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The effect of biodiesel oxidation on engine performance and emissions

Abdul Monyem, Jon H. Van Gerpen *

Department of Mechanical Engineering, College of Engineering, Iowa State University, 2025 H.M. Black Engng. Bldg., Ames, IA 50011-2161, USA

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Abstract

Biodiesel is an alternative fuel consisting of the alkyl monoesters of fatty acids from vegetable oils or animal fats. Previous research has shown that biodiesel-fueled engines produce less carbon monoxide, unburned hydrocarbons, and particulate emissions compared to diesel fuel. One drawback of biodiesel is that it is more prone to oxidation than petroleum-based diesel fuel. In its advanced stages, this oxidation can cause the fuel to become acidic and to form insoluble gums and sediments that can plug fuel filters. The objective of this study was to evaluate the impact of oxidized biodiesel on engine performance and emissions. A John Deere 4276T turbocharged DI diesel engine was fueled with oxidized and unoxidized biodiesel and the performance and emissions were compared with No. 2 diesel fuel. The neat biodiesels, 20% blends, and the base fuel (No. 2 diesel) were tested at two different loads (100 and 20%) and three injection timings (3° advanced, standard; 3° retarded). The tests were performed at steady-state conditions at a single engine speed of 1400 rpm. The engine performance of the neat biodiesels and their blends was similar to that of No. 2 diesel fuel with the same thermal efficiency, but higher fuel consumption. Compared with unoxidized biodiesel, oxidized neat biodiesel produced 15 and 16% lower exhaust carbon monoxide and hydrocarbons, respectively. No statistically significant difference was found between the oxides of nitrogen and smoke emissions from oxidized and unoxidized biodiesel. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Biodiesel is an alternative diesel fuel consisting of the alkyl monoesters of fatty acids derived from vegetable oils or animal fats. It has been the focus of a considerable amount of recent research because it is renewable and reduces the emission of some pollutants. The most common form of biodiesel in the United States is made with methanol and soybean oil

and is known as soy methyl ester, or *methyl soyate*. In Europe, rapeseed oil-based esters have been widely used as an alternative diesel fuel.

A number of researchers have investigated vegetable oil-based fuels [1–4]. Most have concluded that vegetable oils can be safely burned for short periods of time in a diesel engine. However, using raw vegetable oil in a diesel engine for extended periods of time may result in severe engine deposits, piston ring sticking, injector coking, and thickening of the lubricating oil [1,2,5–9]. The high viscosity of raw oil reduces fuel atomization and increases fuel spray

* Tel.: +1-515-294-5563; fax: +1-515-294-3261.

E-mail address: jvg@iastate.edu (J.H. Van Gerpen).

penetration. Higher spray penetration is thought to be partly responsible for the difficulties experienced with engine deposits and thickening of the lubricating oil [1,3,10]. However, these effects can be reduced or eliminated through transesterification of the oil to form monoesters [11,12]. The process of transesterification removes glycerol from the triglycerides and replaces it with radicals from the alcohol used for the conversion process [13]. This process decreases the viscosity but maintains the cetane number and the heating value. These monoesters have come to be known as *biodiesel*.

Several researchers have observed that the exhaust emissions are affected by the use of biodiesel. Chang et al. [14], Geyer et al. [15], Graboski et al. [16], and Alfuso et al. [17] found that biodiesel generally caused an increase in oxides of nitrogen (NO_x) emissions, and a decrease in HC, CO, and particulate emissions relative to No. 2 diesel fuel. The magnitudes of the emission changes appear to be engine dependent.

Biodiesel from soybean oil is highly unsaturated and this makes it very prone to oxidation. The methylene groups adjacent to the double bonds are particularly susceptible to free radical attack. Multiple double bonds on the same fatty acid chain are more susceptible to oxidation than would be indicated by the number of double bonds alone. Linoleic and linolenic acids, with two and three bonds, respectively, oxidize readily. Both are found at relatively high levels in soybean oil. The autooxidation reaction produces hydroperoxides which can polymerize with other radicals to produce high molecular weight insoluble sediments and gums. In some cases, the oxidized fatty acid chains may break apart producing shorter chain acids and aldehydes.

