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by R.M. Alagu and E. Ganapathy Sundaram
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Nitrogen Oxide Emission in Biodiesel Fuelled CI Engines - A Review

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Abstract- **Biodiesel as an augmenting fuel to petroleum diesel has advantages like lower CO, lower CO_2, decreased particulate matter emission and lower UBHC and few disadvantages like poorer cold flow characteristics, lower heating values, and mostly reported higher emissions of oxides of nitrogen (NOx = NO + NO_2, where NO is nitric oxide and NO_2 is nitrogen dioxide). Emission of oxides of nitrogen is the focus of this review article. Formation of NOx is a complex process and gets affected by several parameters like injection timing, adiabatic flame temperature, radiation heat transfer, and ignition delay. This article provides a review of the rich literature describing these parameters, and provides additional insight into the system responses that are manifested by the use of biodiesel.**

I. INTRODUCTION

Though biodiesel as fuel in compression ignition engine results in substantial reduction in energy consumption from life cycle perspective and reduces the atmospheric carbon concentration [1], is facing challenge in terms of increased NOx [2] emission and the same is termed as biodiesel NOx penalty. Lot of research is going on to understand the mechanisms causing increased NOx emission with biodiesel fuel in compression ignition engines. This paper deals with biodiesel composed of glyceride-free mono-alkyl esters converted from triglycerides such as biologically based fats and oils. Direct usage of such vegetable feedstock oils like rapeseed, soya bean, cottonseed, etc. without modification in engines cause several damages to engine. Hence modification is done with transesterification by reacting with alcohol, usually methanol, in the presence of a catalyst (such as sodium or potassium hydroxide).

The advantages of biodiesel usage include: reduced green gas emission, bio degradable, renewable source of energy and no major modifications needed for traditional CI engine. The disadvantages include: lower heating value, lower volatility, lower cold flow properties and higher NOx emission

A. Objective

The basic objective of this paper is to give more insight on the fundamental understanding through a thorough literature survey and to help continuing study of changes in NOx emission on compression ignition engines using biodiesel as fuel. The authors have reviewed more than 100 articles and a comparison is given for biodiesel and petroleum diesel fueled engines and the corresponding NOx emissions. Also this article deals with the potential factors contributing to the differences in NOx emission between biodiesel and petroleum diesel fueled CI engines include injection timing, ignition delay, combustion stages and heat release, combustion temperature, heat radiation and system response issues.

B. Biodiesel at a Glance

The ASTM (D6751 -09) [2], specification of biodiesel is given in Table 1 for reference. The properties of biodiesel mostly depend on both the transesterification process and feedstock. All AASTM specifications requires the fuels (either 100% biodiesel or blended fuels) to meet the specification standards, regardless of biodiesel feedstock.

Table 2 provides measured properties of diesel, B100 and B20 as reported in several references [3, 1-7]. Some of the properties are not provided exclusively in the reviewed literatures. Interpolation between diesel and B100 may be used to evaluate such properties. Because the fatty acid content varies for various feed stocks, it becomes important to understand which feedstock is used to make biodiesel, as this will affect the number of carbon atoms per molecule and the bond structure of the molecule.

TABLE I
BIODIESEL SPECIFICATIONS

Properties	*Limits*	*Unit*
Calcium and magnesium, combined	5 max	PPM (μg/g)
Flash point	93 min	˚C
Methanol content	0.2 max	mass%
Water and sediment	0.050 max	% volume
Kinematic viscosity,40˚C	1.9—6.0	mm^2 / s
Sulfated ash	0.020 max	% mass
Sulfura	0.0015 max	% mass
Copper strip corrosion	No. 3 max	---
Cetane number	47 min	---
Cloud point	Report	˚C
Carbon residue	0.050 max	% max
Acid number	0.50 max	Mg KOH/gm
Cold soak filterability	360 max	seconds
Free glycerin	0.020 max	% mass
Total glycerin	0.240 max	% mass
Phosphorus content	0.001 max	% mass
Distillation temperature	360 max	˚C
Sodium and potassium, combined	5 max	PPM (μg/g)

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TABLE II

DIESEL, BIODIESELBLEND PROPERTIES (MEASURED)

