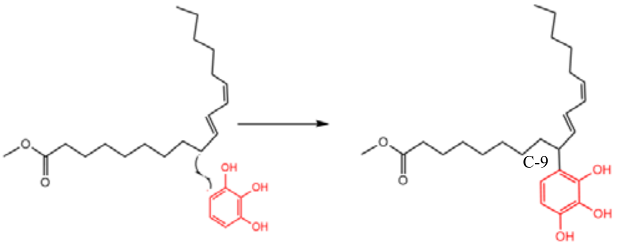

Figure 8. Termination of radical pyrogallol and radical methyl linoleate in C-9

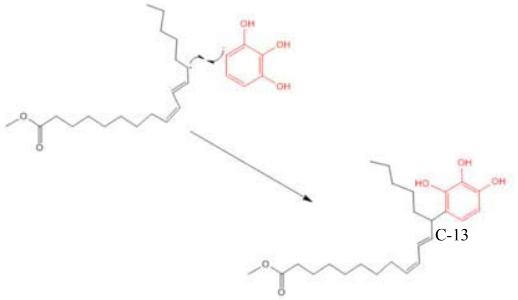

Figure 9. Termination of radical pyrogallol and radical methyl linoleate in C-13

3.7. Solubility Analysis

In this research, pyrogallol was synthesized with methyl linoleate with the purpose to increase its solubility in biodiesel. Therefore, the reaction product was subjected to a solubility test to compare the solubility limit of pyrogallol and the reaction product.

Before the solubility measurement, pyrogallol, methyl linoleate, and the new product were scanned in the UV-VIS spectrophotometer with a wavelength range of 190 nm to 400 nm to determine each maximum wavelength. Two concentrations (lowest and highest) were used to identify the peak. Based on the scanning result, the wavelength of maximum absorbance was 274 nm for the reaction product and 205 nm for methyl linoleate. After the maximum wavelengths were obtained, both pyrogallol and the reaction product was weighed in various concentrations ranging from 500ppm to 3000 ppm. Based on observation, all concentrations of pyrogallol and the reaction product were visually soluble in biodiesel. Hence, the solubility was measured with UV-Vis Spectrophotometer. A dilution of 1:100 was used to measure the absorbance value of each concentration.

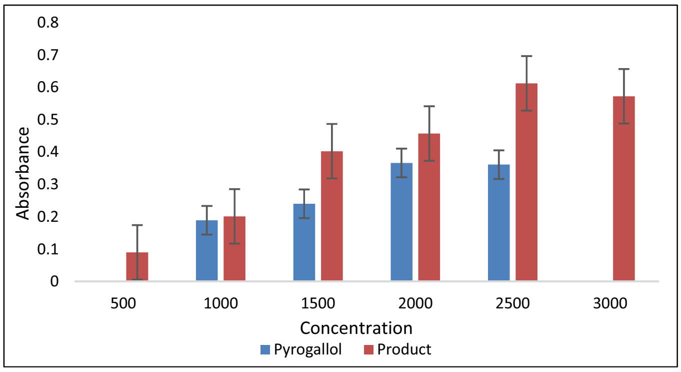

Figure 10. Solubility limit of pyrogallol and the reaction product

As seen in Figure 10 the absorbance value of pyrogallol stopped increasing at 2000 ppm, which implied that the concentration of the antioxidant did not add up in biodiesel after 2000 ppm. Therefore, it could be assumed that pyrogallol's solubility limit in biodiesel was 2000 ppm or 0.002 g/ml in biodiesel. The same analysis was performed with the reaction product's absorbance. The absorbance of the reaction product keeps increasing until 2500 ppm, while pyrogallol's absorbance stopped increasing at 2000 ppm. Hence, the reaction products solubility limit was 2500 ppm or 0.0025 g/ml in biodiesel. This concludes that the solubility of the reaction product in biodiesel was higher than pyrogallol.

3.8. Antioxidant Qualitative Analysis

Although the main objective of this research was increasing the solubility of pyrogallol in biodiesel, the antioxidant activity of the new product must also be evaluated to justify that the new product did not lose its antioxidant ability. Therefore, pyrogallol and the reaction product was analyzed qualitatively. The purpose of this qualitative analysis is to evaluate the DPPH change of color.

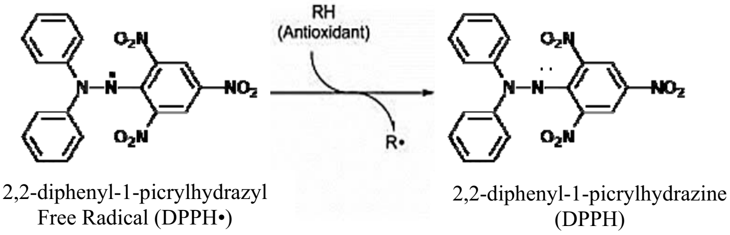

Figure 11. DPPH Antioxidant Mechanism

DPPH as a stable free radical has a violet color in methanolic solution. In the presence of an antioxidant, the color violet (2,2-diphenyl-1-picrylhydrazyl) will change into yellow (2,2-diphenyl-1-picrylhydrazine) because DPPH as a free radical will react with hydrogen which leads to DPPH losing its free radical property. The results showed, pyrogallol as one of the best antioxidants indicated the presence of antioxidant activity with the presence of color change as expect. As for the reaction product, the color of the methanolic DPPH also changed from violet to yellow, which can be qualitatively concluded that the new product had antioxidant activity.