The most likely impact of the sediment and gum formation will be fuel filter plugging and varnish deposits on fuel system components and these phenomena have been observed [18–20]. The acid formation may cause fuel system corrosion. In addition, the hydroperoxides are very unstable and have a tendency to attack elastomers.

The impact of biodiesel oxidation on the engine's performance and emissions is not currently understood. A recent study [21] showed that the cetane number of biodiesel increased as the biodiesel oxidized, up to a peroxide value of 80. Higher cetane number means that the fuel autoignites more easily in the

engine cylinder. However, no research has yet been conducted to determine the impact of oxidized vegetable oil esters on engine performance and emissions.

The objective of this study was to investigate whether the fuel chemistry changes caused by oxidation produce significant changes in the engine performance and emissions. If these changes are significant and deleterious, they could cause engines using oxidized fuel to no longer meet manufacturer performance goals or government regulations for emission certification.

2. Equipment and procedures

The engine used for the experiments was a John Deere 4276T four-cylinder, four-stroke, turbocharged DI diesel engine with a bore of 106.5 mm, a stroke of 127.0 mm, a displacement of 4.53 l, and a compression ratio of 16.8:1. The engine was connected to a 150 HP General Electric model TLC2544 DC electric dynamometer. The engine was operated at two load levels corresponding to full load (257.6 N m) and 20% of full load (51.5 N m) at a constant speed of 1400 rpm. A Lebow load cell was used to measure brake torque. An electronic scale and a stopwatch were used to measure the fuel flow rate.

A Beckman model 402 heated flame ionization detector hydrocarbon analyzer was used to measure the unburned HC emissions. Two Beckman model 864 infrared analyzers measured the concentrations of CO and carbon dioxide (CO_2) in the engine exhaust. A Beckman model 7003 polarigraphic oxygen monitor was used to measure the oxygen in the exhaust gas and a Bosch smoke meter was used to measure the smoke level. A Beckman Model 955 chemiluminescent analyzer was used to measure the concentrations of NO and NO_x .

The emissions data were taken for each fuel at 1-s intervals for two separate 5 min intervals. The first set of data was taken with the NO_x/NO meter set for NO_x emissions and the second set of data was taken for NO emissions. It is general practice to express the emissions data on a "brake specific" basis except for the Bosch smoke number. Brake specific emissions are the mass flow rate of the pollutant divided by the engine power. Humidity correction of the NO_x emissions followed the procedure recommended by

the Society of Automotive Engineers [22]. The emissions data were collected using the Labview program with a 486 computer and a National Instruments Model ATMIO-16 data acquisition system.

2.1. Fuel preparation

The biodiesel was soybean oil-based and purchased from a commercial supplier (NOPEC Corp., Lakeland, Florida). The No. 2 diesel fuel was also a commercial grade and was purchased from a local fuel supplier. Chemical analyses of the test fuels are shown in Tables 2 and 3. The 20% blends were prepared by weight using the same biodiesel and No. 2 diesel fuels tested in the neat form.

The oxidized biodiesel was prepared by heating 83 l of biodiesel to 60°C in a 208 l drum and bubbling oxygen (99.6% purity) at the rate of 0.4 m³/h through the fuel. The fuel's peroxide value, as measured by the American Oil Chemists' Society method Cd 8-53 [23], was used as the indicator of the extent of oxidation. Elevated temperature and oxygen were used to raise the fuel peroxide value quickly without allowing the fuel viscosity to increase by more than one centistoke. Samples were collected each hour and their peroxide value measured. Sampling was continued until the peroxide value of the fuel reached 340 meq/kg ester which typically required about 10 h. At this point, the oxygen flow was stopped and the fuel was allowed to cool down to room temperature. The barrel was then sealed until it was used. A total of three 83 l batches of biodiesel were oxidized to prepare the fuel. Each batch of biodiesel was sufficient for one 3-day test sequence.