Property	***Unit***	***Diesel***	***B100***	***B20***
Carbon content	Mass %	86.7	77.1	---
Hydrogen	Mass %	12.71	11.81	---
Oxygen	Mass %	---	10.97	---
Nitrogen	Mass %	0.0001-0.003	0.002—0.007	---
C/H ratio (mass basis)	---	6.82	6.53	---
Saturated content	Vol %	63	---	---
Olefins content	Vol %	1.3	---	---
Aromatics content	Vol %	35.7	---	---
Sulfur content	Mass %	0.041	<0.005	0.00205
Typical formula	---	$C_{14.09}H_{24.78}$	$C_{18.74}H_{34.43}O_2$	---
Average molecular weight	g/mol	193.89	291.62	---
Density, 21˚C	g/mL	0.8537	0.8814	0.8577
Kinematic viscosity,40˚C	mm^2/s	2.8271	4.2691	2.862
Cloud point	˚C	-20-0	-5-10	-12.0
Cold filter plugging point	˚C	-25-0	-5-10	--
Pour point	˚C	-35-0	-15-10	--
Bulk modulus of compressibility (as a function of pressure)	MPa	12.392*P +1595.1	11.316*P + 1410	--
Speed of sound (as a function of pressure)	m/s	4.5129*P + 1375.8	3.8555*P + 1410	--
Flash point	˚C	67	141	--
Higher heating value	MJ/kg	45.339	39.871	--
Lower heating value	MJ/kg	42.64	37.388	--

These molecular attributes eventually affect fuel properties. Physical and chemical processes within a diesel engine - such as injection timing, fuel vaporization, and ignition delay - are altered for the use of biodiesel fuel relative to petroleum diesel fuel. Table 3 indicates how the relative difference in the property affects various engine parameters. For example, the first row states that the liquid density of biodiesel is relatively higher than that of petroleum diesel (indicated by the plus sign). As a result of biodiesel having a higher density, it: 1) partially influences the fuel injection timing by advancing it (plus sign), 2) partially influences the fuel injection pressure (for non-common rail fuel systems) by increasing it, 3) partially influences fuel spray penetration by decreasing it, 4) partially influences fuel spray angle by decreasing it, 5) has no influence on fuel spray atomization, 6) partially influences ignition delay by increasing it, 7) and 8) has no influence on heat release or combustion temperature.

TABLE III

THE EFFECTS OF PROPERTY DIFFERENCES

Properties	***Difference***	***Injection timing***	***Injection pressure***	***Spray penetration***	***Spray angle***	***Spray atomization***	***Ignition delay***	***Heat release***	***Combustion temp***
density	+	+	+	-	-			+	
Bulk modulus of compressibility	+	+	+	+					
Speed of sound	+	+	+	+					
Viscosity	+	+			-	-	+		
Surface tension	+	+		+		-			
Vapour pressure	-	-				-	+		
Volatility	-					-			
Liquid specific heat	-					+	-		
Vapour specific heat	-					+	-		
Heat of vaporization	+						-		
Chain length	+	-					-		
Oxygen content	+	+						+	+
Aromatic content	-	+						+	+
Sulfur content	-							+	
Saturation	-						+		+
Cetane number	+						-		
Heating value	-								-

Table 3 summarizes explicitly what is reported in the cited literature for a corresponding property. Some fuel properties in Table 3 are closely related, such as bulk modulus of compressibility and speed of sound, as can be shown by their similar effects on engine parameters. Some engine parameters are also closely related, such as injection timing and ignition delay, as can be shown by their dependence on similar fuel properties. In general chemical properties of a fuel (e.g., hydrocarbon structure, aromatic content) are suggested to dominate a fuel's effect on combustion and emission formation [8,9]. Physical properties (e.g., viscosity, density) generally affect injection timing, fuel atomization, and fuel evaporation, which eventually have indirect effects on combustion and emission formation.

C.Generic No_x Formation and Trend

The potential effects of biodiesel on thermal mechanism based on the extended Zeldovich mechanism is represented by reactions (1) through (3), which involves atmospheric nitrogen and occurs during combustion or shortly thereafter in the post-flame gas region [10]:

$$O + N_2 \leftrightarrow NO + N \ldots\ldots\ldots\ldots\ldots(1)$$
$$N + O_2 \leftrightarrow NO + O\ldots\ldots\ldots\ldots\ldots(2)$$
$$N + OH \leftrightarrow NO + H\ldots\ldots\ldots\ldots\ldots(3)$$

Equation (4) below represents the NO concentration.

$$d[NO] / d[t] = (6 \times 10^6 / T^{1/2}) \exp(-69{,}090/T)[O_{2eq}]^{1/2}[N_{2eq}] \times (\text{mole/cm}^3\text{-sec})\ldots\ldots..(4)$$

In above equation (1), [NO] represents the concentration (moles/cm^3) of NO at time t (sec), and $[O_{2,\ eq}]$ and $[N_{2,eq}]$

represent the equilibrium concentrations (moles/ cm^3) at temperature T (K) of oxygen and nitrogen, respectively. Many literatures reveal inconsistent trends in the effect of NOx emissions with the use of biodiesel. The distribution of NOx emission trends between biodiesel and petroleum diesel is represented by pie charts given below.

