Biodiesel Soot Incandescence and NO Emission Studied in an Optical Engine

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High-speed imaging and thermodynamical characterization are applied to an optically accessible, heavy-duty diesel engine in order to compare soot incandescence and NO emission behaviour of four bioderived fuels: rapeseed-methylester, Jatropha oil (pure), Jatropha-methylester and a 50/50 blend of cyclohexanone with a Fischer-Tropsch synthetic fuel. Regular diesel fuel is used as a reference. Soot incandescence is observed at 0.3° crank angle resolution (200 images/cycle). The heat release rate and exhaust NO concentrations are used as indicators of average and peak temperatures, respectively, which are combined with soot incandescence signal to get a relative measure for a fuel's sooting propensity.

Introduction

Currently a lot of discussion is taking place about the use of bio-derived fuels for automotive purposes. Competition with food crops and alleged CO_2 neutrality are some of the issues and for instance Ref. [1] contains a comprehensive review of the many aspects concerning *Jatropha curcas L.* Aspects often overlooked are the emission of soot and NO from combustion of biodiesels. Ever stricter regulations are enforced with regard to these two pollutants and several strategies are possible to reduce their emission, like aftertreatment, exhaust gas recirculation or improved fuel injection techniques. An alternative aspect receiving less attention is fuel composition. Much work has already been done on oxygenated fuels from either biological or fossil feedstock (see Refs. [2-4] and references therein).

This paper focuses on the behaviour of soot incandescence and NO exhaust concentration of four biofuels and a comparison to regular diesel fuel is made.

Experimental setup

All measurements are performed on a six cylinder, heavy-duty Diesel engine. One of the cylinders is modified for optical access through quartz windows at various locations. Full details are given in Ref. [2]. In the measurements reported here, the common-rail pressure is 120 MPa and the cylinder boost pressure is 0.14 MPa (abs). The common-rail pressure, needle lift signal and cylinder pressure signals are recorded during injection and averaged over 20 cycles. An example of the common-rail pressure during injection is presented in Fig. 1. Details of exhaust NO measurements and analysis are given in Ref. [5]. The rate of heat release (RoHR) is calculated from the cylinder pressure.

A Phantom V7.1 digital high-speed camera observes the combustion luminosity through the piston window, which in the visible consists of soot incandescence. The camera is synchronized to the crankshaft of the engine and images are recorded every 0.3° ca ($\approx$35 µs). To avoid overexposure in intensity measurements, an exposure time of 2 µs is used. Detection of the fuel start of delivery (SoD) is enabled by illumination with a continuous-wave Ar$^+$ laser and recording elastic scattering with a longer exposure time of 24 µs. This also allows better localization of the first soot incandescence.
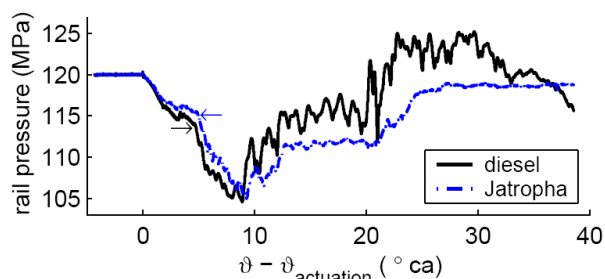

Fig. 1: Common rail pressure transients during injection of diesel and Jatropha oil. The latter exhibits only minor pressure oscillations during injection (due to high viscosity), whereas those of diesel are clearly visible. Pressure curves of the other fuels are almost identical to the one of diesel. Arrows indicate the start of fuel delivery.

Fuels

The bio-derived fuels used are commercially available rapeseed-methylester (RME), pure Jatropha oil (raw vegetable oil), Jatropha-methylester (JME) and a 50/50 blend of cyclohexanone with a Fischer-Tropsch (FT) synthetic fuel (the blend is referred to as CHxnO). Although it is not trivial, liquid cyclic oxygenates like cyclohexanone can be made from lignocellulosic biomass [4]. Regular diesel (EN590) fuel is used as a reference. Elementary data about the fuels is given in Ref. [2]. The low cetane number cyclohexanone is blended with FT to obtain a cetane number similar to the three bio-derived fuels and the oxygen content is

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approximately equal for all biofuels (9 - 10%). Cyclohexanone has a cyclic molecular structure, whereas that of Jatropha oil is branched and for JME and RME it is linear; all biofuels also contain double bonds [2].

