

Reformate Exhaust Gas Recirculation (REGR) Effect on Particulate Matter (PM), Soot Oxidation and Three Way Catalyst (TWC) Performance in Gasoline Direct Injection (GDI) Engines

Maria Bogarra-Macias and Jose M Martin Herreros-Arellano
Univ. of Birmingham

Athanasios Tsolakis
Univ. of Birmingham

Andrew P.E. York and Paul Millington
Johnson Matthey Technology Centre

ABSTRACT

Gasoline direct injection (GDI) engines have become very attractive in transportation due to several benefits over preceding engine technologies. However, GDI engines are associated with higher levels of particulate matter (PM) emissions, which is a major concern for human health.

The aim of this work is to broaden the understanding of the effect of hydrogen combustion and the influence of the three way catalytic converter (TWC) on PM emission characteristics. The presence of hydrogen in GDI engines has been reported to reduce fuel consumption and improve the combustion process, making it possible to induce higher rates of EGR. A prototype exhaust fuel reformer build for on-board vehicle hydrogen-rich gas (reformate) production has been integrated within the engine operation and studied in this work. It is concluded that benefits on engine out soot emissions from the combustion of the reformate-gas are more noticeable in the engine operation conditions with a higher concentration of soot particulates.

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INTRODUCTION

The necessity for cleaner technologies that can also reduce anthropogenic greenhouse gas emissions in the atmosphere by improving fuel economy, has made researchers and vehicle manufacturers focus their attention on more advanced engine concepts, and this includes the development of gasoline direct injection engines (GDI). The potential for higher power and improved fuel economy through boosting and downsizing makes GDI technology an attractive solution for the near future [1, 2, 3, 4].

Despite these technological advantages, GDI engines are widely reported to have increased particulate matter (PM) emissions [5, 6, 7] when compared with Port Fuel Injection (PFI) engines. Until recently, compression ignition engines have been the major source of PM. Thus, the harmful effects of particles in the environment and on human health are well known and have been extensively researched. It has been proven that polycyclic aromatic hydrocarbons (PAHs), which act as soot precursors, can be adsorbed in the carbonaceous core (soot) of particles, presenting mutagenic behaviour that can alter

DNA leading to higher rates of cancer [8]. Furthermore, the inhalation of particulates is associated with the worsening of asthma and allergy-related diseases [9]. It is reported that PM emitted by GDI engines tends to be smaller than those emitted by diesel engines, allowing them to penetrate deeper into the respiratory system and therefore, presenting greater risk to human health [8-9]. Thus, the increasingly restrictive legislation on emissions has been introduced to control PM number in addition to PM mass. For instance, from September 2014, EURO 6 legislation came into force, with a strict limit on PM number from 2017 (Euro 6c) of 6×10^{11} #/km for GDI engines [10].

The possible advantages from the combustion of hydrogen rich gas which is produced in an on-board catalytic fuel reformer, on PM reduction are explored in this work. The improved mixing process, because of the high diffusion coefficient of hydrogen, the more complete combustion on account of the high flame speed and smaller quenching distance, can reduce both PM mass and number [11]. Furthermore, the presence of H_2 promotes OH production that can also slow down the rate of soot formation [12] and enhance the rate

of soot oxidation [13]. On the other hand, hydrogen could increase the in-cylinder temperature, which enhances PM production and PM oxidation [13] thus the trade-off between PM production-PM oxidation is also studied. The reforming process captures waste energy from the engine exhaust and generates hydrogen rich gas (reformate). The combustion of the reformate in GDI engines can also provide combustion stability improvements and gaseous emission reduction [12]. Thus, the aim of this paper is to understand which of the aforementioned effects will dominate during the reformate combustion.

The influence of the reformate on the PM emission characteristics formed from the combustion of the main fuel (i.e. gasoline DI) depends on the PM nature (composition, size, morphology and structure). Also, the combustion inside the cylinder is modified with the presence of reformate gas. Therefore, the PM emission characteristics are also changed. To further understand the influence of reformate on the different nature PMs, two different gasoline injection timings were selected to obtain: i) PM composed of a high proportion of organic components and ii) PM composed mainly of soot. Particulate size distribution (PSD) was investigated before and after the TWC for noEGR, EGR and REGR in order to study its influence. PSD was also measured for engine operation with the reformate products stream to analyse any possible soot formation during the reforming process. Finally, soot oxidation studies through a thermo-gravimetric method were carried out for noEGR, EGR and REGR conditions.