2.2. Statistical analysis

The primary purpose of this project was to characterize the effect on the performance and exhaust emissions of the engine when fueled with oxidized biodiesel compared with unoxidized biodiesel and petroleum-based diesel fuel. In addition to considering these three fuels, the effect of blending 20% of the two biodiesels in diesel fuel was included to give 5 test fuels. Then, the effects of two engine load levels and three injection timing settings were included, giving a total of 3 test variables. Upon further consideration, it was determined that it was not feasible to

produce a single batch of oxidized biodiesel because it may be unstable and continue to oxidize during the time interval between production and use. Therefore, it was decided to produce three separate batches of fuel and to control the time between production and use. This provided two additional test variables, the batch number, which would be used to characterize batch-to-batch variability, and the age of the batch, which would account for on-going oxidation. Later analysis showed that these variables had no significant impact on the test results.

Several different statistical designs were considered for this project. A factorial experiment in which the response of the dependent variables (emissions, fuel consumption, etc.) is observed at all factor-level combinations of the independent variables was considered first. However, a true factorial experiment could not be run because it was not possible to run all of the experiments on the same day. The experiments had to be split into separate days which introduces the uncertainty of day-to-day variability. Also, due to the time required to accurately set the fuel injection timing, only one value could be set per day. In factorial design, a variable like the injection timing needs to change several times during a single day test which was not feasible. Instead of the factorial design, a split plot design was developed for this project.

The split plot design consists of two stages. The first stage is related to the whole plot and the second stage is related to a subplot. This design is thus named a split plot design with a day as a "whole plot" and each of the 10 runs within a day as a "sub-plot". This split plot design is shown schematically in Table 1. The whole plot is a 3 × 3 Latin square and within each whole day plot is a 2 × 5 factorial experiment. The 3 × 3 Latin square design contains 3 rows and 3 columns. The three treatments (injection timings) are randomly assigned to experimental units within the rows and columns so that each treatment appears in every row and in every column. This provides 3 replications of each injection timing. The 2 × 5 factorial experiment identifies the 10 randomly chosen combinations of load and fuel type run each day. More extensive explanation of these topics is provided in Ott [24] and Neter et al. [25].

A SAS program was used to analyze the collected data. The program output was then tabulated using analysis of variance (ANOVA) tables. Another means

Table 1
Split plot design

(a) Whole plot (3×3 Latin square)					
Age →	1	2	3		
Batch					
1	Standard (day 1)	3° advanced (day 2)	3° retarded (day 3)		
2	3° retarded (day 4)	Standard (day 5)	3° advanced (day 6)		
3	3° advanced (day 7)	3° retarded (day 8)	Standard (day 9)		
(b) Randomly assigned subplot within each whole plot (2×5 factorial experiment) ^a					
Fuel →	100% HPVB	100% LPVB	20% HPVB	20% LPVB	No. 2D
Load					
100% load	10th	5th	9th	3rd	1st
20% load	4th	8th	7th	6th	2nd

^aHPVB = high peroxide value biodiesel; LPVB = low peroxide value biodiesel.

to determine statistical significance called a Tukey's grouping table, was also computed. The tables for each of the independent variables are included in Ref. [26].

The statistical analysis identified not only the effect of fuel oxidation on performance and emissions, but also the effect of fuel batch, age of batch, fuel type, load, timing, the interactions between load and fuel, load and timing, timing and fuel type, and the interactions between timing, load, and fuel type. The analysis showed that fuel batch and age of batch did not have a statistically significant effect on any of the measured variables reported in this study. A 95% confidence interval was used throughout the study.

3. Results and discussion

3.1. Engine performance

Fig. 1 shows the brake specific fuel consumption (BSFC) for the five fuels at the full load engine condition. All points shown in this figure were the average of three data points and the error bars show the spread between the maximum and the minimum of the three data points. In this case, the largest error bar was only $\pm 0.6\%$.

The oxidized and unoxidized neat biodiesels had 15.1 and 13.8% higher BSFCs than diesel fuel, respectively. Since the energy per unit mass of biodiesel was lower than No. 2 diesel fuel (see Table 2), the fuel consumption had to be higher to maintain the same brake power. At each of the two load conditions, the torque and the RPM were kept constant, so the brake power was also constant. The heating value of

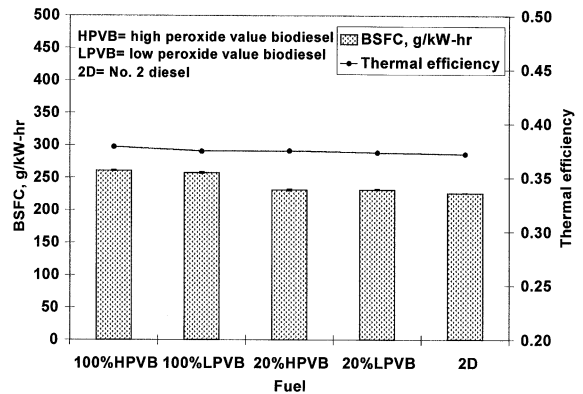


Fig. 1. Brake-specific fuel consumption and thermal efficiency for standard timing at full load.