Results and discussion

Phase averaged images of 10 consecutive injections are shown in Fig. 2. The corresponding heat release rates are shown in Fig 3. The images (Fig. 2) reveal clear differences in the early soot location for different fuels. This is corroborated by the corresponding standard deviation images (not shown).
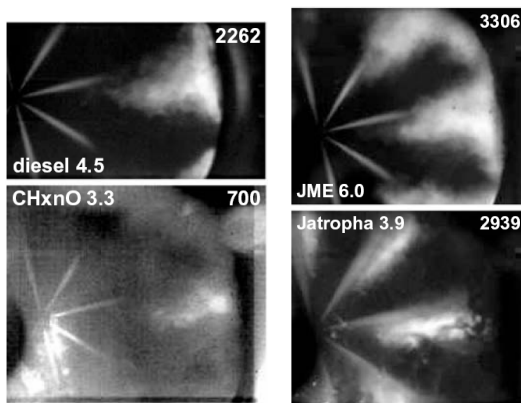

Fig. 2: phase average over 10 injections of various fuels (indication of θ [° aTDC] behind fuel name; θ_{SoD} = -4.5° aTDC, except CHxnO for which θ_{SoD} = -9.5° aTDC; intensity for white indicated in upper right corner).

Diesel, RME and JME have quite similar behaviour, in that first soot is detected between the end of the liquid spray and the cylinder wall and the soot vapour region expands both towards the injector and along the cylinder wall. For CHxnO, very weak soot incandescence is first detected along the full perimeter of the cylinder and only slightly later a brighter region of soot grows towards the liquid core. Jatropha oil, however, behaves markedly different: soot originates leeward of the liquid spray, but only along its downstream half. Later it slowly grows towards the cylinder wall, which is eventually fully exposed to soot.

Figure 3 also presents the total soot incandescence intensity, integrated over the field of view (a more detailed analysis is in progress). The spectral emittance of soot is governed by its temperature distribution: a higher T not only results in a higher total amount of radiation ($\propto T^4$), but also moves the emittance maximum closer to the spectral observation window. This allows the observed soot incandescence to be approximated by T^{13} for the range 1800 - 2700 K in this work [2], so only the hottest soot is observed. This is observed in Fig. 2. For a heavily sooting fuel, the soot cloud may be optically thick, causing some incandescence to be partially obscured by soot particles closer to the detector. This would lead to an underestimation of the total soot incandescence and hence of the amount of soot.
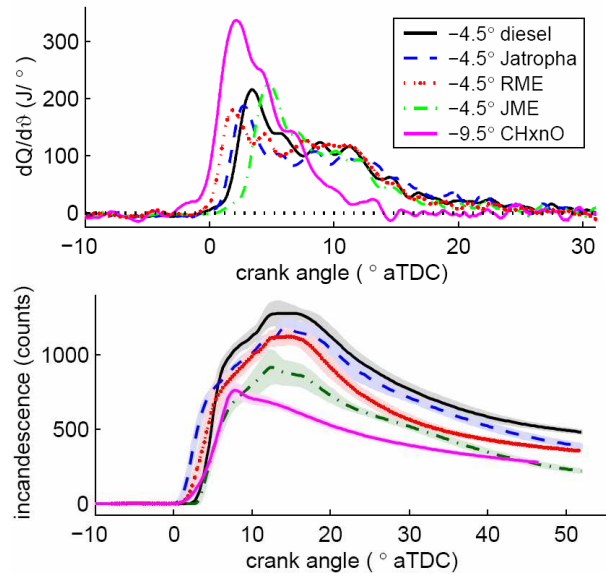

Fig. 3: Heat release rates and soot incandescence (θ_{SoD} indicated). Shaded area represents standard deviation of phase averaging. Note the different horizontal scales.

Significant differences between fuels can be observed in Fig. 2. Ignition delay and air entrainment (which are related) can affect soot incandescence. Soot is observed at a somewhat larger distance from the injector for CHxnO than for the other fuels in Fig. 2 and CHxnO also has a larger ignition delay. Both these factors indicate that air entrainment may be better and consequently soot incandescence lower for CHxnO. A more detailed study involving soot lift-off lengths for all fuels is in progress.

Jatropha oil has an extremely high viscosity, compared to its esterified counterpart JME or commercial diesel. This can already be seen in the much smaller oscillations in its common-rail pressure transient (Fig. 1). It also results in poor fuel atomization. Combined with its relatively short ignition delay, this may explain why its soot is observed much closer to the injector than for most other fuels (Fig. 2). A short ignition delay is not enough on its own, however, since the soot incandescence location of another non-bio-derived oxygenated fuel with similar ignition delay as Jatropha oil (discussed in [2]) resembles much more that of diesel than that of Jatropha oil. For reasons not yet fully understood, it is believed that the dissimilarity in viscosity is (partially) responsible for the observed differences in soot location for Jatropha oil.