II. FACTORS CONTRIBUTING TO NOX FORMATIION

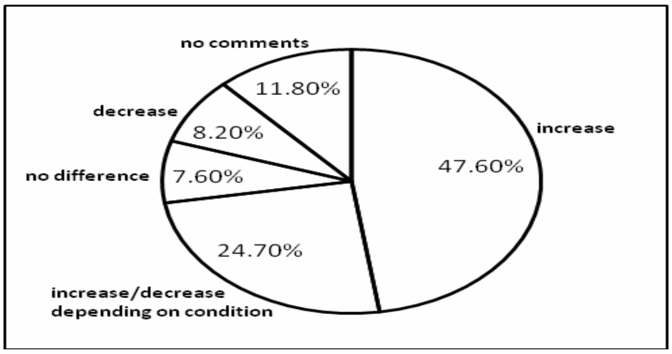
A.Fuel Injection Timing

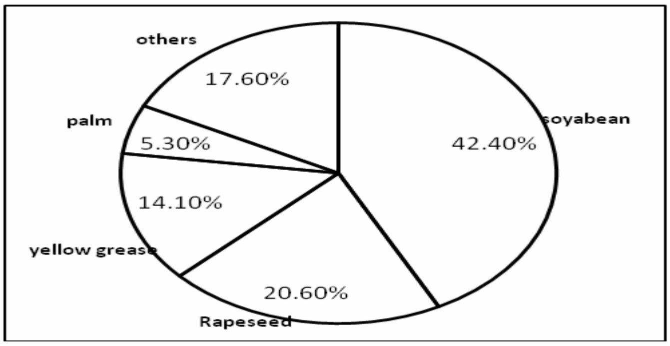
In many literatures it is reported that the start of fuel injection is advanced for biodiesel relative to petroleum diesel. For example, in the experiment carried out by Monyem et al. [11] actual injection timings occurred earlier with biodiesel in spite of no changes to the injection timing setting. The observed artificial advance of injection timing is attributed to differences in the fuels' densities, bulk modulus of compressibility, and speed of sound [12,13].

Essentially, these property differences cause a faster pressure rise within the fuel injector; since diesel fuel injector needle valves are hydraulically opened, a faster rise in fuel pressure will cause the needle to open earlier [14,15]. In connection with the discussion in the above section, this effect on NOx is considered a system-response effect.

Oxygen creates a permanent dipole moment in the molecule; this dipole moment results in stronger hydrogen bonding and increased molecular affinity of oxygenated fuels, compared to

PIE CHARTS





pure hydrocarbon fuels. These factors reduce the free space between molecules in biodiesel, decreasing its compressibility (high bulk modulus) relative to diesel fuel [16].Biodiesel may also introduce controlled system responses resulting from a controlled change to injection timing [5,17,18,19]. This becomes a controlled system response for biodiesel because a longer injection pulsewidth is required to match engine torque in a comparison between petroleum diesel and biodiesel (biodiesel's lower heating value requires a longer injection pulsewidth to deliver roughly the same amount of energy). Injection timing advance generally lengthens the ignition; this allows additional time for premixing fuel and air and generally increases the premix portion of diesel combustion [20]. An increase in the premix portion of diesel combustion increases reaction temperatures when diffusion combustion commences, elevating diffusion reaction temperatures and ultimately post-flame gas temperatures. As described earlier with Equation (1), an increase in post-flame gas temperature increases NO formation rate.

B. Ignition Delay

For biodiesel, the ignition delay is generally shorter than that for petroleum diesel [21,9,15,22,23-27]. For example, Nagaraju et al. [25] show no statistical change in start of injection between bio- and petroleum-based fuels, yet show a 29% shorter ignition delay (about 2 crank angle degrees) for B20. Fig. 1 (a) shows the normalized mass fraction burned curves (calculated from measurements of in-cylinder pressure [28]) as functions of engine crank angle (in degrees after top dead center, or deg ATDC) for petroleum diesel (reference) and biodiesel at 1400 rev/min, normal-load condition [29]. Notice that, in spite of having the same injection timing, biodiesel ignites (i.e., shows positive mass fraction burned) sooner than petroleum diesel fuel.

