



Study of soot formation during the combustion of Diesel, rapeseed methyl ester and their surrogates in turbulent spray flames

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HIGHLIGHTS

- ▶ The sooting behaviour of rapeseed methyl ester (RME) has been studied.
- ▶ The addition of RME to Diesel reduces the quantities of soot and soot precursors.
- ▶ Different RME surrogates (2 alkanes, 1 alkene and 1 ester) have been tested.
- ▶ The effects involved in soot reductions have been identified and quantified.
- ▶ Biodiesel soot are bigger than Diesel ones but oxidize faster.

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ABSTRACT

Effects induced by the use of rapeseed methyl ester (RME) as additive or Diesel substitute on the soot formation process have been studied in turbulent spray flames. Investigations have been carried out by coupling Laser-Induced Incandescence and Fluorescence (LII/LIF) at 1064, 532 and 266 nm. LII and LIF profiles obtained with fuels containing various amounts of ester (from 10 to 100 vol.%) showed that the addition of RME to a European low-sulphur Diesel or to a Diesel surrogate (a n-decane/1-methylnaphthalene blend derived from the 'IDEA' fuel) induces significant reductions of the quantities of soot and soot precursors (including high-number ring aromatic species and light soot precursors). The study of different RME surrogates (n-decane, n-hexadecane, 1-octadecene and methyl oleate) also revealed that the details of the oxidation of biodiesels could be mimicked only using large methyl esters as surrogates. N-alkanes and n-alkenes were found to be unable to reproduce the soot formation process occurring during the combustion of large fatty acid methyl esters (FAME) such as those contained in RME. The analysis of the correlation existing between the threshold soot index (TSI) and the peak soot volume fraction measured in flames of m-IDEA/RME blends containing up to 80% of ester allowed the identification of the different effects involved in the soot reduction (i.e. the dilution and the ester functional group effects). The estimation of their relative contribution has also been investigated. Finally, the LII fluence curves and time decays obtained at different heights above the burner in flames burning Diesel and a Diesel/RME mixture have been compared. By this way, it has been demonstrated that biodiesel soot are bigger than Diesel ones in the soot formation region. On the other hand, particles oxidize much faster when RME is added to Diesel.

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1. Introduction

The growing use of oxygenated biofuels appears to be an interesting means to reduce the energetic dependence on petroleum as a fuel source. In addition, biofuels also contribute to reduce net

emissions of greenhouse gases since these alternative fuels are derived from renewable sources [1,2]. Among the wide variety of alternative fuels currently studied, fatty acid methyl esters (FAME) also called biodiesels have received a particular attention since these monoalkyl esters of animal or vegetable derived long chain fatty acids are quite similar to conventional Diesel in its main characteristics [2,3]. FAME can thus be used pure and more generally in blends with diesel fuel in Compression Ignition engine with no major modifications in the engine hardware or performance [2–4]. Moreover, positive effects are globally associated to the addition

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of FAME to conventional Diesel in terms of pollutant emissions. This includes lower emissions of carbon monoxide (CO), unburned hydrocarbons and particulate matter (PM). Increases of NO_x concentrations in exhaust gases are generally reported on the other hand [1–5]. Many studies have thus been carried out during the past two decades to analyse and better understand the effects associated to the use of FAME on Diesel engines performance and emissions. Among the wide variety of works conducted in this field, the following literature survey will focus on studies that deal with the impact of methyl esters on the formation of regulated pollutants. It will especially focus on the formation of PM and hence soot which are generally the main constituents of Diesel exhaust particulates.

