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Full Length Article

Effect of propylene glycol ether fuelling on the different physico-chemical properties of the emitted particulate matters: Implications of the soot reactivity

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ABSTRACT

The research work presented here focuses on the particulate matter (PM) number concentration and its physicochemical properties from the combustion of di and tri-propylene glycol methyl ether/diesel blends (D20 and T20 respectively). Exhaust PM were characterised using a Scanning Transmission electron microscopy equipped with energy dispersive X-ray spectroscopy (S/TEM-EDX) to quantify the nanostructure, morphology and elemental composition of the agglomerates, Raman spectroscopy (RS) to further examine the particulate graphite like structure and thermo-gravimetric analyser (TGA) to analyse the oxidative reactivity. The increase in the fuel oxygen content reduces both, exhaust PM levels and NOx emissions. TGA analysis confirms that the oxygenated blends enhance the particulates oxidative reactivity and increase their volatile fraction in the following order T20 > D20 > Diesel. EDX analysis shows that the combustion of both D20 and T20 lowers the PM carbon fraction and ash precipitations but increases its oxygen functional groups in the same order. Furthermore, a notable reduction in the primary particles size was recorded, whose carbon layers were found to be more tortuous than diesel, but no significant modification was shown in their length. Unconventionally, smaller separation distance was seen between the carbon layers, and higher graphitisation order was seen from the RS analysis. It was finally concluded that concerning the nanoscale parameters, the initial curvature of the carbon layers present a stronger influence in dictating the particles reactivity compared to the graphitisation order or the initial length and separation distance of the carbon layers. As for the macroscale, primary particle size and the portion of oxygen in the particle could be another possible reason for the better soot reactivity seen from the propylene glycol ethers combustion. The significance of the reduced exhaust particles' level and modified PM's physical and elemental properties can improve Diesel Particulate Filter (DPF) regeneration and allows engine calibration that can favour NOx emissions reduction (i.e. lower NOx - PM trade-off lines).

1. Introduction

Particulate Matter (PM) emissions from modern vehicles, including diesel and gasoline direct injection (GDI), consist of carbonaceous materials (soot) and various types of inorganic and organic substances, some being mutagenic and carcinogenic by nature [1]. It is commonly accepted that oxygen-borne fuels blended with diesel can effectively reduce the PM levels and other unregulated emissions such as unburned hydrocarbons and carbon oxides [2]. They also play an important role in enhancing the oxidation reactivity of the soot particles (ability to oxidise) [3], leading to improved particulate filters function and durability by lowering the regeneration temperatures and minimising the intensity of the soot burn-off cycles [3].

If the biodiesel production and utilisation is increased to cover the energy demand in transportation, then the associated increase of the glycerol volumes (main by-product from biodiesel production) may create ecological hazards, since they cannot be safely disposed [4]. One possible application to benefit from these products could be to synthesise propylene oxides from the resulted glycerol, following the process described by Yu et al [5]. Afterwards, propylene glycol ethers could be produced by the reaction of propylene oxides with methanol [6]. Similar to biodiesel, glycol ethers are known for their suitability with compression ignition engines in terms of safety, accessibility, price and compatibility with diesel fuel [7]. Their atomic structure contains both ether and alcohol moieties and generally characterised with a high oxygen content [8].

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Tri-propylene glycol methyl ether (TPGME) was nominated by Natarajan et al. [9] and also suggested by Gonzalez et al. [2] as a potential oxygenate to be blended with diesel among 71 candidates, taking into account several factors such as exhaust emissions reduction, blend toxicity, lubricity and cetane number improvement. Following Gonzalez's work, both Mueller et al. [10] and Burke et al. [6] incorporate numerical simulations along with chemical kinetic modeling to examine the effect of TPGME molecular structure on the ignition mechanism and the soot formation process. As a result, TPGME was shown to effectively reduce the soot precursors inception, especially acetylene, and that was directly linked to its optimal atomic structure where all the oxygen atoms are available for soot reduction. Dumitrescu et al. [11] studied the ignition delay and flame lift-off length produced by TPGME/Diesel blend (50% vol.) and confirmed a reduction in soot formation and/or increase in soot oxidation. Another glycol ether candidate is the di-propylene glycol methyl ether (DPGME), with similar chemical structure as TPGME but present shorter carbon chain length and one less oxygen atom, resulting in approximately the same oxygen content. Natarajan et al. [9] nominates DPGME as a viable candidate for mixing with diesel in terms of PM reduction and blend stability. Gomez-Cuenca et al. [8] reported that DPGME/diesel fuel blends enhanced the mixture cetane number and reduced CO, while no clear trend was seen in production of THC, NOx and PM emissions. Hilden et al. [12] reported that both DPGME and TPGME diesel blends, with 6.5 wt% oxygen content, present comparable PM reduction capability, while other authors [9] found that under the same oxygen content of 7 wt%, TPGME/diesel blends are more efficient in reducing PM emissions than DPGME blends.