The relative shortening of ignition delay is reported to vary with engine load and speed. Lu et al. [13] report that biodiesel ignition delay is 10%-42.9% shorter (relative to petroleum diesel) from a low engine load to a high engine load, respectively, at 1300 rev/ min. Similarly, biodiesel ignition delay is 17.2%-35.3% shorter from a low engine speed to a

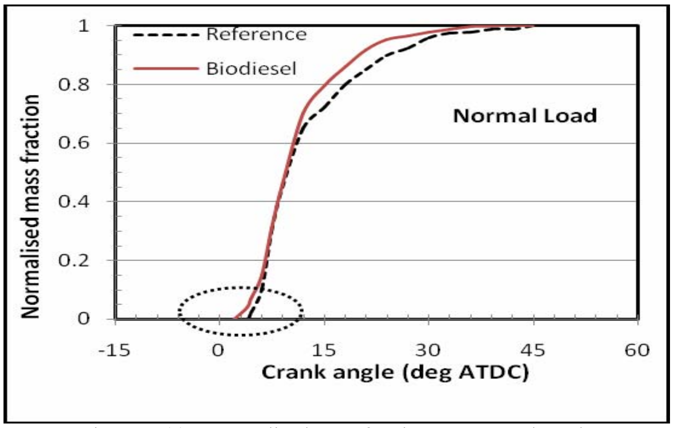


Figure. 1 (a). Normalized mass fraction versus crank angle

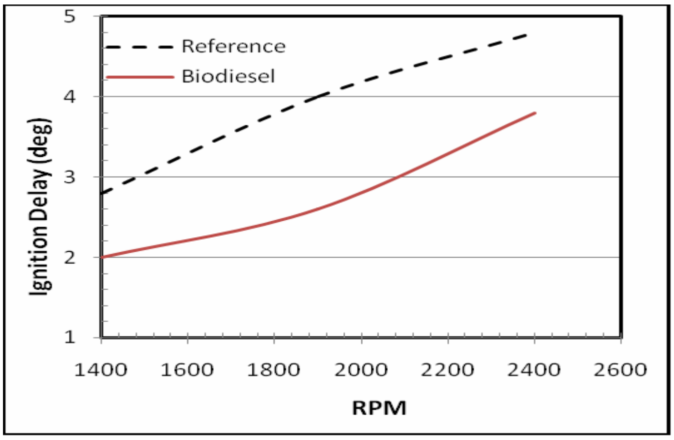


Figure. 1 (b). Ignition delay versus RPM

high engine speed, respectively, at 75% engine load. These trends may depend on the type of injection system (or other parameters), however, as Fig. 1 (b) reveals varying relative changes to ignition delay between petroleum diesel and biodiesel.

Several studies consider the physical and chemical properties affecting biodiesel ignition delay [30,9,10,27]. These studies identify that biodiesel vapor pressure is relatively lower than petroleum diesel. The lower vapor pressure results in slower evaporation rate, which tends to increase ignition delay. Biodiesel's higher surface tension contributes to an advance in injection timing; its influence on ignition delay is complex due to competing interactions with atomization, penetration, droplet size, and evaporation [31]. Biodiesel's lower heat capacity allows its temperature to rise at a faster rate once injected, resulting in evaporated droplets sooner than petroleum diesel. This effect presumably contributes to a shorter ignition delay. Related to chemical properties, most studies tend to identify the increased chain length of biodiesel as the primary contributor to shorter ignition delays [9,10,27]. Double-bond structuring of fatty acid chains (i.e., increased unsaturation of the fatty acid ester) tends to lengthen ignition delay.

C. Combustion and Heat Release

According to many reports, the overall combustion characteristics of low concentration blended biodiesel with petroleum diesel (B5) are quite similar to those of pure petroleum diesel [32]. Comparisons between pure biodiesel and pure petroleum diesel, however, reveal interesting dissimilarities, as shown in Fig. 2 (a) (b). It is observed that a short ignition delay generally results in a smaller fraction of premixed burn and a correspondingly higher fraction of diffusion burn [20]. Petroleum diesel at the normal-load condition exhibits mostly premixed burn; diffusion burn becomes dominant late in the heat release (around 15˚ ATDC). Biodiesel, on the other hand, exhibits diffusion burn much earlier (around 6˚ ATDC).

In addition to the shorter ignition delay of biodiesel (e.g., higher cetane number), its lower volatility is cited as also contributing to decreased fractions of premixed burn. The lower volatility results in fewer evaporated droplets during the ignition delay period (which is already shortened due to biodiesel's naturally shorter ignition delay), further compounding a reduction in premixed burning (relative to petroleum diesel).

