

After-Treatment Investigation on Particulates Characterization and DPF Regeneration of a Naphtha Fuel in a Compression Ignition Engine

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ABSTRACT

Adaptation of both oil based fuel and engine technologies are key enablers to reduce CO₂ footprint as well as pollutant emissions. Recent work has demonstrated the potential of gasoline-like fuels to reduce NOX and particulate emissions when used in compression ignition engines. In addition, properties of naphtha produced directly from the atmospheric crude oil distillation process in a refinery offer significant CO₂ benefits.

When introducing such innovative fuel and engine, after-treatment investigations are mandatory to meet pollutant regulations. In that respect, this work focuses on investigating structure and properties of the particulates produced with naphtha fuel to validate Diesel Particulate Filter (DPF) design requirements. First, soot mass measurement techniques are detailed. Then, characterization of soot is performed through DPF pressure drop, soot oxidation rates with and without Fuel Borne Catalyst (FBC), composition & structure analysis. Lastly, particulate size distribution is assessed for different operating conditions.

Results show that regardless of the fuel, FBC use has the same impact on soot oxidation. A slightly lower oxidation rate of naphtha soot was observed, most likely linked to oxygenated species present in Diesel fuel. Lastly, engine combustion process, more than fuel composition seems to be the most important factor for particulates size distribution.

Finally, differences between naphtha and conventional Diesel soot are very low or negligible for all the parameters investigated in this work. Based on these results, DPF design rules and standards usually used with Diesel fuel could be transferred for a naphtha based fuel powertrain application.

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INTRODUCTION

Global transport energy demand, as well as world energy demand, is increasing, driven by economic and population growth, especially in non-OECD (Organization for Economic Co-operation and Development) countries. Petroleum-based fuels currently supply around 93% of the global energy for transport. Despite involvement of regulators, automakers or refiners to increase engine efficiency, develop alternative fuels or introduce hybridization and BEV (Battery Electric Vehicle), the position of oil-based fuels will remain largely dominant in this sector. The 2040 projection for oil-based fuels over global transport energy demand remains high at 85% [1]. Commercial activities for freight transportation such as trucking, air and marine modes are the main factor of the growth in energy demand in the transportation sector. The demand for petroleum-based fuels will consequently be skewed toward middle-distillate fuels, such as Diesel and jet fuel [2]. The worldwide existing global refinery network is not configured to meet this expected mismatch in demand. Consequently,

considerable investments in complex and expensive conversion units could be necessary. In addition to the economic considerations, additional refinery units also imply higher CO₂ emissions at the fuel refining stage. This scenario leads to an unwanted increase in the well-to-tank carbon footprint of petroleum-based fuels.

In this context, straight-run naphtha, a refinery stream directly derived from the crude oil distillation process under atmospheric pressure, was identified as a valuable fuel for engines in the transport sector. Naphtha is a generic term describing the fraction of crude oil that boils within the 30-180°C range and contains C5 to C11 hydrocarbons. Naphtha has a low Research Octane Number (RON) value (40-70). It is a refinery product that can potentially be beneficial to reduce the CO₂ footprint from well-to-tank due to less processes. Compared to Diesel fuel, naphtha-based fuels also present intrinsic chemical properties which are advantages compared to Diesel.

higher H/C ratio along with a higher Low Heating Value (LHV). These characteristics enable a reduction of CO₂ emissions from tank-to-wheel assuming the same engine efficiency (Figure 1).

Within the scope of reducing engine-out emissions, naphtha-based fuels also offer advantages. Firstly, naphtha auto-ignition properties lie in an intermediate position within the range delimited by gasoline and Diesel fuel. Its low Cetane Number (CN) allows for greater air/fuel mixing before combustion due to the increased ignition delay. The results of an enhanced air/fuel mixture are:

1. a reduction of NOx and smoke when associated with an adjustment of the fuel loop calibration of the engine;
2. a lower swirl requirement, enabling further CO₂ saving with lower heat losses and volumetric efficiency improvement.

Secondly, straight-run naphtha has less aromatic compounds, which may intrinsically lead to less soot formation.

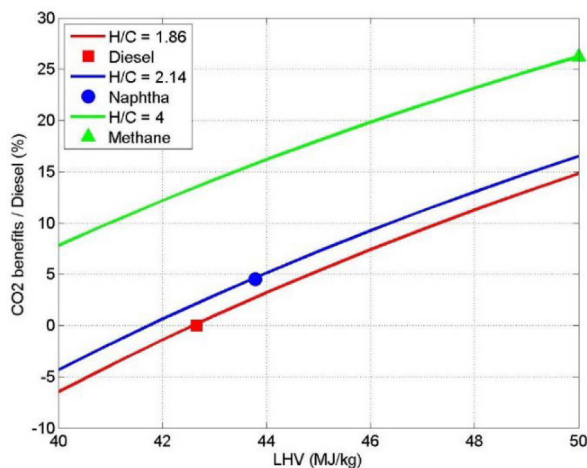


Figure 1. Theoretical CO₂ benefit (%) as a function of LHV (MJ/kg) and H to C ratio for different fuels

Many studies reported activities using gasoline-like fuels or straight-run naphtha to run compression ignition engines [3, 4, 5]. Several issues need to be addressed to adapt a Diesel engine to run with naphtha fuel. Usually authors mention the following:

- combustion stability and high level of HC and CO, due to an over-mixing between air and fuel at low load conditions. This problem becomes even more pronounced when operating in cold conditions
- noise issues when increasing the engine load as higher premixed combustion leads to a higher peak of heat release.

