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Research paper

Analysis on NO_X Formation of Biofuels

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Abstract

This paper proposes on analysis of NO_X formation of biofuels by using Corn Oil Methyl Ester (COME) and Palm Oil Methyl Ester (POME). The main contribution of this work is to study and analyse NO_X emission from biodiesel, which can improve biodiesel content and component in order to reduce NO_X formation coming from Compression Ignition (CI) engine. This is achieved by using alkaline base catalyst transesterification process to convert from palm and corn oil, containing low Free Fatty Acid (FFA), to biodiesel or biofuel. Biodiesel ratio, which is 400 ml of cooking oil: 100 ml of methanol: 2.8 g of potassium hydroxide (KOH), is used in transesterification process. The ratio is used to reduce alcohol consumption and cost. Almost 97% of biodiesel are yielded by using this ratio with direct heating from hot magnetic stirrer. The Fourier Transform Infrared Spectroscopy (FTIR) results determine the biodiesels illustrate the exhibition of C=O and C-O, which is the functional groups of esters, whereas the conventional diesel fuel does not have any of the functional groups. In gas emission testing, the biodiesel blends are burned in ceramic beaker including B20, B60 and B100. NO_X formation increases when the percentage for biodiesel blends are increase. In ThermaCAM P65 inspection, the testing shows that the temperature is directly proportional with the percentage of biodiesel blends. COME produced the highest amount of NO_X than POME and PBDF; POME is the suitable alternative biodiesel fuel that can be used for Compression Ignition engine beside PBDF. The analysis is useful for researchers who intend to reduce NO_X emission and improve air cleanliness by determining parameters and factors that can influence NO_X formation.

Keywords: *Biodiesel; Corn Oil; Diesel; Palm Oil; Nitric Oxide.*

1. Introduction

"Emissions" is a collective term that is used to describe the undesired gases and particles which are released into the air or emitted by various sources, its amount and the type change with a change in the industrial activity, technology, and a number of other factors, such as air pollution regulations and emissions controls [1]. The use of biodiesel in combustion engines greatly reduces the emissions compared to Petroleum Base Diesel Fuel (PBDF) by up to 100% sulphur dioxide (SO_2), 48% carbon monoxide (CO), 47% particulate matter (PM), 67% total unburned hydrocarbon (HC) and up to 90% reduction in mutagenicity [2].

Nitrogen oxides (NO_X) in the ambient air is divided into two gaseous, which are nitric oxide (NO) and nitrogen dioxide (NO_2). Significant pollutant of the lower atmosphere is caused by the two forms of nitrogen oxides. Nitrogen oxide is colourless and odourless. The presence of oxygen will convert from nitric oxide into nitrogen dioxide. Nitrogen dioxide is a reddish-brown gas with sharp and it is combined with water to produce nitric acid (HNO_3). Nitric acid will progressively destroy lung tissue [1].

NO_X emissions can be reduced with corrections in injection timing and combustion temperatures [3]. The potential increases in NO_X are always being addressed through research on engine thermodynamic mechanisms to improve emissions, catalytic conversion type technology to address the problem [4] or other else the additions of compounds such as antioxidants [5]. In term of diesel engines, almost all affords to reduce emissions at the source are to balance between the levels of nitrogen oxides and other compounds [6].

Many factors have an effect on NO_X formation from diesel engine such as pressure, reaction temperature, residence time of combustion products, premixed portion of combustion, availability of excess oxygen, ignition delay period, heat removal rate and the operational parameters of the engine [18]. An increase occurred in NO_X emissions for 3.5% increase in the fuel density was expressed by Signer et al. Furthermore, the number of double bonds, quantified as iodine number, correlated with the NO_X emissions were founded by Peterson et al. and McCormick et al. However, other researchers observed that there are no change or reduction NO_X emissions.

Furthermore, changes in the physical properties of biodiesel like the speed of sound and bulk modulus, can affect the fuel injection timing and this can increase NO_X. Several numbers of researches on the effects of NO_X formation have shown that in pump line nozzle (PLN) injection system, the increasing of fuel mass delivery and residence time owing to earlier injection timing will result very high reaction temperature and more NO_X formation. Vegetable oil and its methyl esters are less compressible than diesel; the pressure wave from the fuel pump is rapidly transferred to the injector nozzle which will cause the needle to open earlier [19].

