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Gasoline direct injection engine soot oxidation: Fundamentals and determination of kinetic parameters

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ABSTRACT

Current emissions legislation for road transport vehicles, including modern gasoline vehicle fleet limits the mass and the number of Particulate Matter (PM) emitted per kilometre. The introduction of a gasoline particulate filter (GPF) is expected to be necessary, as was the case for diesel vehicles, the traditionally recognised source of PM in transportation. Therefore, for the design of efficient GPFs and the regeneration strategies, soot oxidation characteristics in gasoline must be understood.

Extensive research has been carried out mainly to investigate the oxidation of diesel soot, however, in the most cases soot were collected on microfiber filters and the activation energy was calculated with the logarithm method assuming mass and oxygen reaction orders equal to one. Identified limitations that lead to inconsistent and inaccurate trends and results are presented in this paper. As a consequence, a novel methodology to accurately obtain the oxidation kinetic parameters for soot emitted from a Gasoline Direct Injection (GDI) engine has been developed and presented in this paper. The particles collected in a silicon carbide wall-flow particulate filter are directly exposed to oxidation conditions in a thermogravimetric analysis (TGA) without the use of microfiber filter.

The significance of more accurate and consistent calculations of soot oxidation kinetic parameters as a result of this methodology will aid modelling and experimental work of the aftertreatment systems and will lead in improving the GPF regeneration process in modern GDI vehicles. Avoiding high peak temperatures during regeneration and large thermal stress gradients and thus increasing the operating life of the filters is amongst the benefits can be seen.

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Definitions, Acronyms, Abbreviations

A'	Exponential prefactor defined as $A' = A \cdot p_{O_2}^r$
A	Exponential prefactor
BMEP	Brake mean effective pressure
bTDC	Before top dead centre
CAD	Crank angle degree
DMF	2,5-dimethylfuran
E_a	Activation energy
EGR	Exhaust gas recirculation
Exp	Experimental
GDI	Gasoline direct injection
GPF	Gasoline particulate filter
IMEP	Indicated mean effective pressure
m	Sample mass
m_{exp}	Mass obtained experimentally
MMLRT	Maximum mass loss rate temperature
m_{model}	Mass obtained with Arrhenius-like equation
n	Mass reaction order
NEDC	New European driving cycle
p_{O_2}	Oxygen partial pressure
PAHs	Polyaromatic hydrocarbons
PFI	Port fuel injection
PM	Particulate matter
r	Oxygen reaction order
R	Universal gas constant
REGR	Reformate exhaust gas recirculation
T	Temperature
t	Time
TEM	Transmission electron microscope
t_f	Number of points

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TGA Thermogravimetric analysis
TWC Three way catalyst
VOC Volatile organic compounds

1. Introduction

Particulate matter (PM) emissions are composed of a carbonaceous core, known as soot, onto which different hydrocarbon species, especially polyaromatic hydrocarbons (PAHs), can be adsorbed. Internal combustion engines are the main source of PM. Although the introduction of Gasoline Direct Injection (GDI) has proven to be an efficient technology capable of significantly reducing fuel consumption and CO_2 emissions from road transport vehicles, its wider implementation has also raised interest due to the increase in PM emissions [1,2].

In general the effect of PM in the environment and on human health is diverse; it is reported to cause building soiling and reduced visibility after high PM level episodes [3]. Moreover, PM can penetrate into the human body through the respiratory system causing asthma, bronchitis or exacerbating allergies [4]. From the lungs, PM can spread to different organs, such as the liver or brain, through the blood stream [5]. PM can block the arteries increasing stroke risk and provoking cardiovascular- related complications [6]. As the awareness of the deleterious effects of PM is increasing, stricter limitations are being imposed to gasoline and diesel powered road vehicle, including the Euro 6c which will limit PM levels to 6×10^{11} particles per kilometre.

One of the most widely used techniques to obtain oxidation rate parameters is thermogravimetric analysis (TGA). In diesel engines, extensive efforts have been made to characterise the kinetic parameters of soot in both non-catalysed [7] and catalysed samples [8,9]. Understanding and modelling the soot oxidation processes can provide useful data for automotive engineers to design optimised strategies for filter regeneration, saving fuel and extending the catalyst's life, as well as reducing local temperature peaks in the catalyst. Studies performed in diesel engines shown that the particle oxidation characteristics can be influenced by the particle morphology (i.e. primary particle size, fractal dimension), particle nanostructure (size of the graphene layers and curvature) as well as particle composition [10–12]. However, in GDI engines this knowledge is still not well established and the different local in-cylinder conditions might result in different particle characteristics. TGA has been used to quantify the composition of PM: volatile organic compounds (VOC)/soot ratio and ash percentage [13,14], and to prepare the PM sample for subsequent studies [14,15]. For instance in [15], soot samples were partially oxidised under 8% oxygen and 1000 and 2000 ppm NO_2 in 8% oxygen to examine the physicochemical properties of soot during the oxidation process through Transmission Electron Microscope (TEM).