Table 2
Biodiesel analysis

Fatty acid composition	
Palmitic (16:0)	10.76%
Stearic (18:0)	4.37%
Oleic (18:1)	24.13%
Linoleic (18:2)	51.83%
Linolenic (18:3)	6.81%
Free glycerin (%)	0.004
Monoglycerides (%)	0.352
Diglycerides (%)	0.132
Triglycerides (%)	0.152
Total glycerin (%)	0.131
Viscosity (cSt)	4.63

oxidized and unoxidized biodiesel were shown in Table 3 to be 14.7 and 12.7% lower, respectively,

Table 3
Fuel analysis

Fuel	No. 2 diesel	Biodiesel	Oxidized biodiesel
<i>Molecular wt.</i>	198	291.6	—
% Carbon	86.23%	76.14%	76.06%
% Hydrogen	13.14	11.75	11.51
% Sulfur	0.034	< 0.005	0.010
Cetane number (D613)	47.4	51.1	72.7
<i>Heat of combustion</i>			
Gross (kJ/kg)	45504	39766	38896
Net (kJ/kg)	42716	37273	36454
<i>Hydrocarbon types</i>			
Saturates (%)	64.1	—	—
Olefins (%)	4.9	—	—
Aromatics (%)	31.0	0	0

than that of No. 2 diesel fuel. Another reason for the increase in BSFC with biodiesel may be a change in the combustion timing caused by the biodiesel's higher cetane number as well as injection timing changes to be discussed later. These results are similar to those of MacDonald et al. [27] who fueled a Caterpillar 3304 PCNA engine with low-sulfur diesel fuel and methyl esters of soybean oil. In their research they found a 13 to 14% increase in BSFC.

There was a 1.2% higher BSFC measured for the oxidised biodiesel than the unoxidized biodiesel and this was statistically significant. The heating value of oxidized biodiesel was about 2.2% less than that of unoxidized biodiesel. Thompson et al. [28] also found that the heat of combustion decreased as the peroxide value of the biodiesel increased. They found that the heating value decreased about 1.4% after 24 months of storage.

Fig. 1 shows the thermal efficiency of the engine at full load. For the purposes of this calculation, the measured lower heating values given in Table 3 were used. Fig. 1 also shows that the thermal efficiency of the biodiesel and its blends is the same as for No. 2 diesel fuel. This indicates that the engine's ability to convert chemical energy to mechanical energy is the same for all the fuels. The thermal efficiency for all five fuel blends was about 37% at the full load engine condition and was 21% at the light load engine condition (not shown). Similar results were found by Schumacher et al. [29] and Chang et al. [30].

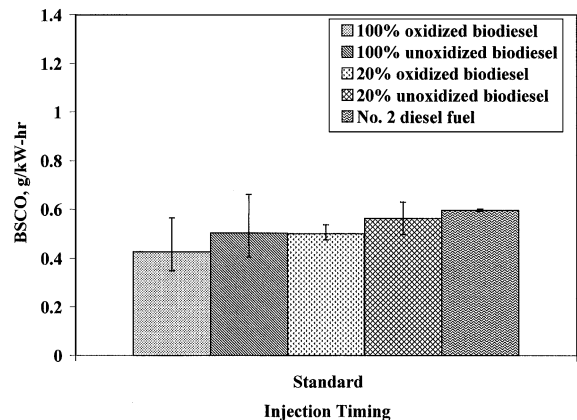


Fig. 2. Brake-specific CO emissions at full load.