The other interesting feature to evaluate between biodiesel and petroleum diesel fuels is the combustion duration. Several studies [21,4,15,33,34,35,36] report a general increase in the combustion duration with biodiesel. However, as per Bittle et al. [29], biodiesel combustion duration is generally shorter than petroleum diesel combustion duration. In all cases, biodiesel seems to terminate combustion sooner. If combustion duration is defined as the 10%-90% burn angle, then biodiesel has shorter combustion durations across several speeds and loads.

The observed shorter durations may result from biodiesel exhibiting a faster diffusion burn rate than petroleum diesel. This is evidenced in Fig. 3 (a) (b), where initially biodiesel and petroleum diesel may have similar burn rates, but eventually petroleum diesel's burn rate slows as its combustion becomes predominantly diffusive.

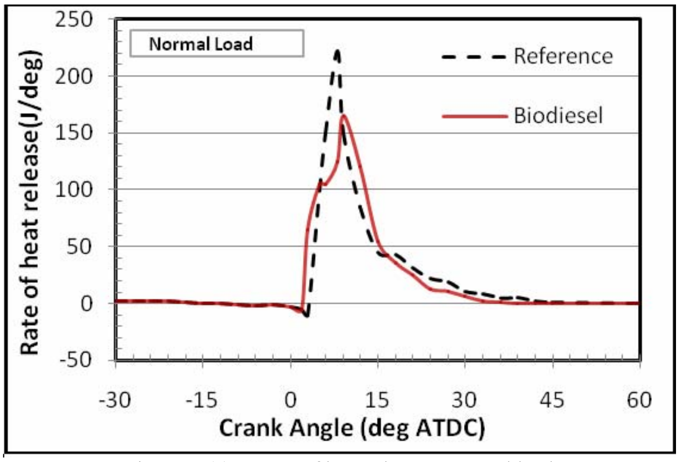


Figure. 2 (a). Rate of heat release at normal load

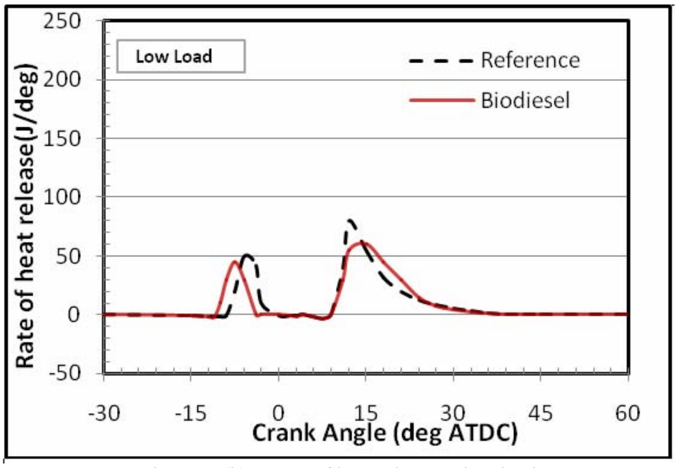


Figure. 2 (b). Rate of heat release at low load

In literature, not only there seems to be inconsistency regarding the relative combustion durations between biodiesel and petroleum diesel, there also seems to be some inconsistency in the magnitudes of heat release between the two fuels. They attribute these changes in combustion characteristics to the presence of oxygen in the fuel and late burning of heavy compounds found in biodiesel. Bittle et al. [29], however, reveal lower rates of heat release for biodiesel fuel, as shown in Fig. 2 (a) (b). In a related study [37], similar trends (lower heat release of biodiesel relative to petroleum diesel) are observed at other speed and load conditions.

D. Heat Radiation by In-Cylinder Soot

All the review papers report significant reductions in PM (or soot) emission when substituting biodiesel into petroleum diesel. An example of PM decreasing with increased biodiesel content, taken from [38] is shown in Fig. 4. There are reports [10,39] attributing the reduction of in-cylinder soot as a possible cause for the differences in NOx emissions between biodiesel and diesel combustion. Carbonaceous soot, which mostly forms through premixed reactions in the core of a diesel spray jet, acts as an effective heat radiator as it oxidizes through the diffusion flame [35]. This radiative heat transfer, which is an energy transfer mode, results in a reduction in flame temperature (relative to the case of no radiation heat transfer).