EXPERIMENTAL SETUP

A 2 L, four-cylinder air-guided GDI engine with dual scroll turbocharger, side-mounted solenoid injectors and a centrally located spark plug is used for this study. Further details of the engine specification are listed in Table 1. Standard EN228 gasoline with 5% (v/v) ethanol content provided by Shell has been used for this research. Fuel properties can be found in Table 2.

Table 1. Engine specifications.

Compression ratio	10:1
Bore x stroke	87.5 x 83.1 mm
Turbocharger	Borg Warner k03
Rated power	149 kW at 6000 rpm
Rated torque	300 Nm at 1750-4500 rpm
Engine management	Bosch ME17

The aftertreatment system of the engine is composed of a commercial three-way catalyst (TWC) which meets the Euro6 legislation. The fuel reformer is positioned after the TWC. The reformer was designed by Johnson Matthey and it consists of five metallic plates coated with Pt - Rh based catalyst, offering high sulphur tolerance, excellent thermal stability under the conditions studied and high resistance to coke formation. The reformer recovers heat from the exhaust stream to promote chemical reactions [14,15], for example the conversion of HCs in a hydrogen-rich gas. Gasoline is injected in a bypass of the

main exhaust stream, which is not treated by the TWC. In the reformer, heat from the main exhaust is exchanged to the gasoline-exhaust flow without mass contact. A schematic of the engine reformer integrated system is shown in Figure 1 together with the experimental set up.

Table 2. Gasoline properties.

Analysis (Test method)	Result
Density at 15°C (kg/m ³)	743.9
IBP (°C)	34.6
20% v/v (°C)	55.8
50% v/v (°C)	94.0
FBP (°C)	186.3
C m/m %	84.16
H m/m %	13.48
O m/m %	2.36
Paraffins (% vol)	43.9
Olefins (% vol)	11.7
Naphthenes (% vol)	7.8
Aromatics (% vol)	26.9
Oxygenates (% vol)	7.7
Sulfur (RFA, ppm) (mg/kg)	6
Calorific values (MJ/kg)	42.22
MON	85.3
RON	96.5

For the continuous measurement of gaseous emissions a Fourier Transform Infrared Spectroscopy (FTIR) 2100 MKS was used. Legislated emissions (CO, CO₂, NO_x and THC) were sampled before and after the TWC, as well as the reformate products downstream the on-board reformer. The sample was filtered to avoid any damage of the optical part by PM and pumped at a rate of 1L/min through the equipment. The heating lines and the pump were maintained at 191°C to eliminate any condensation of HCs and water in the pipes.

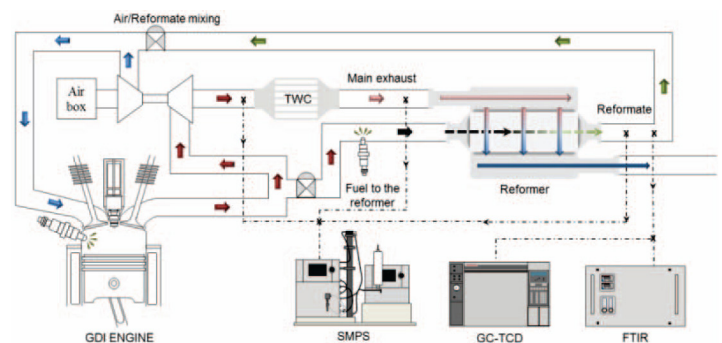


Figure 1. Schematic of the experimental set-up

A TSI scanning mobility particle sizer (SMPS) composed by a series 3080 electrostatic classifier, a 3081 Differential Mobility Analyser (DMA) and a 3775 Condensation Particle Counter (CPC) was used for the measurement of particles size distribution. The sample and sheath flow rates were set to provide a particle size distribution range between 7.5 and 294 nm. The dilution ratio (DR) was set at 11 using an ejector diluter system to precondition the sample for the SMPS measurement. The DR was continuously measured based on NO and CO₂ concentration on the exhaust and diluted streams, equation 1,

using the FTIR. Samples were taken before and after the TWC to evaluate its effectiveness in the removal of volatile organic compounds (VOC).

$$DR = \frac{\text{ppmNO}_{\text{rawexhaust}}}{\text{ppmNO}_{\text{dilutedsample}}} \approx \frac{\% \text{CO}_2_{\text{rawexhaust}}}{\% \text{CO}_2_{\text{dilutedsample}}} \quad [1]$$

Hydrogen production from the on-board reforming process was measured using a HP 5890 Series 2 gas chromatograph (GC) coupled with a thermal conductivity detector (GC-TCD) and HP 3395 integrator. The carrier gas was argon and the detector was calibrated with 30% hydrogen in nitrogen. The hydrogen production downstream the reformer and the hydrogen after-combustion are measured.