A change of fuel properties results the increase of ignition delay period. An increasing of NO_X generation is caused by premixed burn fraction, the ignition delay become longer when premixed burn fraction is increase. Choi and Reitz [20] stated that when biodiesel used

as a diesel engine fuel, fuel mass delivery and spray tip penetration become increase. Benajes et al. [21] observed adiabatic flame temperature and stoichiometric can affluence the effects on NO_X formation in diesel engines. The increasing of fuel bound oxygen yields high adiabatic flame temperature of the fuels to complete the combustion. An increasing of NO_X is caused by the enrichment of oxygen intake and the use of oxygenated fuels [22]. Then, cetane number is created to measure the ignition delay between the injection timing and combustion timing. When the ignition delay become shorter, it will reduce combustion temperatures and decrease residence time.
This study aims on NO_X formation during combustion in diesel engine by using biodiesel production from palm oil and corn oil. Moreover, this study is also to evaluate the effect of oxygen content in biodiesel and NO_Xformation. Besides, the objective of the study is specifically to evaluate the relation of elevated temperature with NO_X formation in diesel engine.

2. Method & material

2.1. Preparation of biofuels

New palm and corn oils that are collected do not need to be filtered and heated as the oil does not have any contaminant and water content. The oil only undergoes two types of pre-treatment which are filtration and heating. The oil must be filtered to remove food residue, solid participate and large particle which can interrupt the transesterification process. After the filtration has been done, the oil is heated at 60˚C for 10 minutes to remove water content. Finally, the pre-treated palm oil is stored in a 500 ml conical flask sealed with aluminium foil.

Table 1: Vegetable feedstock (corn and palm) composition.

Feedstock composition per 14 g serving	Vegetable feedstocks	
	Corn oil	Palm oil
Monounsaturated fatty acid	4.0 g	6.3 g
Polyunsaturated fatty acid	7.5 g	1.7 g
Saturated fatty acid	2.2 g	6.0 g
Trans fatty acid	0.0 g	0.0 g

Transesterification process is performed by using either alkaline base catalyst or acid-catalyst transesterification. For this experiment, alkaline base catalyst transesterification is used as the oil has contained low Free Fatty Acid (FFA) shown in Table 1. According to Atadashi et al. [23], the amount of FFA in vegetable feedstock composition shall not exceed than 3wt% to make reaction working. Otherwise, acid-catalyst transesterification has to be implemented in order to reduce FFA level. Commonly, potassium hydroxide (KOH) is a catalyst that dissolves with methanol. Preventive action should be taken for KOH from absorbing atmosphere to avoid ineffectiveness in the transesterification. The quantities of methanol, potassium hydroxide and cooking oil are flexible as long as the ratio is followed which is 400 ml of oil: 100 ml of methanol: 2.8 g of potassium hydroxide [7].
Thereafter, the KOH and methanol are mixed by using magnetic stirrer at 60˚C for 15 minutes. This process is known as methoxide process. Then, the cooking oil is poured into conical flask with the methoxide. The methoxide will be reacted with palm and corn oils by using 1500 RPM of magnetic stirrer at which it stirs the mixture for 1, 2, 3 and 4 hours and maintain the temperature of mixture at 65˚C.
When the reaction is completed, a lump of glycerol is removed from methyl esters. The low solubility of glycerol in the alkyl esters will generally cause the separation occurred quickly and can be accomplished by settling [8]. Biodiesel and glycerine have different density, which biodiesel is less dense than glycerine and by using concept of gravity introduced by Stoke's law, the settling can be done. A separatory funnel is commonly used since it is easy to drain off the glycerine. This process only takes 24 hours at room temperature.
After the glycerol has been removed, the biodiesel is contained with residual catalyst, water, unreacted alcohol, free glycerol and soaps, which are obtained during transesterification process. Glycerol is insoluble in biodiesel oil. A free glycerol is a problem of biodiesel after settling because it will remain either as suspended droplets or the very small amount that does not dissolve in biodiesel. The most glycerol in biodiesel is treated during washing process. A free glycerol is quite low in water-wash fuel especially by using warm water to wash biodiesel [8].
The washing process is carried out by taking 20 ml of distilled water and mixing with the biodiesel. Then, the mixture is stirred gently about 10 minutes. Two layers of the biodiesel can be observed in separatory funnel at which the lighter biodiesel is on the top; hydrated methanol is at the bottom of separatory funnel. The washing process can be repeated approximately in 5 times to separate the biodiesel from the glycerol. Confirmation test can be done by using pH paper where the biodiesel must be neutral to ensure no more catalyst and methanol left in the biodiesel.
After the biodiesel is washed, it must be dried until it is crystal clear. Reacted, washed and dried biodiesel may be used in some diesel engine. It should not have a methanol left in it and it should have pH close to 7.
Typically, biodiesel is generally used a blend of pure biodiesel and diesel. The biodiesel which are produced is pure biodiesel and known as B100. The blending percentage of biodiesel with diesel is set up to 20%, 60% and 100% which are known as B20, B600, and B100. However, in order to prepare 100 ml of B20, 20 ml of pure biodiesel is mixed with 80 ml of diesel. Finally, all the biodiesel blends are stored in different conical flask and sealed with aluminium foil

