

Evaluating the potential of a long carbon chain oxygenate (octanol) on soot reduction in diesel engines

I. Ruiz-Rodriguez¹, R. Cracknell², M. Parkes², T. Megaritis¹, L. Ganippa¹

¹Mechanical Aerospace and Civil Engineering Department, Brunel University London, Uxbridge, UK

²Shell Global Solutions UK, Cheshire, UK

ABSTRACT

The automotive industries are facing challenges to meet stringent CO₂ and air-quality regulations. One of the factors affecting emissions performance is the fuel's chemical composition. The presence of oxygen in a fuel offers the potential to lower soot emissions, however, these oxygenates must be compatible with existing hardware and fuels. To this end, the combustion of a long carbon-chain oxygenate, 1-octanol, was studied. It was injected at high pressures into a constant volume chamber, and a high-speed, two-colour pyrometry system was used to evaluate the spatial and temporal variations of soot and temperature. A significant reduction of soot was obtained whilst the flame temperature remained similar to that for diesel.

1 INTRODUCTION

Internal combustion engines (ICEs), in particular diesel engines, are widely used in many industrial sectors such as in agriculture, rail, marine and automotive because of their high power output, thermal efficiency, reliability, and low maintenance costs. Significant developments have been made to improve the emissions of noxious compounds such as NO_x and soot [1]. Nonetheless, regulations are getting tighter, and whilst light-duty vehicles will see increasing levels of electrification ranging from mild hybrids to battery electric vehicles (BEVs), for the heavy-duty fleet, it is difficult to envisage a complete switch in the near future because of weight and range constraints. It seems clear that ICEs will remain important in the near future, so solutions that address both legislation and demand must be sought. One such approach is to improve the in-cylinder combustion processes, as this would offer several socio-economic advantages: by making use of the current engine infrastructure, implementation costs can be reduced and; reducing in-cylinder emissions would reduce the cost and load on aftertreatment systems, potentially allowing old vehicles to still operate on the roads.

In recent years, the interest for oxygenated fuels has increased due to their potential use as a drop-in fuel to help reduce emissions in diesel engine, in particular soot. In this context, a drop-in fuel is an alternative fuel to diesel that can be directly used in ICEs without major modifications to the engine. Drop-ins are normally blended with diesel at different percentages. Some oxygenates can be produced from renewable sources and even biomass, and others can be synthetically produced via microbial engineering [2-4]. One such group of oxygenates are alcohols, which are characterised by their hydroxyl functional group (-OH). The combustion of low carbon chain alcohols such as ethanol and methanol has been widely studied and characterised, and their properties are well known [5-7]. More recently, work has been done on higher carbon chain alcohols such as pentanol, hexanol, and octanol [8-10]. Higher carbon chain alcohols tend to have a higher energy content; furthermore, longer carbon chains reduce compound polarity,

meaning that they are more likely to form stable blends with diesel. In an industrial context, this means that higher alcohols have the potential to be compatible with current engine hardware and would not necessitate the addition of surfactants to mix with diesel- offering a cost-effective solution. In other studies, octanol has been implemented directly in engines and has shown promising results in terms of emission performance [10, 11]. However, most studies have been done in engines, and thus optical data on its in-cylinder combustion process and soot formation are still lacking. Spatial information of how the temperature and soot distribution in spray flames change when fuelled with octanol could help to characterise its potential as a drop-in fuel for diesel engines.

This work aims to determine the suitability of octanol for engines and its effectiveness in reducing in-cylinder soot formation when compared to diesel. In order to address this, 1-octanol was injected neat (in its pure form without blending with other components) into a high-pressure, high-temperature constant volume chamber. This allowed for the decoupling of complex cylinder motions from the combustion event and for precise control of the ambient conditions. Its combustion was studied by using the two-colour pyrometry method coupled with high-speed imaging. This was done under conditions simulating an exhaust gas recirculation (EGR) environment that is typically used to control NO_x emissions- albeit normally having a trade-off on increased soot emissions. If under these conditions a fuel can produce lower soot than diesel it could potentially help address the NO_x-soot trade-off.