The general shape of the soot incandescence curves (Fig. 3) is influenced by the amount of soot and its temperature, as discussed above. An average temperature indication can be derived from the heat release rate, but locally temperatures and

hence soot incandescence may deviate appreciably. As a measure for the local peak temperature the exhaust NO concentration is used [2], since most of it is formed in diesel combustion through the thermal process [5].
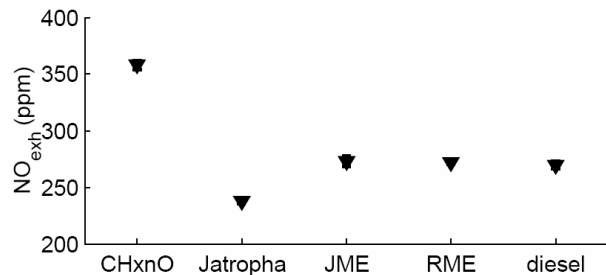

Fig. 4: NO exhaust concentration of the fuels used (uncertainty is approximately symbol size).

The results of exhaust NO measurements are presented in Fig. 4 and clearly indicate that CHxnO has a much higher NO production than other fuels used here. A higher NO production by cyclohexanone blends is also observed in Ref. [3]. Consequently, the temperature is expected to be higher as well during combustion of these two fuels. This is also reflected in the magnitude of the RoHR curves (Fig. 3). Therefore the effect of temperature on soot incandescence will be significant. Yet CHxnO has the lowest of all soot incandescence signals presented. This is a strong indication that CHxnO combustion produces the least amount of soot from all fuels discussed here.

It may be argued that the relatively large ignition delay of CHxnO plays an important role in its lower soot production, since this leaves more time for mixing and combustion in regions closer to stoichiometric, thus reducing soot formation. But in Ref. [3] a fuel blend of cyclohexanone with only 5% oxygen is compared to a dibutylmaleate blend containing 9% oxygen: their ignition delays are found to be almost identical, as are their particulate matter (soot) emissions. So in that case the mixing times are equal, yet with much less oxygen incorporated, the cyclohexanone blend reaches the same soot reduction. Thus, although ignition delay may be important in soot abatement, clearly another yet undisclosed mechanism, of physical or chemical nature, is expected to be responsible for the large soot reduction capabilities of cyclohexanone blends.

Comparing JME, RME and diesel, their heat release rates and NO exhaust concentrations are all quite similar, suggesting only small differences in temperature for these three fuels. Therefore the soot incandescence signal of these fuels in Fig. 3 can be considered a relative measure for their sooting propensity. For these fuels, soot incandescence is still higher than that of CHxnO, but it may be deduced that the sooting propensity decreases from diesel to RME to JME. The soot incandes-

cence of Jatropha oil is similar to that of diesel and its NO exhaust concentration (hence temperature) is only slightly lower, therefore Jatropha oil's sooting propensity is expected to be quite similar to that of diesel and thus higher than that of the other biofuels.

From the aforementioned results, it may be deduced that CHxnO produces a lower amount of soot than the other biofuels. Apart from the role of ignition delay (see above), this may imply that the strength of CHxnO lies more in suppression of soot formation, rather than enhanced soot oxidation. But this needs further confirmation from ongoing research.

Acknowledgments

References
[1] W.M.J. Achten, L. Verchot, Y.J. Franken, E. Mathijs, V.P. Singh, R. Aerts and B. Muys, *Jatropha biodiesel production and use,* Biomass Bioenergy **32**, 1063-1084 (2008).
[2] R.J.H. Klein-Douwel, A.J. Donkerbroek, A.P. van Vliet, M.D. Boot, L.M.T. Somers, R.S.G. Baert, N.J. Dam, J.J. ter Meulen, *Soot and chemiluminescence in diesel combustion of bio-derived, oxygenated and reference fuels,* Proc. Combust. Inst. **32**, 2817-2825, (2009).
[3] M.D. Boot, P.J.M. Frijters, R.J.H. Klein-Douwel, R.S.G. Baert, *Oxygenated fuel composition impact on Heavy-Duty diesel engine emissions,* SAE Technical Paper 2007-01-2018 (2007).
[4] M.D. Boot, P.J.M. Frijters, C.C.M. Luijten, L.M.T. Somers, R.S.G. Baert, A.J. Donkerbroek, R.J.H. Klein-Douwel, N.J. Dam, *Cyclic oxygenates: a new class of second generation biofuels for diesel engines?,* Energy & Fuels, (2008) (doi: 10.1021/ef8003637)
[5] K. Verbiezen, A.J. Donkerbroek, R.J.H. Klein-Douwel, A.P. van Vliet, P.J.M. Frijters, X.L.J. Seykens, R.S.G. Baert, W.L. Meerts, N.J. Dam, J.J. ter Meulen, *Diesel combustion: In-cylinder NO concentrations in relation to injection timing,* Comb. & Flame **151**, 333-346, (2007).