3. Experimental tests

3.1. Fourier transforms infrared spectroscopy (FTIR)

Fourier Transform Infrared Spectroscopy (FTIR) is a device to determine an infrared spectrum of absorption, mission, photoconductivity of a solid, liquid or gas where the information is collected and converted from interference pattern into spectrum. The objective of FTIR invented is to identify unknown materials, obtain the quality or consistency of the sample and determine the quantity of component in mixture.
FTIR has five components. The source of infrared emits a different wavelength of infrared radiation. An optical inverse Fourier transform on the entering infrared radiation is performed by using the interferometer. Gas sample is penetrated by the modulated infrared beam where various molecules present absorbed to various extents at different wavelengths. Thereafter, the intensity of infrared beam is

detected by the detector and the signal is converted into data by using Fourier Transform from computer to get an infrared spectrum of the sample gas.

3.2. Gas analysis of fuel at various biodiesel/diesel blends by using flue gas analyser

After fuels are burned, a certain number of gas components such as CO_2, OH, NO_X, CO and PM will emerge. If the gas' components are still containing combustion heat, they are known as heating gases. After they conveyed their energy to absorbing surfaces of a heat exchanger, they are called flue. The device, which is called as Flue Gas Analyzer, is used to determine the content of flue in biodiesel after combustion. Flue gas analyser is used to determine the flue of the gases in biodiesel. Flue gas analyser is carried out by using a device is known as Testo 350-XL. A structure of the flue will be obtained by using flue from heating oil in ceramic beaker based on biodiesel blends from B20, B60, B100 and conventional diesel. After Flue is generated, it will be placed in a funnel of the Flue Gas Analyzer and the data will be displayed on the screen.

3.3. Flame temperature test by using ThermaCAM P65

Thermography unit as known as the ThermaCAM P65 infrared condition monitoring system has been built with several components such as infrared camera with built-in 36mm lens, a visual color camera, a laser pointer, an IrDA (infrared communications link), a 4" color LCD on a removable remote control, and a range of accessories. This camera is measured and images the emitted infrared radiation from combustion of the fuels in ceramic beaker. Actually, the fact states that radiation is a key function to detect flame temperature from combustion fuels, which makes it possible for the camera to calculate and show a temperature.

4. Results and discussions

4.1. FTIR results

The purpose of this testing is to analyse the functional group in Corn Oil Methyl Ester (COME), Palm Oil Methyl Ester (POME) and Petroleum Base Diesel Fuel (PBDF) based on the infrared spectrum obtained. The result of FTIR was compared and analysed to determine the functional group respectively. Based on the Fig. 1, FTIR signal was captured to compare the infrared spectrum between biodiesels and PBDF.

According to Fig. 1, the light was absorbed moderately from 4000 cm^{-1} to 3500 cm^{-}1 and the range, referring to the theory, possibly shows the present of O-H functional group. O-H functional group is the component of glycerol. The range from 3400 cm^{-1} until 3100 cm^{-1} informed that the functional group of unsaturated C=C-H was existed and the highest peak among the others.

On the other hand, the range from 2900 cm^{-1} until 2400 cm^{-1} was flat but the functional group still absorbed the light. This range had been observed that the functional group for each type of fuels has some difference. For COME, the functional group of saturated C-H did not emerge compared with POME and PBDF. Biodiesel and biodiesel blends produce significant reduction in HC between 10% and 25% in heavy-duty engines and vehicles [10].