3.9. Rancimat Test

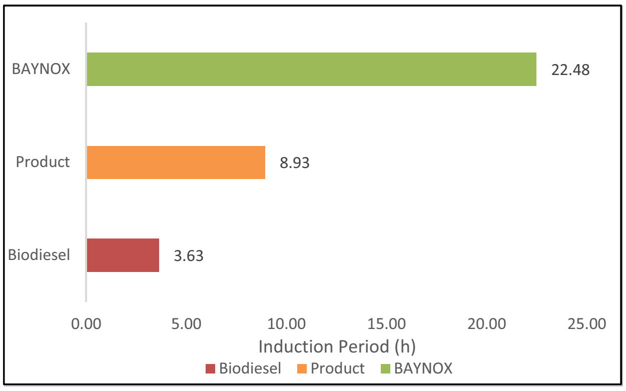

Figure 12. Induction period results from rancimat test

The Rancimat test was conducted based on EN 14112 method, the samples prepared was blank biodiesel, the reaction product, and BAYNOX as the commercial antioxidant for comparison. The concentration for BAYNOX and the reaction product was 2000 ppm in biodiesel. The result confirmed that the reaction product can slow down the oxidation of biodiesel as the reaction product has a higher induction period compared to blank biodiesel. Based on the international standard for biodiesel, the induction period standard successfully fulfills the ASTM D6751 (United States) standard which is induction period higher than 3 h and successfully fulfilled the national standard of biodiesel SNI 7182 which was induction period higher than 8 h.

3.10. Acid Value

Acid Value is used as an indicator to determine the amount of free fatty acid (FFA) in biodiesel sample. Acid value monitors the degradation of biodiesel during storage since acid value increases with degradation. The Acid value was calculated using Eq. 1.

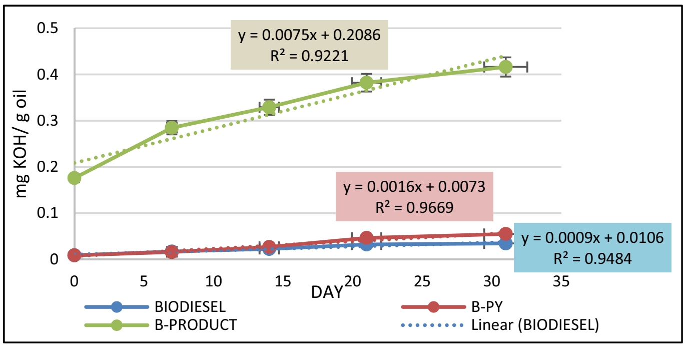

Figure 13. Acid value vs Biodiesel additive

As seen on Figure 15, biodiesel with no additives and biodiesel-pyrogallol has similar acid values (around 0.0089) at day 0, while the acid value of the biodiesel- reaction product was much higher which is 0.17. The reaction product had much higher acid value compared to biodiesel and biodiesel-pyrogallol, possibly because of the formation of formic acid which was a product from biodiesel secondary oxidation. The oxidation of biodiesel might have been caused by the remaining unreacted benzoyl peroxide in the mixture since benzoyl peroxide is a strong oxidizing agent.

All samples have an increase in acid value during the four weeks of analysis. On the 31st day, biodiesel-product has a higher acid value compared to biodiesel-pyrogallol and biodiesel with no additives which doesn't show a positive result of the product in slowing down the increase of acid number caused oxidation. However, the biodiesel-product at the end of analysis has an acid value of 0.41 which was not more than 0.5. Based on EN 14214 and ASTM D6751 acid value of biodiesel must not exceed 0.5 to fulfill the antioxidant requirement, thus the new product can still be considered as an antioxidant for biodiesel.

4. Conclusion

The synthesis between pyrogallol and methyl linoleate to produce a molecule with higher solubility in biodiesel produced a molecule (10*E*,12*Z*)-9-(2,3,4-trihydroxyphenyl) octadeca-10,12-dienoate and its isomer methyl (9*Z*,11*E*)-13-(2,3,4-trihydroxyphenyl) octadecyl-9,11-dienoate. However, from the HPLC result, it was found that the yield of the product was only 0.2%.

The TLC analysis indicates that the new product was more non-polar compared to pyrogallol and methyl linoleate because of its higher Log P value. The HPLC showed the product's peak at 9.779 min with a yield of 0.2%, confirming that the product was a non-polar compound. As for the LC-MS/MS, the result shows the desired product molecular mass which was 418 g/mol.

The solubility test cannot be performed to evaluate the solubility limit of the desired product, however, the test result revealed that the reaction product had a higher solubility limit which was 2500 ppm in biodiesel, compared to pyrogallol's solubility limit at 2000 ppm. The antioxidant activity evaluation concluded that the reaction product fulfilled the ASTM D6751 (United States) standard, European EN 14214, and Indonesia National Standard SNI 7182 induction period requirements of 3 h, 6 h and 8 h. However, the reaction product had a lower acid value stability compared to pyrogallol and blank biodiesel.

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