1.1. Literature survey

In a 1996 paper, Schmidt and Van Gerpen reported significant reductions of PM and hydrocarbon emissions at the exhaust of a Diesel engine when adding up to 50% of various pure esters to a standard diesel fuel [6]. Additional experiments were performed by adding octadecane (a long chain paraffin that does not contain oxygen) to Diesel in order to investigate effects of cetane number, long chain hydrocarbons and oxygen on emission levels. The obtained results lead to the conclusion that the particulate reducing effect of biodiesels was due to a combination of their oxygen content and to the displacement of aromatic species with long-chain esters (a phenomenon called “dilution effect” [7]). The same year, Krahl et al. summarized the published emissions measurements issued from different experimental studies carried out at the exhaust of Diesel engines fuelled by rapeseed methyl ester (RME) [8]. Significant reductions of the emissions of hydrocarbons (–20% to –30%), CO (–10% to –30%), PM (–20% to –40%) and aromatic compounds (up to –40%) were thus reported. Reductions of PM (–20% to –53%) and CO (–2.4% to –48%) were also observed by McCormick et al. during a series of engine tests based on the use of various biodiesels and esters produced from pure fatty acids [9–11]. In 1997, Choi et al. lead experiments in a Direct Injection Diesel engine fuelled with blends composed of 80 vol.% Diesel and 20 vol.% ester or octadecene (a good representative of the soyate hydrocarbon composition with one double bond but without oxygen) [12]. By this way, the authors wanted to isolate the effect of oxygenates on the reduction of emissions such as PM. The obtained results showed that the 20% biodiesel blend reduced PM emissions an additional 36% versus the 20% octadecene blend. The authors thus concluded that the presence of oxygen in the ester was the main factor inducing the reduction of PM as opposed to the dilution of aromatics and sulphur in the baseline diesel fuel. This result contrasts somewhat with the conclusions from Schmidt and Van Gerpen who adopted a similar experimental strategy using an alkane as additive as seen before [6]. In 2002, the US Environmental Protection Agency (EPA) published a report including results issued from 39 experimental studies conducted with heavy-duty engines [13]. The processing of all the data presented in this reference report lead to correlate the concentration of biodiesel added in conventional diesel with changes in regulated pollutants. The obtained results revealed that the use of biodiesel generates significant reductions of PM (up to 47% when using pure biodiesel), hydrocarbons (up to 67%) and CO (up to 48%) while slightly increasing NO_x emissions (up to 10%). Many works have then confirmed these trends with reduction of pollutant emissions sometimes far more important [14–17]. Knothe et al. especially compared the regulated exhaust emissions issued from a Diesel engine fuelled by three fatty acid methyl esters (methyl laurate, methyl palmitate and methyl oleate), biodiesel, low sulphur petrodiesel and alkanes (dodecane and hexadecane) [17]. They concluded that the use of a commercial biodiesel as well as pure fatty acid methyl esters sig-

nificantly reduces PM emissions (from 75% to 83%) compared to petrodiesel. The tested alkanes achieved lower PM reductions (from 45% to 50%) suggesting that the methyl ester moiety influences PM emissions considerably more than neat straight-chain hydrocarbons. Szybist et al. then conducted motored engine experiments and noted that significant amounts of CO₂ were produced during the low-temperature heat release part of the ignition process when using an ester (the methyl decanoate) instead of Diesel. The authors argued that this CO₂ production was the result of the decarboxylation of the ester group and not the product of an oxidation process [18]. This phenomenon also pointed out by Westbrook et al. in a previous modelling work is of interest as it eliminates C atoms from the product pool of species that can subsequently make soot in fuel-rich Diesel ignition zone [19]. On the other hand, Zhang et al. showed that this early CO₂ production could be significantly influenced by the presence and the position of double bounds in the aliphatic chain of fatty acid esters as observed in the case of various C₉ FAME [20]. In an extensive literature survey devoted to the analysis of the effect of biodiesels on Diesel engine emissions, Lapuerta et al. noted that a global consensus was found on the main trends presented in the EPA report even if a wide disparity of results was observed due to the number of different engine technologies used or to the operating conditions varying from test to test [21]. Based on the analysis of numerous experimental works, the authors concluded that the addition of FAME to Diesel induces significant reductions of PM emissions with a global decrease of the size of soot particles. Presence of oxygen as well as absence of aromatic compounds in biodiesels have been pointed out as being the main reasons explaining this trend. The authors explained that the oxygen contained in the ester molecule enables a more complete combustion even in regions of the combustion chamber with fuel-rich diffusion flames. This explains the observed reductions of hydrocarbons, carbon monoxide and PM emissions. Lapuerta et al. also argued that the oxygen content of biodiesels promotes the oxidation of soot. This process that is well correlated with the local flame temperature [22] would be influenced by modifications in the structure of biodiesel soot as observed in various works from Boehman et al. [23,24,26] and Jung et al. [25]. Finally, the authors noted that the concentrations of aromatic compounds such as Polycyclic Aromatic Hydrocarbons (PAHs) in exhaust gases were generally lower during the combustion of biodiesel. This observation is of interest since these species are directly involved in the soot formation process. Since then, many recent engine-based experimental works have confirmed these trends especially in the case of RME [27–29]. Nevertheless, there are a multitude of factors that contribute to a wide variability in the obtained results during engine studies [21]. Moreover, experiments conducted in engines do not allow a direct analysis of pollutants formation mechanisms and kinetics as pointed out in a recent paper from Maricq [30]. Well defined and reproducible laboratory scale experiments have thus been conducted to better understand fundamental effects induced by the use of esters on the formation mechanisms of pollutants such as soot.

In a computational study from 2000, Fisher et al. developed detailed chemical kinetic models for the combustion of two small methyl esters (methyl butanoate and methyl formate). These models have been tested against data obtained in static vessels. They were found to overestimate the overall reactivity of the two esters prompting demand for more complete and well-characterized experimental databases to test the developed kinetic codes thoroughly [31]. In 2007, Dagaut et al. proposed for the first time an experimental study of the oxidation behaviour of RME in jet-stirred reactor (JSR) [32]. The oxidation of the long saturated alkyl chains present in RME was found to be very similar to the oxidation of large alkanes. The JSR results obtained with RME were thus compared with the predictions issued from a kinetic scheme

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