2 EXPERIMENTAL METHODS

2.1 Fuels

Diesel of EN590 compatibility was used as a reference fuel, with a known cetane number (CN) of 53, density of 850kg/m³ and a lower calorific value (LCV) of ~43MJ/kg. The oxygenate studied, 1-octanol, is a linear alcohol with a carbon chain of 8 and with a hydroxyl group at one of the chain ends, as shown in Figure 1. It has a chemical formula of C₈H₁₈O with a CN of 39, density of 824kg/m³ and an LCV of ~38MJ/kg [12, 13]. From toxicity reports [14], its only known health impacts lie in the irritant category, which makes it a chemical with a relatively lower toxicity profile than diesel. This is an important fuel property, as it would make its handling relatively safe for end-users.



Figure 1. The chemical structure for 1-octanol.

2.2 The constant volume chamber

Both diesel and octanol were injected separately in their neat forms into a constant volume chamber (CVC). In these experiments, the mass of octanol injected was matched to that of diesel when injected at a duration of 1.5ms. One of the advantages of using a CVC is that the ambient conditions can be carefully controlled. For these tests, the conditions were set to an ambient temperature of ~1300K and to an ambient oxygen concentration of ~10%. This was obtained by chemically preheating the chamber with a premixed homogeneous mixture of acetylene and air, ignited by a spark plug. The combustion of the mixture would then reach high temperatures and pressures, closely monitored by a fast-response pressure transducer. Upon the decay of both temperature and pressure, the test fuel was injected at the desired conditions

mentioned previously. A multi-hole common rail diesel injector was used, but only one of the sprays was imaged to maximise the camera's resolution. The injection pressure was set at 700bar to allow for a compromise between imaging area and camera speed, set at 18,000 frames per second (fps); seven combustion events were acquired for each fuel.

The schematic of the experimental set-up that was used in this work is shown in Figure 2. The window at the bottom allows for optical access for the high-speed diagnostics and the one at the side allows for additional illumination if needed for spray visualisation. For the two-colour pyrometry method (which will be described in more detail in the following subsection) the flame emission was imaged at two different wavelengths by using two narrow band-pass filters centred at 543.5nm and 670nm, having a full-width half max (FWHM) of 10nm. This was achieved by aligning a 50/50 beam splitter with a mirror directly under it, which enabled two images of the same flame to be acquired using one camera after passing through either a green or a red filter. The data was then post-processed to extract the flame from the background and to solve for soot and temperature values.

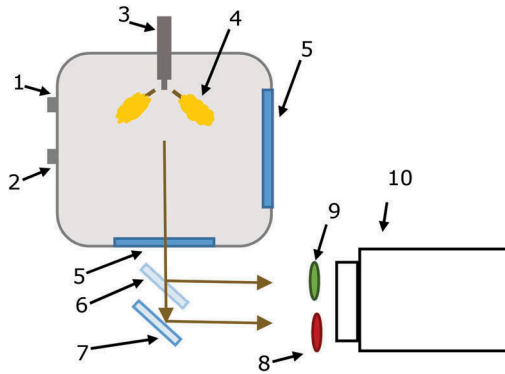


Figure 2. Experimental set-up showing the CVC and the high-speed two-colour pyrometry system. 1. Exhaust valve; 2. Gas inlet; 3. Multi-hole injector; 4. Sample flame; 5. Optical access; 6. 50/50 beam splitter; 7. Mirror; 8. Red narrow band-pass filter; 9. Green narrow band-pass filter; 10. High-speed camera.

2.3 Two-colour pyrometry

The two-colour (2C) method has been used extensively in the literature to characterise the soot and temperature distributions in flames [15-18]. The basic concept relies on the soot radiance (I_{soot}) being a multiple of the blackbody radiance (I_{BB}) and the emissivity of the body investigated (ϵ), which takes a value of <1 for a non-blackbody emitter, equation 1. The radiation emitted by the soot particles is dependent on both the wavelength (λ) and the temperature (T). By using Planck's laws one can further express I_{BB} in terms of λ , T and two constants, c_1 and c_2 known as Planck's constants, equation 2.

$$I_{soot} = \epsilon I_{BB} \quad (1)$$

$$I_{BB} = \epsilon \frac{c_1}{\lambda^5 \left[\exp\left(\frac{c_2}{\lambda T}\right) - 1 \right]} \quad (2)$$