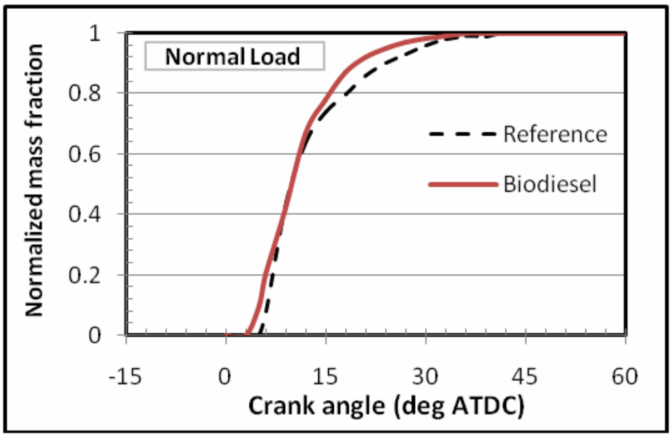


Figure. 3 (a). Normalized mass fraction at normal load

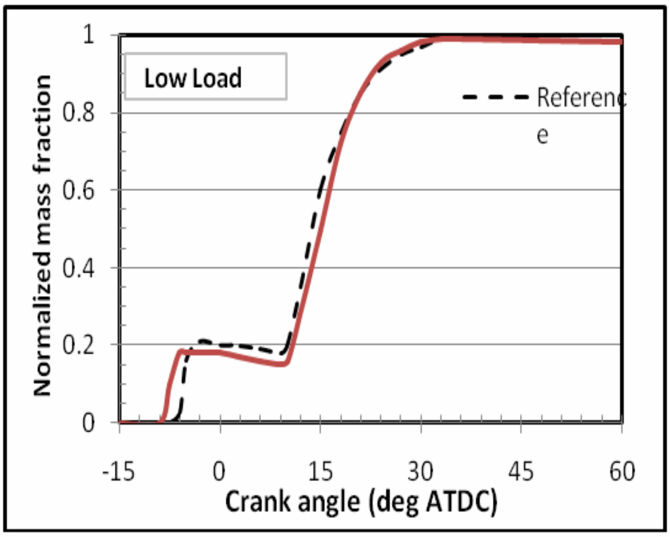


Figure. 3 (b). Normalized mass fraction at low load

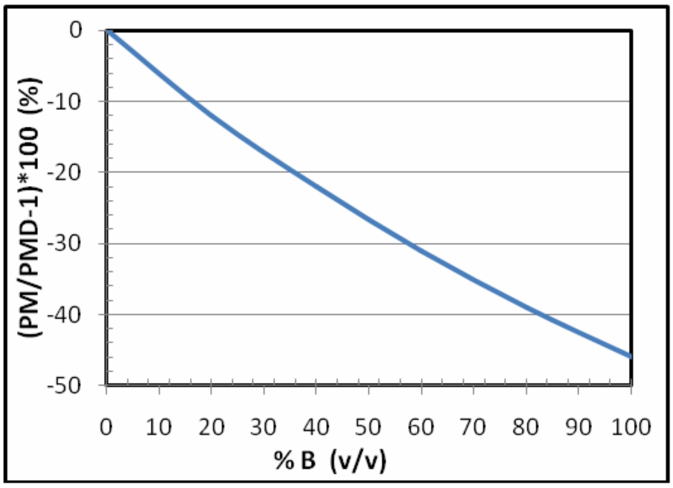


Figure. 4. Reduction in PM

The important mechanisms that are reported to contribute to lower soot formation in biodiesel are (a)increased fuel-bound oxygen, (b) lower stoichiometric air-fuel ratio (decreased equivalence ratio for same actual air-fuel ratio), (c) advanced start of combustion (due to artificial advance and shorter ignition delay), (d) decreased boiling point and (e) different sooting tendencies of various biodiesel esters [5,8,40-41,42,43,44-46]. The presence of fuel-bound oxygen suppresses soot nucleation early in the formation on the fuel side of the mixing-controlled flame. This nucleation suppression inhibits peak soot production, leads to more complete burning, and reduces the formation of soot precursors (cracked hydrocarbons such as acetylene and ethylene).

Other literatures [40,45,46] report that overall oxygen content should be about 30-40% to reduce soot precursors to close to zero. Considering that the oxygen content in biodiesel is around 10% the oxygen-induced soot reduction is noticeable but not extreme. Furthermore, blends of biodiesel with conventional diesel assumedly would have proportionally smaller effects on in-cylinder soot reduction. Also, a study by Mueller et al. [46] suggests that the biodiesel-bound oxygen may be underutilized for reducing soot precursors.