A schematic of the experimental setup can be found in Figure 1. The dotted lines represent the sample points for the SMPS, GC-hydrogen and FTIR respectively. For the soot oxidation behaviour analysis, a Perkin Elmer thermogravimetric analyser (TGA) model Pyris was employed. Whatman GF/F Glass microfiber $\Phi 47$ mm filters were pre-heated at 600°C during 6 hours to limit any influence of filter material losses during the thermogravimetric analysis. Immediately after this pre-treatment, the filters were loaded for one hour. In order to avoid any loss of PM during the sampling process, instead of scratching the filters, a sharp cylindrical hollow pipe was used to cut the filters at the same diameter as the TGA pan. As for the SMPS measurements, filters were loaded for noEGR, EGR and REGR conditions bTWC keeping the dilution ratio at 11. Also, the study included soot oxidation behaviour in the TWC. Further information of the parameters chosen for the TGA experiments can be found in the following section.

TEST PROCEDURE/METHODOLOGY

Particulate size distribution and soot oxidation behaviour for high rates of EGR and REGR were characterised in this investigation. With this aim, two injection timings producing PM with different nature, as set out previously, were selected for the study: i) 303 CAD bTDC, the ECU injection, forming small nuclei and mostly VOC particulates and ii) 335 CAD bTDC, an advanced injection, forming highly sooty particulates due to the wall-piston wetting [16]. In addition, a very early injection (335 CAD bTDC) produces enough soot to analyse the oxidation through TGA. Furthermore, the performance of the TWC in EGR and REGR conditions is analysed as well as the possible trapping effect that can occur in this aftertreatment device and its possible influence in soot oxidation. To assure the robustness of the results, each test point was repeated three times. Furthermore, confidence intervals have been added to the reference condition in order to quantify the variability of the data.

The percentage of EGR that was used for the study was the maximum achievable that maintained the coefficient of variation (COV) of maximum pressure averaging 200 cycles below 5%, to ensure the correct operation of the engine and avoid misfire. In order to compare EGR and REGR, the position of the peak of maximum pressure was held constant to make the combustion process comparable, 12 and 16

CAD aTDC for 303 CAD bTDC and 335 CAD bTDC injections timing, respectively. Therefore, when the EGR is inducted, the spark timing was advanced with respect to the baseline condition in order to maintain the combustion phasing (i.e. peak of maximum pressure centred on the same CAD position). However, in the case of REGR the spark timing was similar to the baseline condition. This information is summarised in Table 3.

Table 3. noEGR, EGR and REGR settings.

	Injection CAD bTDC	EGR (%)	Spark timing CAD	Position max in-cylinder pressure (CAD aTDC)
Baseline	303	0	24 (ECU)	16
	335	0	24 (ECU)	12
EGR	303	19	8.25(*)	16
	335	19	17.25(*)	12
REGR	303	19	3.75(*)	16
	335	24	-0.25(*)	12

(*) + advance, - retard, spark timing with respect to ECU settings.

Estimation of the Soot Oxidation Kinetic Parameters From TGA Analysis

The reactivity of particulate matter emitted by diesel engines has been broadly characterized as they have been historically the major source of PM. Several studies can be found in the literature discussing the methodology for the estimation of kinetic parameters obtained from the derivative curve (dm/dt) given by TGA [17, 18, 19, 20, 21]. In the present work, a similar method has been used to study the oxidation behaviour of soot produced from GDI engines.

The calculation of kinetic parameters from the TGA soot oxidation curve is based on the formal Arrhenius-like kinetic equation:

$$-\frac{dm}{dt} = A \cdot e^{\frac{-E_a}{RT}} \cdot m^n \cdot p_{O_2}^r \quad [2]$$

Where m is the mass of the soot sample, t, the time, E_a , the activation energy, R, the gas constant (8.314 J/mol·K), T, the sample temperature and n and r are the soot and oxygen reaction orders respectively. This form m^n is generally acceptable and often applied by researchers for heterogeneous thermal processes such as soot oxidation [17].

In the majority of studies the authors hypothesise first order reaction for oxygen partial pressure and soot mass [17, 22]. However, there is still no consensus and some other authors have reported that reaction orders with respect to oxygen partial pressures vary between 0.5 and 1 [18, 20, 23, 24]. For the present work first order reaction has been assumed, in accord with most of the literature on non-catalytic soot combustion [24].