At the range of 2400 cm^{-1} to 2200 cm^{-1}, some absorption of the light had occurred moderately and it showed the functional group of C≡C was exhibited clearly. The differences among three types of fuels were the intensity of light where functional group of C≡C had absorbed and reflected it. PBDF had absorbed more light compared with COME and POME. Furthermore, Fig. 1 had shown PBDF contained more C≡C than biodiesels. The functional group of C≡C as known as acetylene gives fuels to be burned easily. Typically, acetylene has a very wide flammability range; acetylene has higher flame speed and also faster energy release. Larger compression ratios are allowable because of high self-ignition temperature of acetylene compare with diesel engines do [11].

At the range of 1700 cm-1 to 1750 cm-1, The presence of C=O functional group was clearly significant. COME and POME have C=O content lower than PBDF. It means that COME and POME maybe contained more oxygen and promote cleaner and complete combustion. The range of 1500 cm-1 to 700 cm-1 is known as fingerprint regional. It indicated that the functional group of C-O possibly existed in PBDF but lesser than POME and COME.

PBDF was certainly contained oxygen composition but very small composition compared with biodiesel. Biodiesel is containing 11% of oxygen by weight and high content of oxygen in biodiesel caused increasing emission of NO_X formation due to the high combustion efficiency and higher reaction temperature [12].

4.2. Gas emission of fuel at various biodiesel/diesel results

Gas emission was produced when the fuels had been burned in ceramic beaker. Gas emission testing was used for this research to determine the exact amount of NO_X in diesel and biodiesel fuels. Furthermore, the intention of this testing is to determine a correlation between oxygen emission in fuels and NO_X formation. A small part of the paper was consumed to ignite the biodiesels in ceramic beaker as biodiesel is hard to be burned without any starter due to higher viscosity.

Fig. 2 showed the relation between COME, POME and PBDF in term of O_2 and NO_X. Based on the graph and the data given, the correlation can be seen between O_2 and the components of NO_X for each fuel. The recent research states NO_X can be increased with the enrichment of oxygen intake and the usage of oxygenated fuels [13]. The adiabatic flame temperature and NO_X emissions decreased linearly with the oxygen content in biofuels [14].

According to the Fig. 2, the graph is clearly stating the comparison between COME, POME and PBDF in O_2 and NO_2 term where the oxygen content in POME is higher than COME. As the oxygen content of POME and COME are higher than PBDF, a lot of NO_2 gas amount is produced and exposed to the environment. the difference could be seen on Fig. 2. The oxygen content of COME and POME fuels is 20.79% and 21%. NO_2 content produced from COME fuel combustion 0.4 ppm higher than POME. In term of normalization, which can be referred to Fig. 3; the COME and POME are increased by 1% and 1.9% of NO_2, which the oxygen emission for both biofuels are higher than the PBDF. From this result, the higher oxygen emission in fuels will increase NO_X formation in combustion.

A comparison of gas emission between blends including B20, B60 and B100 among COME, POME and PBDF was made and recorded. Referring to the recent research, the use of PBDF as a blend into biofuels form resulted in substantial increases in NO_X formation. By the way, the use of B100 can cause a measurable increase in NO_X emissions compared with PBDF [10].

Based on Fig. 4, the O_2 emissions gradually increased with B20, B60 and B100 of COME. Same goes with NO_2 formation, which it was increase in ratio from 1 to 3 times. From the observation, it was clearly shown that the NO_X formation was increase when the blends of biofuels were also increase. One of the researcher states biodiesel blends and biodiesel for B20 produce significant increase in NO_X around 2 until 3% [15]. However, the uses of B20 can lead NO_X emissions to increase cannot widely accepted [10]. Fig. 4 concluded that the percentage of oxygen emissions in fuels was proportional to NO_X formation. The NO_X formation in the POME from B20 to B100 can be seen in Fig. 3. For B20 and B60 of the POME, the NO_2 formation ratio was the same. However, it was increase when the blends were increased till B100.

Fig. 4 concluded NO_X formation can be increased when the blends increased. Benchmarking with COME and PBDF, POME yielded NO_X formation lesser than COME but higher than PBDF from B20 until B100. It means that oxygen emissions are the important factor that can change the amount of NOx formation.