There are a limited number of studies on soot oxidation kinetics in the literature to date. These studies mainly focus on the effects of fuel and engine operating condition on activation energy and reaction order for soot oxidation. Luo et al. [16] analysed the activation energy for soot samples derived from gasoline, E10 and E20 blends under four different operating engine conditions using the Arrhenius-like equation logarithm method and assuming first reaction order for mass and oxygen. The activation energy for gasoline soot was found to lie between 197 and 256 kJ mol^{-1} whereas for ethanol blends soot, higher oxidative reactivity was reported. E10 activation energy was in the range of 183–221 kJ mol^{-1} and E20 from 163 to 189 kJ mol^{-1}. Higher activation energies and higher oxidation temperatures were found for higher engine loads that the authors hypothesised were on account of the greater ordered structures. Wang et al. [17] analysed the PM composition and soot characteristics under two different engine conditions using a single cylinder engine fuelled with gasoline, ethanol and 2,5-dimethylfuran (DMF). The effect of the heating ramp and sample load was analysed in order to develop a TGA methodology for GDI soot. The kinetic parameters were obtained using the logarithm method, again assuming one for both reaction orders. The activation energy found for gasoline soot was 131 and 153 kJ mol^{-1} at 1500 rpm, 5.5 and 8.5 bar indicated mean effective pressure (IMEP) respectively. Ethanol and DMF were also reported to reduce the activation energy of soot oxidation when compared to gasoline. Due to the low oxygen concentration present in the gasoline exhaust, it is important to estimate the oxygen reaction order to predict the soot oxidation behaviour in actual gasoline exhaust environment. In the majority of the investigations carried out for diesel soot, reaction order one for both oxygen and mass has been assumed, while for GDI engines, several investigations have estimated the reaction orders in order to separate the effect of mass and oxygen concentration. Wang-Hansen et al. [18] analysed the reactivity and kinetics of particles emitted by diesel, gasoline Port Fuel Injection (PFI) and GDI soot in comparison to a Printex U reference. Temperature programmed oxidation and isothermal experiments were carried out under different concentrations of oxygen and NO_2. The reported activation energy for GDI soot was 146 kJ mol^{-1} and similar to soot from PFI engines [9], and both were higher than diesel soot. On the other hand, ethanol soot was reported to be more reactive. Choi et al. [19] calculated the activation energy, prefactor and mass reaction order of GDI soot samples under five different engine conditions following the Arrhenius-like equation logarithm method. The activation energy ranged from 125 to 142 kJ mol^{-1}, the prefactor from 13,500 and 127,000 s^{-1} and the mass reaction order from 0.512 to 1.011. The authors in [19] did not find any correlation between the engine operating condition and the kinetic parameters while a catalytic effect of the ash was observed in the GDI soot oxidation process.

A summary of the kinetic parameters reported in the literature is presented in Table 1. The activation energy of gasoline soot lies in the wide range between 125 and 256 kJ mol^{-1} depending on engine operation condition, fuel formulation, soot collection and soot reactivity method. This is a similar activation energy window to that reported for diesel soot in the literature, 100–300 kJ mol^{-1} [20]. There is an agreement in the literature that the microstructure of soot emitted from gasoline engines is less ordered than the microstructure of diesel soot [14], whereas the effects of engine speed and load are not fully understood yet [19]. It is also reported that soot produced under ethanol combustion has higher soot reactivity compared to gasoline soot. The authors claimed that ethanol enhanced the oxidation activity due to i) smaller particle size and ii) more disordered microstructure [16]. It has also to be noted, the lower concentration of oxygen in a gasoline exhaust could become a challenge for particulate filter regeneration, thus further analysis is needed to understand GDI soot characteristics.

The aim of this research work is to develop a new robust methodology in order to obtain accurate and consistent information of the GDI soot oxidation parameters such as activation energy, prefactor, mass reaction order and oxygen reaction order. The first section of the paper studies the capabilities and limitations of the state of the art particle collection methods where microfiber filters are used to collect particles under different engine exhaust conditions. Based on the identified limitations of the methodologies in the first section of the paper in the second section particles are collected in a vehicle exhaust particulate filter to better reproduce the oxidation process in the actual after treatment system, and were then removed for direct exposure to the TGA analysis. Soot oxidation kinetic parameters are estimated using the Arrhenius-like equation and assessed and compared to the proposed methodology using isothermal processes instead of heating ramps. The proposed particles collection method and methodology enable to characterise the gasoline soot oxidation parameters

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