Finally, the emissivity can be obtained by using semi-empirical equations expressed in terms of the soot absorption (KL factor) and λ [19]. One of the most widely used correlations for ϵ is that presented in equation 3, where α is the dispersion coefficient and takes a value of 1.39 in the visible wavelength range [19].

$$\epsilon = 1 - \exp\left(-\frac{KL}{\lambda^\alpha}\right) \quad (3)$$

Calibration parameters are then required to relate the pixel's arbitrary units registered in the sensor of the camera to a physical parameter such as radiance. In this work, this was done by using an integrating sphere and a tungsten halogen light source. With these data, the above equations, were solved using an in-house developed numerical solver to obtain the flame temperature and KL factor for every pixel location in the flame. The sensitivity of this technique due to the choice of the detection wavelength, selection of the dispersion coefficient and other setting-specific errors have been discussed in [20, 21]. This technique provides semi-qualitative relative information about soot and temperature between fuels.

3 RESULTS AND DISCUSSIONS

The results and discussions section has been divided into three subsections. Some generic combustion characteristics based on soot luminosity will be discussed first in §3.1. Following this, in §3.2, the spatial and temporal changes in temperature will be presented. Finally, in §3.3, the spatial and temporal characterisation of the soot evolution will be discussed.

3.1 Fuel and combustion characteristics

Attempts were made in this work to report any deterioration caused to the fuelling system when injecting with neat octanol. From purely qualitative observations, there was no significant alteration of the system behaviour based on the consistent combustion behaviour. As well, the injector remained operative as there were no injection failures. After each injection, the components were visually inspected for signs of damage but none could be observed. Additional tests were also carried out,



Figure 3. Sample of qualitative soaking test carried out to identify potential material incompatibilities with neat octanol. Sample 1, stainless steel; Sample 2, steel; Sample 3, galvanised steel; Sample 5, PVC hose; Sample 6, PTFE hose. Besides a slight swell of the PVC pipe, no material alterations were observed.

where sample materials from the fuel system and CVC were soaked in neat octanol for a period of three hours. Some of the materials tested were stainless steel, steel, galvanised steel, PVC, Viton, silicone, and nitrile, as shown in Figure 3. The only effect observed in any of the materials tested was a slight swelling of the PVC hose. This

indicates that for longer storage periods, PVC components might not be compatible with neat octanol. These tests show that octanol is potentially compatible with existing injection systems, and more so as a drop-in fuel at low percentages, as the concentration of octanol would be relatively lower than that tested here. The three-hour period was chosen to ensure that the fuel did not cause any break down of the high pressure chamber set-up, however, longer time periods would be required to ascertain its full compatibility characteristics. Even though the outcome is encouraging, these are only preliminary qualitative observations and a quantitative evaluation is required to fully characterise any material incompatibilities.

Regarding the combustion characteristics, two parameters were studied. First, the soot lift-off length (sLOL), Figure 4, and second, the end of (visible) combustion (EOC), Figure 5. The sLOL is the distance between the injector tip and the point at which soot has stabilised. The data in each figure represent an ensemble average of the seven injections for each fuel, with the error bars representing two standard deviations (SDs). The values were time-averaged over the stable sLOL period, which occurred from 985 μ s after the start of injection (aSOI) to 2217 μ s aSOI.

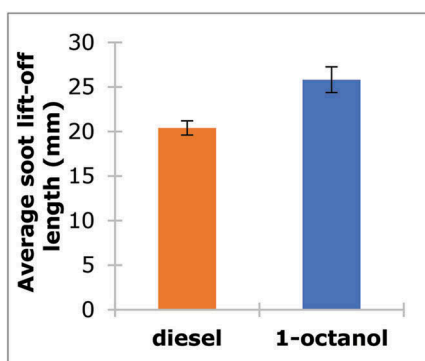


Figure 4. Soot lift-off length for both fuels averaged over the period where the sLOL was stable. This was done for the seven injections performed for each fuel and the error bars correspond to two SDs.