3.2. The effect of fuel oxidation on exhaust emissions

3.2.1. Carbon monoxide (CO) emissions

The changes in CO emissions that resulted from the changes in injection timing, fuel type, and load were found to be statistically significant. Only the brake-specific CO emissions for full load at standard injection timing are shown in Fig. 2 but the trends were similar at the other timings. The highest CO emissions for all of the injection timings were found for the No. 2 diesel fuel, while the oxidized biodiesel fuel had the lowest. The oxidized biodiesel had about 15% less CO emissions than the unoxidized biodiesel and 28% less than No. 2 diesel fuel, both of which were statistically significant.

The 20% load CO emissions are shown in Fig. 3. The brake-specific CO emissions were much higher for the light-load condition than at full load. The CO emissions for the oxidized biodiesel were 26% lower than for the unoxidized fuel and 55% lower than the No. 2 diesel fuel.

3.2.2. Unburned hydrocarbon (HC) emissions

The full load HC emissions for all five fuel blends are shown in Fig. 4. The HC emissions for all the biodiesel fuels were less than for the No. 2 diesel fuel. The changes in HC emissions that resulted from the changes in fuel and load were statistically significant. This HC emissions reduction was also found for blends of vegetable oil esters and diesel fuel by

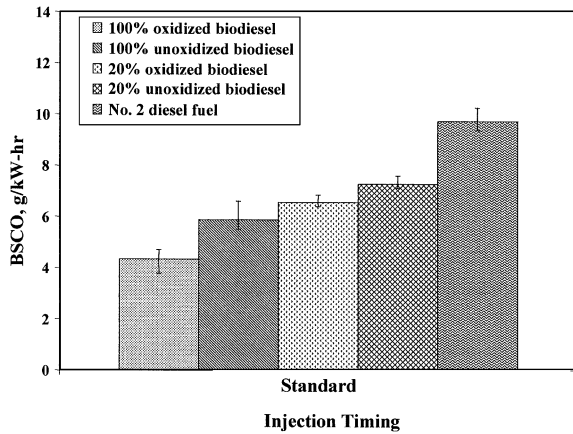


Fig. 3. Brake-specific CO emissions at light load.

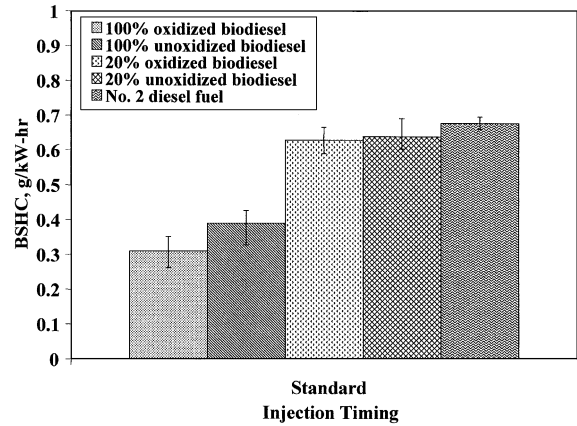


Fig. 5. Brake-specific HC emissions at light load.

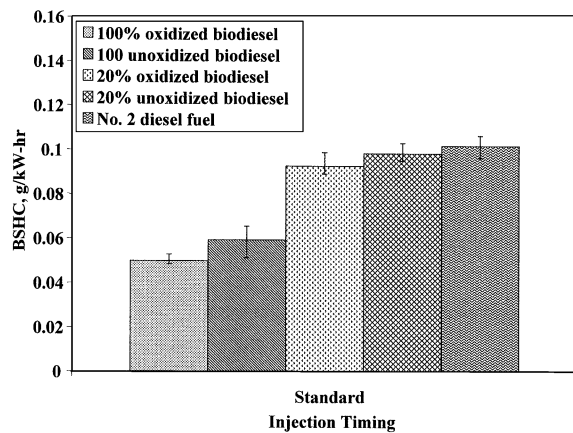


Fig. 4. Brake-specific HC emissions at full-load engine condition.

Chang et al. [30] and Rickeard et al. [31]. In the same manner as the CO emissions, the oxidized biodiesel was found to significantly reduce HC emissions compared with unoxidized biodiesel.