4.3. Flame Temperature Results

This experiment was conducted by burning the fuels in ceramic beaker and the ThermaCAM P65 captured the radiation from the flame of the burning fuels. The NO_X was formed by the reaction of an atmospheric nitrogen with an oxygen due to the combustion at elevated temperature around 537˚C (1800˚K) [16]. Basically, an increasing of temperature may be influenced by oxygen content, which is helping the combustion to be completed, in biodiesel. Biodiesel was reported that the adiabatic flame temperature for biodiesel is slightly higher than PBDF due to complete combustion resulting from fuel bound oxygen [17].

According to Fig. 5, the 20% of biodiesel in the POME yielded was the lowest temperature which the T_{upper} value was 171˚C

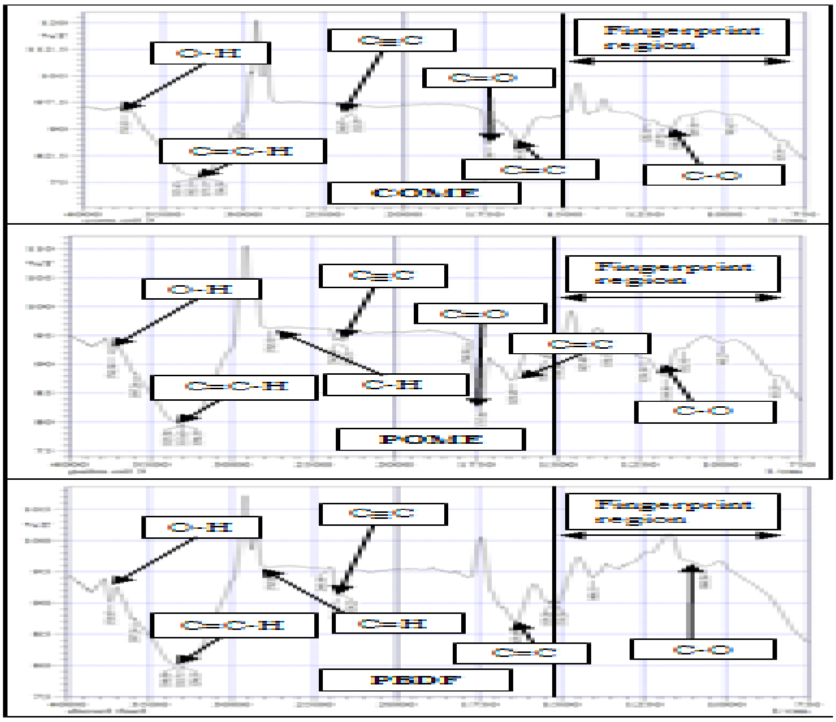

Fig. 1: The FTIR spectrum of COME, POME and PBDF.

Lesser than 172˚C for B60 and 177˚C for B100. The 100% of biodiesel had yielded the highest temperature among the blends.

Based on recent researches and previous results, the fuel bound oxygen may be the main factor of NO_X formation and high temperature. Next, Fig. 5 showed that the 20% of biodiesel in the COME had yielded the lowest temperature, which the T_{upper} value is 170 ˚C lesser than 176 ˚C for B60 and 185 ˚C for B100. The 100% of biodiesel had yielded the highest temperature among the blends. However, the result demonstrated the COME yielded the highest temperature from B20 until B100 compared with the POME and PBDF. Previous results had shown that the oxygen content in the POME should be higher than the COME and PBDF. Thus, it should be the POME to yield higher temperature than the COME and produces a large amount of NO_X. PBDF yielded the lowest T_{upper} value compared than B100 of the POME and COME. Therefore, the PBDF should yield less NO_X because of the lowest temperature compared than the COME and POME. Fuels with higher oxygen content might yield high temperature and NO_X.

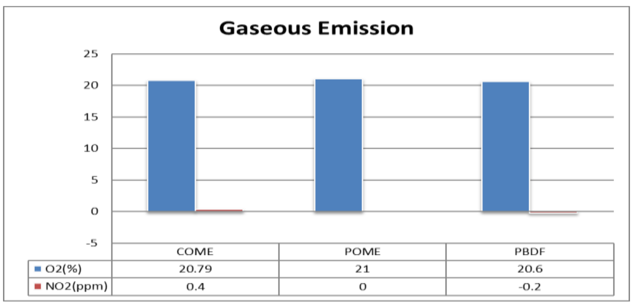

Fig. 2: The graph for COME, POME and PBDF in term of O_2 and NO_2.