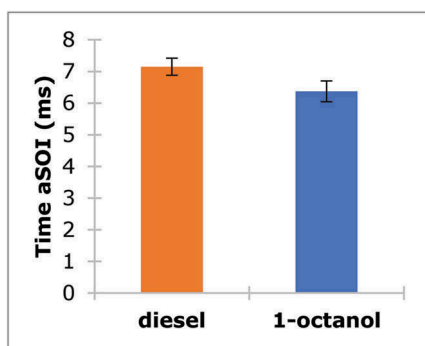


Figure 5. Point in time at which the last visible pocket of soot can be observed, referred to in the main body of the text as the end of (visible) combustion (EOC).

From Figure 4, it can be seen that for diesel, soot stabilizes on average ~20% closer to the injector tip than octanol. This indicates that spatially, the onset of soot formation is delayed for octanol to a more downstream location. It could be that either its lower

reactivity or its oxygen concentration (or a combination of both) offsets the formation of soot. In practical terms, this means that soot will have less space and time to grow beyond this location before oxidation starts to control the soot development processes. This could limit the development of larger soot particles, which would also make them easier to oxidise in engines.

From Figure 5, it can be seen that soot is consumed slightly earlier for octanol than for diesel. It is interesting to note that even though 1-octanol has a lower reactivity profile than diesel, its oxidation is clearly faster, as the last pocket of soot disappeared around 700 μ s earlier than for diesel. In an engine, this would mean that for the same amount of injected mass of fuel, due to its faster oxidation, less soot will remain “un-oxidised” when the exhaust valve opens. This can potentially reduce engine-out soot emissions as well as enhance the life of diesel particulate filters where applicable.

Though the same mass of fuel was considered in this work, as seen in §2.1, the density and energy between the fuels are also comparable and hence the outcome based on the same energy content may not be far from that presented in these results.

3.2 Spatial and temporal characterisation of the flame temperature

The spatial and temporal changes in temperature for diesel are shown in Figure 6, and those for octanol in Figure 7. For both graphs, each data point corresponds to the temperature value averaged over the detected flame, and subsequently ensemble averaged over the seven injections performed. The errors bars shown correspond to two standard deviations. The images above each graph correspond to one of the seven injections performed for each fuel, and are presented to illustrate the spatial temperature distribution throughout the flame as combustion proceeds. Each image in the sequence corresponds to one of the data points in the graphs, presented in chronological order.

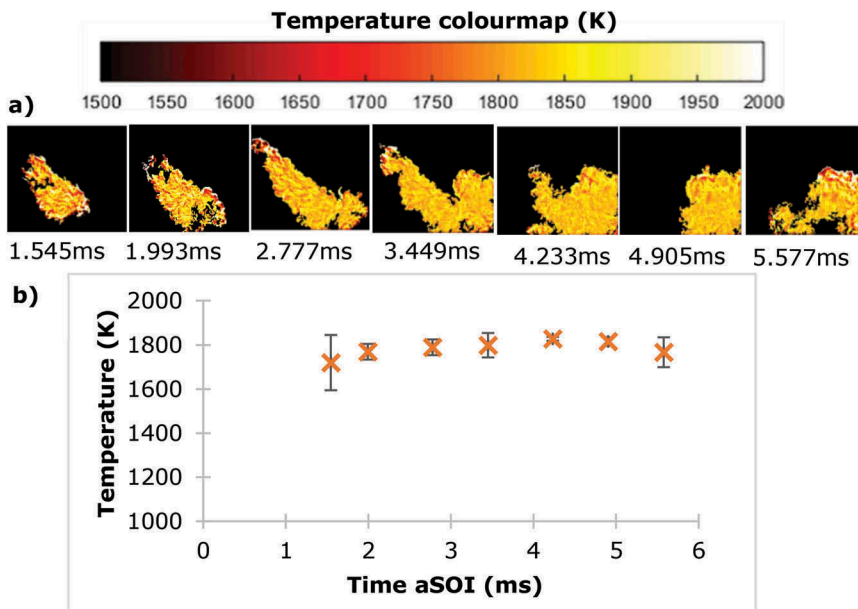


Figure 6. Temperature results obtained for diesel a) Spatial distribution of the temperature in the flame b) Temporal development of the temperature for points selected throughout the injection event.