Although propylene glycol ethers are proved to be suitable for mixing with diesel and can reduce the PM discharges, there's still no study highlighting their effect on the resulted PM characteristics. The PM physical and chemical properties, such as primary particle size, carbon layer structure, oxygen functional groups and the corresponding ash content are reported to guide the particles reactivity [13]. Despite the extensive analysis conducted to understand the correlation between these variables and the oxidative behaviour of the particles, this relationship is still quite complex and not fully understood in the literature [13–15]. The current paper aims to obtain an improved understanding of the level at which DPGME and TPGME diesel blends (20% vol.) will influence the PM oxidative reactivity, and determine the effect of the different physico-chemical properties, such as morphological parameters, nanostructure characteristics and elemental composition, on the corresponding reactivity.

2. Experimental set up and methodology

2.1. Engine test and combustion analysis

The experiments have been conducted in a four-stroke research diesel engine featured with a common rail fuel injection system that permits the control of multiple injection strategies. The main specifications of the test engine can be found in Table 1. All the tests were performed at a constant speed of 1500 rpm and fix load of 4 bar IMEP. The fuel was injected at a stable pressure of 600 bars and split between pilot and main-injections, with timings of 15 and 8 CAD BTDC respectively. In-cylinder pressure was recorded over 200 cycles using an AVL GH13P pressure sensor mounted in the cylinder head and the signal was amplified by an AVL FlexiFEM 2P2 Amplifier. The corresponding Digital shaft encoder producing 360 pulses per revolution was used to measure the crank shaft position [16]. Heat release rate (HRR) was integrated from the pressure data collected by designing a simple model neglecting the heat losses differences between the fuels tested. Exhaust temperatures from the combustion of each fuel blend are recorded with the use of K-type thermocouples (with a range of 0-1250 °C and an accuracy of \pm 2.2 °C) located directly at the exhaust port.

Table 1

Diesel engine	specifications.
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Engine Parameters	Specifications
Engine type Stroke type Cylinder bore x stroke (mm) Connecting rod length (mm) Compression ratio Displacement (cc)	Diesel 1-cylinder Four-stroke 84 × 90 160 16:01 499
Ingine speed range (rpm) IMEP range (bar) Common Rail fuel injection pressure range (bar) Number of injections	 900–3000 < 7 500–2000 3 injection events

2.2. Fuel used

Ultra-low sulphur diesel (ULSD) was supplied from Shell Global Solutions UK, DPGME and TPGME were purchased from Sigma Aldrich. Glycol ether diesel blends, so called D20 and T20 in this analysis, were prepared by mixing diesel with each of the candidates (DPGME and TPGME respectively) separately to form 6.5% wt. oxygen content. The properties of the oxygenated hydrocarbons and the blends are obtained from the literature and presented in Table 2.

2.3. Gaseous emissions analysis and particle number concentration

Multi-Gas 2030 FTIR spectrometry-based analyser was utilised to measure the gaseous emission from the exhaust such as nitrogen oxides (NOx), carbon oxides (CO-CO₂) and total hydrocarbons (THC).

Electrical mobility particle size distribution (dp) and the total particle number concentration were measured using a TSI scanning mobility particle sizer which includes a 3080-electrostatic classifier, 3081-Differential Mobility Analyser and a 3775-Condensation Particle Counter [22]. The exhaust dilution ratio was set to 1:11 using an ejector diluter system before testing the sample.

2.4. PM collection method

Tissue quartz (TQ) filters (type TISSUEQTZ 2500 QAT-up, 47 mm diameter) supplied from VWR International Ltd UK were applied to collect the needed PM samples. The filters were pre-heated at 650 °C for 3 h before starting the collection process. Afterwards, they were placed in an inline filter holder connected to a partial exhaust line (vacuumpumped assisted, as shown in Fig. 1) and loaded to a target mass of 3 mg, following the method proposed by Mühlbauer et al. [13]. The samples collection point was placed before the Diesel Oxidation Catalyst (DOC) to maintain the particles volatile fraction and prevent any possible low-temperature PM oxidation that may be caused by the increased amounts of NO_2 produced downstream the catalyst. This type of reactions may alter the particles structural order and affect their oxidative reactivity [23].

2.5. PM characterisation techniques

A schematic diagram presenting the production process of the tested oxygenated hydrocarbons (from literature) and the experimental procedure used for the soot collection and the different methods used to analyse the particles physico-chemical properties is plotted in Fig. 1.

2.5.1. TGA analysis

The TQ-filters were cut into small sections and used for the TGA testing (PerkinElmer) in a way that an equal PM mass of ~ 0.12 mg was examined in all the experiments [24]. The sample was first heated under an inert atmosphere of nitrogen at a constant flow of 40 ml/min with 10 °C/min ramp to reach 400 °C. The PM sample was maintained

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