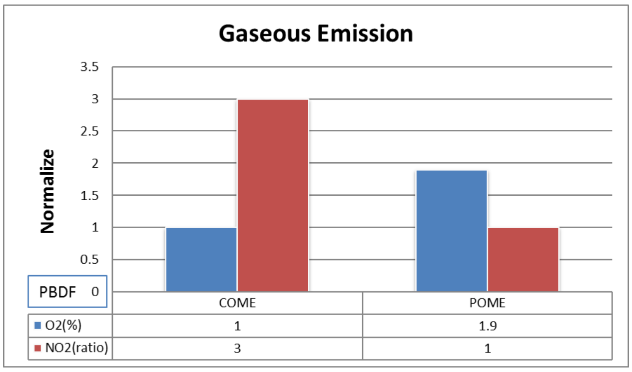

Fig. 3: The graph for COME, POME and PBDF after normalisation.

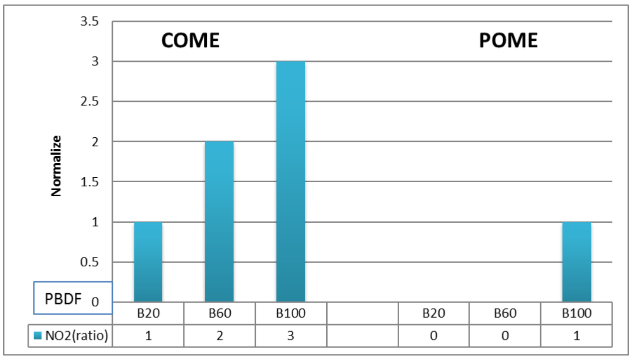

Fig. 4: The graph for COME and POME compare with PBDF in term of NO_2.

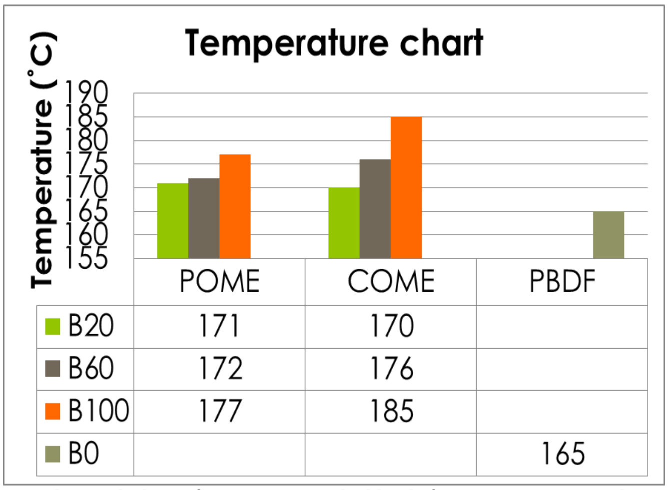

Fig. 5: The graph shows for T_{upper} among the blends for COME, POME and PBDF**.**

5. Conclusions

Palm Oil Methyl Ester (POME) and Corn Oil Methyl Ester (COME) have been successfully made from palm and corn cooking oil by using transesterification process. However, the results are achieved in satisfactory state as the objectives of the project have been fulfilled, including:

1. The FTIR results have shown the presence of C=O and C-O indicate the functional group of esters. It means that COME and POME may be contained higher oxygen composition compared with PBDF. Higher content of oxygen in biodiesel causes increasing emission of NO_X formation due to the high combustion efficiency and higher reaction temperature.
2. For gas emissions test, it was found that when the percentage of biodiesels increased, more NO_X formation was produced as the flame temperature increased due to 20% the oxygen content increased in the biodiesels. The B60 of each biodiesel produced temperature lesser than B100 but higher than the B20 and PBDF. The percentage of the biodiesels may affect the temperature since an increasing of the oxygen content in the biodiesels can lead to higher combustion.
3. Based on observation, COME produced the highest amount of NO_X than POME and PBDF; POME is the suitable alternative biodiesel fuel that can be used for Compression Ignition engine beside PBDF.

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