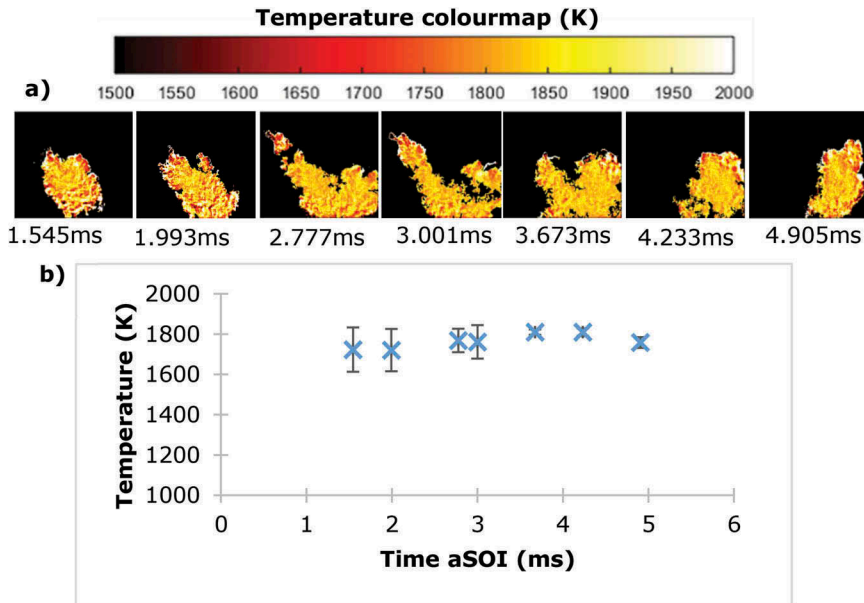


Figure 7. Temperature results obtained for octanol a) Spatial distribution of the temperature in the flame b) Temporal development of the temperature for points selected throughout the injection event.

For both fuels, the temperature was slightly lower at the SOC relative to the main combustion phase, and it then increased slightly and remained nearly constant until the combustion approached an end. Both fuels followed the expected combustion behaviour, that is, an increase in temperature during the SOC as reactions developed and then an almost constant temperature until the final reactants and products were consumed during the EOC. When looking at the average temperature, it remained between 1700K-1800K for both fuels. It could be that under the conditions studied, the similar LCV of both fuels produced this similar average flame temperature. Extrapolating these findings to an engine, this suggests that under the selected ambient conditions, octanol could produce a similar amount of thermal NO_x as that of diesel, provided the flame areas are similar. Nonetheless, this suggestion is based on the findings of this work performed in a CVC and further works on engines are needed to provide a quantitative assessment.

When looking at the spatial distribution of the flame temperature (Figures 6a and 7a), similar trends were identified for both fuels. At the SOC, there were flame regions with high and low temperature values due to the localised fuel-rich regions that had not had the time to mix and therefore burned at different temperatures depending on their mixture ratios. As combustion proceeded, these regions dissipated and the temperature distribution became more uniform except for some lower temperature patches at the tip of the flame and at the flame edges that were closer to the chamber wall. This was caused by heat transfer from the hot flame gases to the cooler CVC wall. As combustion approached an end, high temperature regions were observed at the tip of the flame, where more air was available for oxidation reactions to develop and consume the remaining pockets of fuel and soot.

3.3 Spatial and temporal characterisation of soot

The spatial and temporal distributions of soot (Figure 8 and Figure 9) have been presented in a similar manner to those for the temperature in the previous sub-section (Figure 6 and Figure 7). The KL factor represents the amount of soot as an averaged line-of-sight value. It is commonly presented in 2C pyrometry literature without units, and following this convention, it is presented as dimensionless in this work. The data points in the graphs represent averaged values over the flame and are themselves an ensemble average of the seven injections performed for each fuel. Each image also corresponds to a single shot of one of the combustion events, corresponding chronologically to the data points in the graphs.

As opposed to the similarities observed for the temperature in the previous subsection (Figure 6 and Figure 7), the soot distribution for diesel and octanol showed differences both spatially (Figure 8a and 9a) and temporally (Figure 8b and 9b). Octanol consistently showed lower KL factor values than diesel and achieved a significant reduction in peak soot of 67%. When looking at the temporal evolution of the curves for both fuels, the soot formation rate for octanol was slower, as it took longer than diesel to reach its peak KL factor value. Before the KL peak is reached, when the KL factor has a positive rate of change, soot formation dominates over oxidation. On the other hand, after the KL peak is reached and the KL factor has a negative rate of change, soot oxidation dominates over formation. The oxidation rate of octanol appeared slightly faster than that of diesel, partly because of its lower peak KL factor. However, from these observations, it seems that the effect of octanol on soot suppression is the major drive behind its soot reduction potential. Overall, this finding makes octanol an attractive drop-in fuel for engine applications because of its lower soot production rate that makes it easier to oxidise before the exhaust valve opens.