E. Combustion Temperature

Higher reaction temperature of biodiesel combustion than petroleum diesel combustion [21,47,15] is the most-commonly cited reason for differences in NOx emissions between the two fuels [48,49]. Conceptually, increased NOx could result with biodiesel if the stoichiometric adiabatic flame temperature of biodiesel is higher than that of diesel fuel [39]. In spite of its lower heating value, biodiesel also has a lower stoichiometric air-fuel ratio. Thus, when computing the stoichiometric adiabatic flame temperatures for petroleum diesel and biodiesel, they may closely match. In all cases, biodiesel has a lower adiabatic flame temperature than petroleum diesel. From microscopic points of view, characteristics of biodiesel such as degree of saturation have significant influence on the flame temperature. The number of carbon single bonds in the molecule reflects degree of saturation. Increased single bonds mean increased saturation. The iodine number is a measure of saturation, where low iodine number indicates high saturation (increased single bonds). This connection between degree of saturation and flame temperature/ NOx emissions is supported by the tendencies of different biodiesels with different degrees of saturation to produce different amounts of NOx [50].

F. System Response

System responses broadly fall into two categories, namely "passive" and "active". The passive system response is due to difference in certain properties and active system response is due to changes to the operating point on the calibration map as a result of extending the injector pulsewidth to match engine torque. The advance in injection timing may be manifested by a difference in property (i.e., bulk modulus) between biodiesel and petroleum diesel- a passive system response. A change in injection timing may also be manifested by a change to the operating map from which control parameters are specified- an active system response. In other words, because the injection pulsewidth is typically longer with biodiesel (due to its lower heating value) and most engine control systems use pulsewidth as an input to the calibration map (which controls parameters such as injection timing), the changed injection pulsewidth may change other controlled parameters (in spite of the same engine speed and torque).

The elimination of a passive system response would require a substantial design change to the system component being affected. In the example of bulk modulus manifesting an earlier injection timing of biodiesel, this response could be designed out of the system by employing common-rail fuel system. The manifestation of an active system response, however, is rather arbitrary. Tat et al. [51], for example, observe an advance in injection timing as injector pulsewidth increases. Such system responses make it difficult to quantify specific reasons that may cause differences in NO emissions between biodiesel- and diesel-fuelled engines and likely contribute to the variations in NO emissions that are reported in the literature.

III. SUMMARY

The major factors that seem to contribute to changes in NO emissions between biodiesel and petroleum diesel fuel are provide in a summary fashion in Fig. 5. A "shaded bold box" connecting two parameters suggests that an increase (or advance) in the lower order parameter increases (or advances) the higher order parameter. For example, a "non-shaded bold box" line connects radiation heat transfer to post-flame gas temperature; that is, an increase in radiation heat transfer decreases post-flame gas temperature.

A lower order parameter that is "dashed box" indicates that such a parameter is higher with biodiesel than with petroleum diesel. For example, fuel-bound oxygen is given as dashed box; indicating that biodiesel has higher fuel bound oxygen than the petroleum diesel. For some lower order parameters, either through reported uncertainty in the literature or no found literature discussing the topic, the relative differences between biodiesel and petroleum diesel are not known. In these cases, the lower order parameters are given as dashed boxes. Thermodynamically, adiabatic flame temperature is a function of heating value and stoichiometric air/ fuel ratio.