The brake-specific HC emissions for the light-load engine condition are shown in Fig. 5. The HC emissions were higher at the light-load engine condition than at the full-load engine condition by approximately a factor of 10. At the light-load engine condition, the oxidized biodiesel reduced the HC emissions by 21% compared to the unoxidized biodiesel. Compared to the No. 2 diesel fuel, the highest HC emissions improvement was 54% and was found for the oxidized biodiesel. Although not shown in the figure, the HC

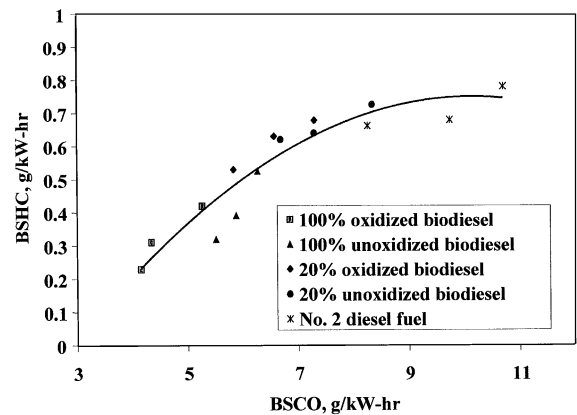


Fig. 6. BSHC vs. BSCO emissions curve at light load.

emissions increased as the injection timing advanced and this was true for all five tested fuel blends.

Fig. 6 shows the relation between the light-load HC and CO emissions for all the fuels and injection timings. The CO emissions consistently increase as the HC emissions increase. Even under the fuel-lean conditions of a diesel engine, incomplete HC oxidation can result in an increase in CO levels. The HC in the overmixed regions oxidizes very slowly and can produce more products of incomplete combustion, such as CO.

The origin of biodiesel's lower HC and CO emissions is not clear. The reductions might be due to changes in the stoichiometry of the spray caused by the oxygen in the biodiesel fuel. Part of the HC reduction could also be due to loss of unburned biodiesel

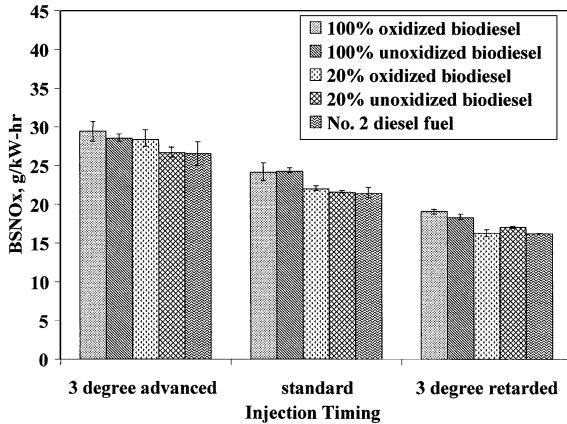
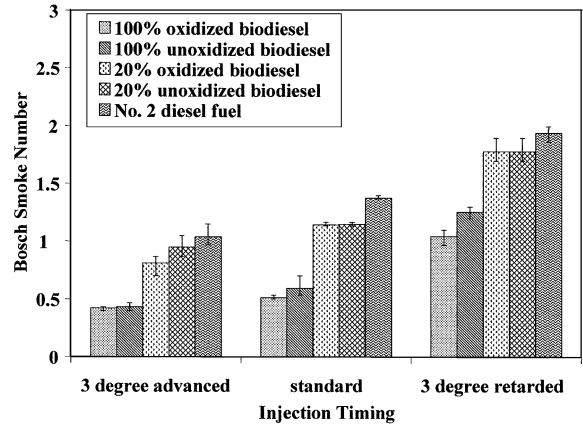
Fig. 7. Brake-specific NO_x emissions at full load.

Fig. 8. Bosch smoke number at full load.

in the emissions sampling system because of its low volatility. The shorter ignition delay associated with biodiesel's higher cetane number could also reduce the amount of overmixed fuel which is the primary source of unburned HC at light load.

3.2.3. Oxides of nitrogen (NO_x) emissions

Nitric oxide (NO) and nitrogen dioxide (NO₂) are usually combined together as NO_x although NO is the dominant oxide of nitrogen produced inside the engine cylinder. The brake-specific NO_x emissions are shown in Fig. 7 for the full-load engine condition, at 3 different injection timings. The NO_x emissions for biodiesel were higher than for the No. 2 diesel fuel. The differences between the NO_x emissions of the oxidized biodiesel and the unoxidized biodiesel were not statistically significant. The NO_x emissions of the 20% blends were not significantly different from the No. 2 diesel fuel.