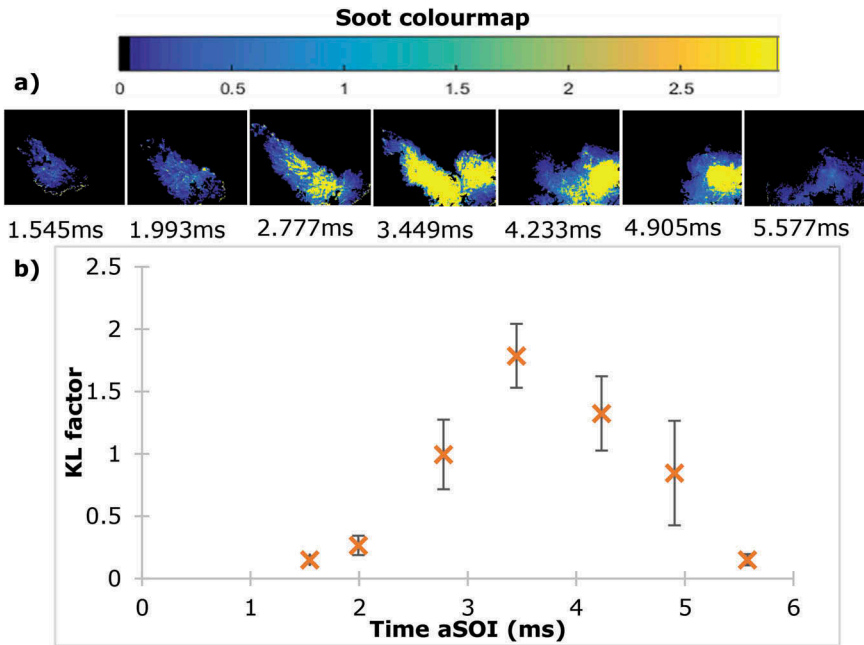


Figure 8. Soot (KL) results obtained for diesel a) Spatial distribution of the soot in the flame b) Temporal development of the soot for points selected throughout the injection event.

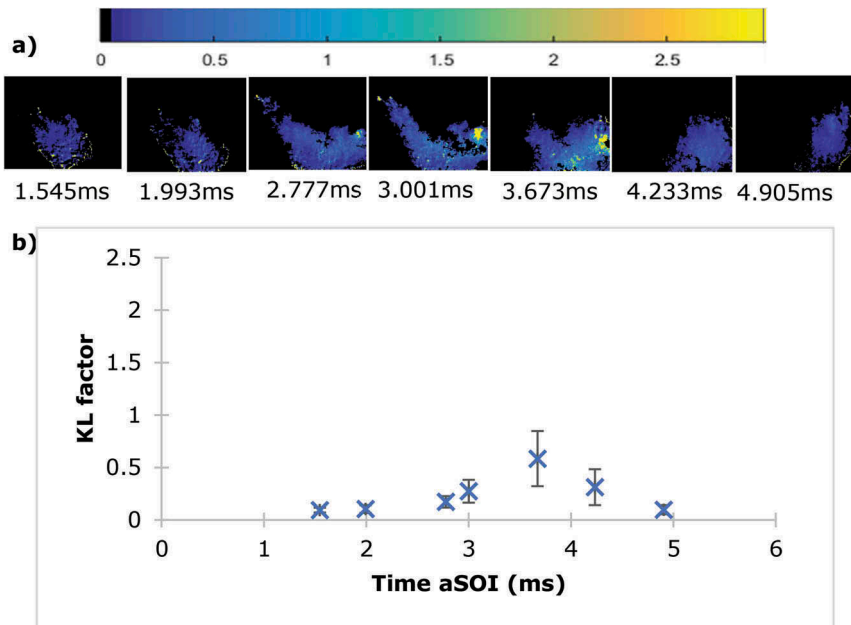


Figure 9. Soot (KL) results obtained for octanol a) Spatial distribution of the soot in the flame b) Temporal development of the soot for points selected throughout the injection event.