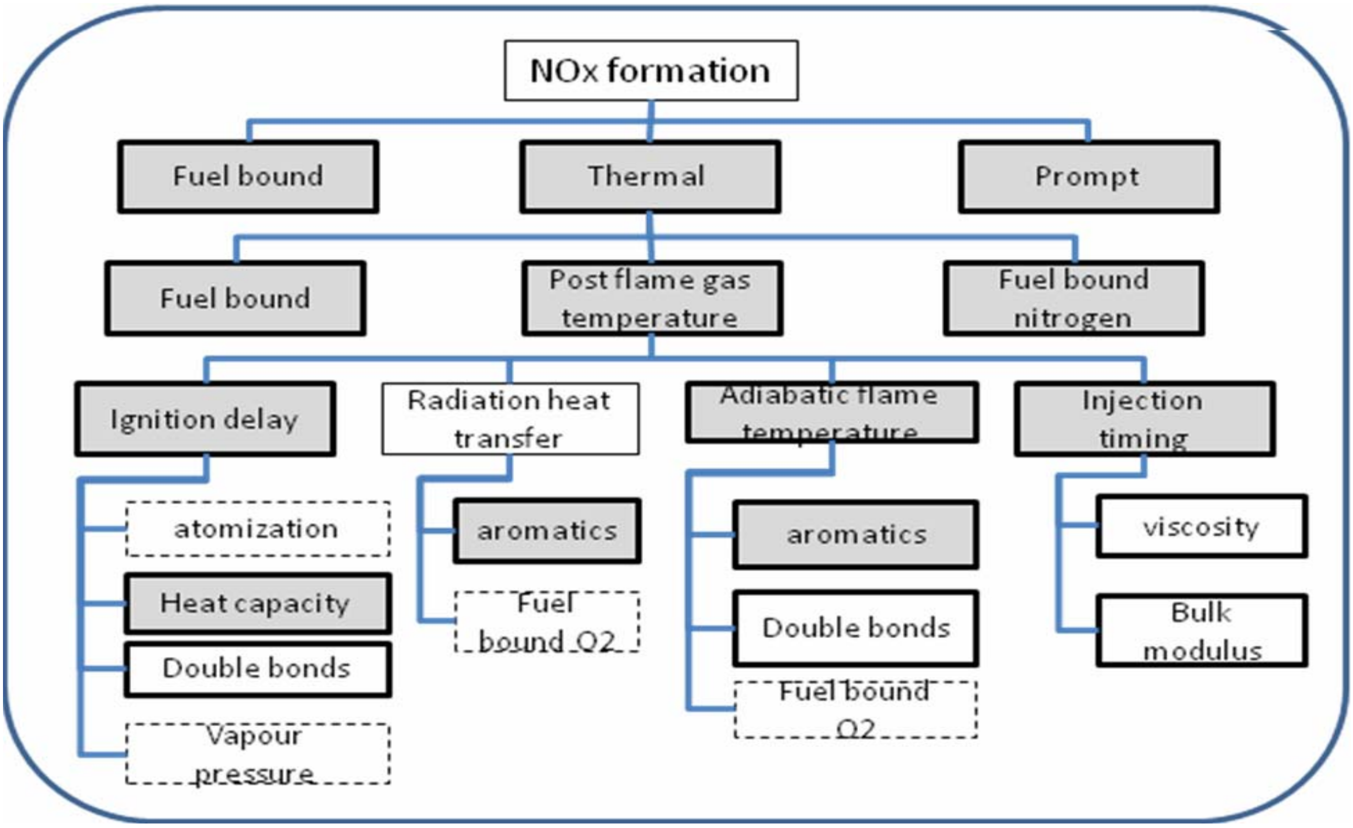


Figure. 5. Nitrogen Oxide formation summary

IV CONCLUSION

The major reported reasons for observed differences in NOx emissions between biodiesel and petroleum diesel fuels are as follows:

- Combustion related: Advances in start of combustion, manifested either by biodiesel's inherently shorter ignition delay or the combination of biodiesel's inherently shorter ignition delay and artificial advances in injection timing. Further, biodiesel has differences in heat capacity, vapor pressure, and atomization which affect physical ignition delay. In addition to elevating combustion temperatures, a shorter combustion duration coupled with shorter ignition delays creates longer residence time for post flame nitrogen chemistry to occur.
- Adiabatic flame temperature: Contributing factors for adiabatic flame temperatures are the fuels' heating values and stoichiometric air/fuel ratios. Biodiesel fuels consistently have lower heating values than petroleum-based fuels. They correspondingly also have lower stoichiometric air/fuel ratios, which in the balance may yield stoichiometric adiabatic flame temperatures that are similar between the fuels. Several fuel parameters participate in affecting adiabatic flame temperature, including the amount of fuel-bound oxygen, the level of aromatics in petroleum diesel, and the number of double bonded species in biodiesel.
- Radiation heat transfer: Decreased radiation heat transfer with biodiesel, manifested by the decreased concentrations of aromatic species and increased concentrations of fuel-bound oxygen, tends to increase post-flame gas temperature (and thus increase NO emissions). Although the decreased radiation heat transfer from decreased aromatic species may raise post-flame gas temperature in biodiesel, the higher aromatic species concentrations in petroleum diesel may correspondingly raise flame temperatures in petroleum diesel.
- Engine design and technology: Inconsistencies in reported NOx emissions behavior between biodiesel and petroleum diesel fuels among studies may largely be linked to differences in engine design and technology. Study comparisons using different sized engines (based on displacement), different fuel systems (i.e., common rail versus in-line/mechanical pump systems), different air systems (e.g., naturally aspirated versus turbocharged), and different control systems (mechanical versus electronic) will likely repor variability in NOx emissions behavior between fuels

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