The neat oxidized biodiesel at the standard injection timing had about 13% higher NO_x emissions than the No. 2 diesel fuel while the unoxidized biodiesel had about 14% higher. Compared to No. 2 diesel fuel at the same timing, the neat oxidized biodiesel at 3° advanced and 3° retarded injection timings increased NO_x emissions by 11 and 18%, respectively. Unoxidized biodiesel at the 3° advanced and 3° retarded injection timings increased NO_x emissions by 7.5 and 12.9%, respectively. The neat biodiesels consistently produced higher full-load NO_x emissions than the No. 2 diesel fuel at all three injection timings.

Mittelbach and Tritthart [32] tested methyl esters of used frying oil and found lower CO and HC emissions and smoke level but increased NO_x emissions compared to No. 2 diesel fuel. Rickeard et al. [31] also mentioned that the NO_x emissions increased for the bio-fuels they tested.

The reason for the increase in NO_x is not clear. The cetane numbers of the biodiesels shown in Table 3 are higher than for the No. 2 diesel fuel and this is usually associated with lower NO_x emissions. However, data presented in [26] and to be published later has shown that the different physical properties of biodiesel, including its sonic velocity and bulk modulus, cause an advance in the injection timing of biodiesel-fueled engines. Rotary injection pumps like that on the engine used in the study can also cause the injection timing to change if the quantity of fuel delivered changes. A greater quantity of biodiesel was injected in these tests in order to deliver the same engine torque. The timing advance associated with these effects could be at least partially responsible for the increase in NO_x emissions.

3.2.4. Smoke number (SN)

The Bosch smoke numbers for all five fuels are shown in Fig. 8 for the three injection timings. The No. 2 diesel fuel had the highest smoke number at all injection timings, while the oxidized biodiesel fuel had the lowest. All fuel blends showed an increase in smoke number as the injection timing was retarded. Compared with the No. 2 diesel fuel, the unoxidized

biodiesel reduced the smoke number from 1.4 to 0.6 at standard injection timing. The oxidized biodiesel had an even greater reduction in smoke number. Even though the average smoke number for the highly oxidized biodiesel was 14% lower than for the unoxidized biodiesel at standard timing and full load, the difference was not found to be statistically significant. Similarly, the smoke numbers for the 20% blends of biodiesel were not significantly different from each other. The smoke number for the No. 2 diesel fuel had a statistically significant difference compared with the other four fuels. All three groups of fuels, the neat biodiesels, the 20% blends, and the No. 2 diesel fuel were significantly different from each other. The minimum significant difference in the smoke number was 0.05 for the fuel blends.

4. Conclusions

The objective of this project was to characterize the effect of oxidation on the exhaust emissions from a biodiesel-fueled engine. Based on the experimental results, the following conclusions can be drawn.

1. The engine performance of the oxidized and unoxidized biodiesels and their blends was similar to that of No. 2 diesel fuel with nearly the same thermal efficiency, but with higher fuel consumption reflecting their lower energy content.
2. The neat biodiesels and the biodiesel blends produced lower CO emissions. The oxidized biodiesel reduced the CO emissions over 28% compared with diesel fuel and 15% compared to the unoxidized biodiesel at the full-load engine condition.
3. All neat biodiesels and biodiesel blends produced lower emissions of unburned hydrocarbon with a maximum reduction of 51% for the neat oxidized biodiesel compared with No. 2 diesel fuel. The oxidized biodiesel produced 16% lower HC emissions than the unoxidized biodiesel.
4. The neat biodiesels produced NO_x emissions that were 13–14% higher than the No. 2 diesel fuel. However, the difference in NO_x emissions between the No. 2 diesel fuel and the 20% blends was not significant. The effect of fuel oxidation on the NO_x emissions was also found to be insignificant.
5. The Bosch smoke number was significantly reduced when the diesel engine was fueled with neat biodiesel and its blends with No. 2 diesel fuel. The highest reduction was found for the oxidized biodiesel. It reduced the smoke number from 1.4 to 0.5 compared with diesel fuel. The effect of oxidation on the Bosch smoke numbers of the biodiesel fuels was not statistically significant.

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