When looking at the soot spatial distribution images, the same conclusions can be drawn than those from the graphs: overall, octanol soots less. From the images, it can be seen that diesel shows higher KL values throughout the times considered, whereas octanol has lower KL values, which indicates a lower sooting tendency. Some interesting information could also be extracted when looking at how the soot develops spatially. That is, looking at where the high and low KL regions are in the flame and how they develop with time. For diesel, soot accumulated at the centre of the flame and towards the tip as combustion proceeded. For octanol, the higher sooting regions relative to its sooting propensity occurred -and remained- mostly at the tip. This suggests that the physio-chemical mechanisms causing a reduction in overall soot for octanol are also altering the way in which soot is distributed across the flame when compared to diesel flames. Even though at this stage it is unclear whether the soot reduction capabilities of octanol are due to the chemical effect of its hydroxyl group or due to dilution [22, 23], it is clear that octanol shows the potential to reduce soot emissions without a significant increase in flame temperature (i.e., without a significant effect on the thermal NO_x). More studies are currently being carried out by the authors to determine the extent of the effect of the hydroxyl moiety on soot reduction.

In line with the outcomes reported in this work, previous works on single-cylinder engines have also shown promising results regarding the potential of fuelling with octanol [10, 24, 25]. Under the conditions in their studies, they showed that octanol had a longer premixing time than diesel, and that this partly contributed to its low sooting levels. They have also shown a reduction in PM of up to a factor of five when fuelling with octanol, which indicates that depending on the load operation, soot suppression could be even higher than the one reported in this work.

Of interest for the manufacturers is the performance of higher carbon chain alcohols in multi-cylinder diesel engines. Whilst currently there is not much information available on the behaviour of octanol in multi-cylinder engines, in [26] they have shown that with alcohol additions of 30% to diesel and with the help of cetane improvers, a significant reduction in soot emissions is possible whilst keeping similar heat release and thermal efficiency profiles to those of diesel. Furthermore, in line with the preliminary material compatibility tests presented in §3.1, other works have not reported any major damages to their equipment when using octanol as a neat fuel or as a drop-in.

Whilst more research is required in CVCs, single, and multi-cylinder engines to understand the fundamentals behind the reduced sooting propensity of octanol, it is clear that from an industrial and application point of view, both its physical and combustion characteristics make it an attractive compound for future consideration as a drop-in fuel.

4 CONCLUSIONS

The combustion characteristics of octanol and diesel were studied by using high-speed two-colour pyrometry, which allowed to elucidate the spatial and temporal data for both soot and temperature at every flame location. The soaking test performed with octanol revealed no operational or physical alterations of the injection and pumping systems, but it was observed that octanol did cause a slight degree of swelling of the PVC hose.

The spatial and temporal changes of the temperature and soot in the spray flame were also analysed. Both fuels showed a similar average flame temperature throughout the combustion event as well as similar temperature distributions under the conditions studied. Nonetheless, large differences were observed between the fuels for the sooting propensity, with octanol consistently showing lower sooting values. Octanol also had a slower soot formation rate, which in an engine would facilitate its prompt oxidation. It was also found that for octanol, the soot only accumulated at the flame tip, whereas for diesel it was more evenly distributed throughout the flame. From our work, it can be concluded that octanol offers the potential to reduce engine-out soot emissions, which would also help reduce the load on the particulate filters.

ACKNOWLEDGEMENTS

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NOTATION

ε	Emissivity (-)
λ	Wavelength (nm)
2C	Two-colour
BEV	Battery electric vehicle
CN	Cetane number
CVC	Constant volume chamber
EGR	Exhaust gas recirculation
Fps	Frames per second
FWHM	Full width at half maximum
EOC	End of combustion
I_{soot}	Soot radiance (W/sr m ² nm)
I_{BB}	Blackbody radiance (W/sr m ² nm)
ICE	Internal combustion engine
KL	Soot optical thickness (-)
LCV	Lower calorific value (MJ/kg)
sLOL	Soot lift-off length
SOC	Start of combustion
SOI	Start of injection
T	Temperature (K)

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