Worldwide agencies, engine and vehicle manufacturers, emission control system suppliers and fuel refiners are strongly working to decrease pollutant and CO₂ emissions. In Europe [6], particulates mass levels have been divided by 10 over the last 15 years (Figure 2).

First, only particulates mass was regulated but since 2011, particulates number is also limited for CI engines. In order to meet these regulations, a DPF is not mandatory but is a suitable solution to reduce PM (Particulate Matter) emission and advantageous for CO₂/NOx trade off. A reduction of particulates emissions through the use of straight-run naphtha, will help to meet pollutant regulations, shift the NOx/PM trade-off, decrease DPF size or reduce the number of regenerations between two oil services. But investigations are required to validate soot regeneration and ensure DPF integrity.

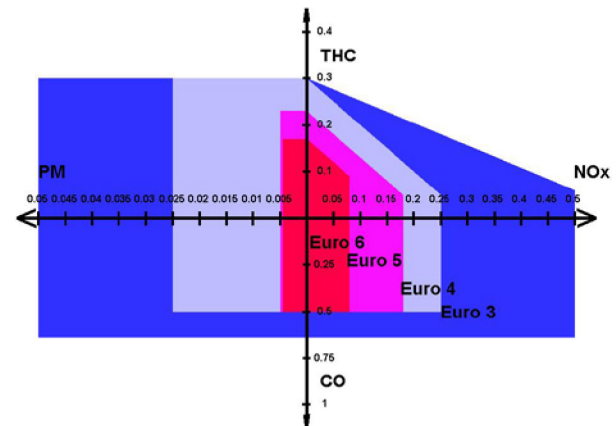


Figure 2. European Union emission standards for passenger cars (category M1) for CI (Compression Ignition) engine

This paper focuses on analyzing and comparing naphtha soot characteristics to Diesel ones. Before studying the impact of naphtha on soot characteristics, preliminary checks were performed for measurement techniques. During loading phase, weighing and particulate emission estimation were compared. Thereafter, during the regeneration phase, DPF soot mass estimation with carbon balance, based on CO emission, was also compared to weighed mass.

Weighing techniques allow to follow DPF mass evolution and consequently ash mass. The ash mass evolutions of different fuels (Diesel and naphtha), with or without FBC, were compared to literature data.

Upstream DPF temperature target and regeneration duration are linked to soot oxidation rate. Comparison of Diesel and naphtha soot oxidation rates was assessed with different temperature targets. In order to confirm engine bench references, TGA (Thermogravimetric Analysis) were also performed in laboratory facilities. Then, results of both methods were compared. Impact of FBC on naphtha and Diesel soot was investigated. Soot reactivity with uncontrolled regeneration process was assessed with FBC for both Diesel and naphtha fuels. This process was used to determine maximum loaded soot mass in order to be compliant with DPF constraints such as maximum thermal gradient.

DPF pressure drop is used in engine ECUs' (Engine Control Unit) for DPF regeneration management and DPF diagnosis. DPF pressure drop generated with naphtha soot was also investigated to validate or define a new threshold to launch the regeneration process or to detect a clogged DPF. Then, particles size distribution was analyzed for different operating conditions for both fuels. Lastly, soot composition and structure were detailed to explain potential differences.

EXPERIMENTAL SETUP

Fuels

For this after treatment study, only a straight-run naphtha with cetane number equal to 35 (CN35) and a standard EN590 Diesel fuel were used. Diesel includes 5% of FAME (Fatty Acid Methyl Esters) as bio content to respect fuel regulation.

Table 1 lists the main chemical properties of Diesel, a reference RON95 gasoline and the straight-run CN35 naphtha. The relative CO₂ benefit related to the intrinsic fuel properties (LHV and H/C ratio) is also reported. The distillation curves are presented in Figure 3.

Table 1. Chemical fuel properties of Diesel, gasoline and straight-run naphtha (CN35)

		Diesel	Gasoline	Naphtha
RON	[-]	-	96.6	41
Cetane	[-]	53.6	-	35.8
% C	[% vol]	86	84.65	84.85
% H	[% vol]	13.3	13.4	15.15
% O	[% vol]	0.7	1.92	0
Ratio H/C	[-]	1.85	1.89	2.14
Density (15°C)	[kg/L]	0.834	0.748	0.736
Viscosity (40°C)	[mm ² /s]	2.67	0.47	0.6
LHV	[kJ/kg]	42595	42650	43787
CO ₂ benefit / Diesel	[%]	-	-	4.7

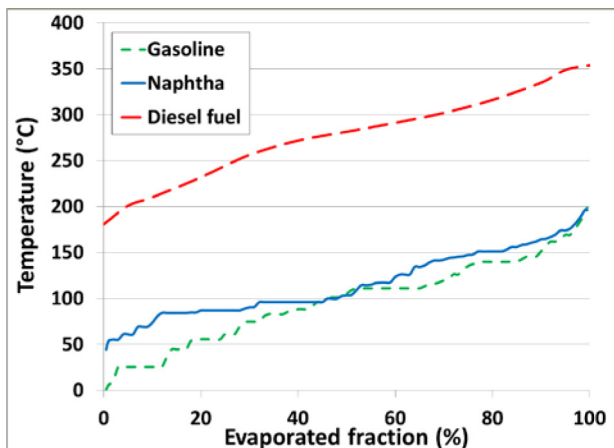


Figure 3. Distillation curves of Diesel, gasoline and straight-run naphtha (CN35)

Compared to Diesel, CN35 fuel has a lower kinematic viscosity (Table 1) and a reduced lubricating capability. Consequently, 1,000 ppm of a commercial lubricity additive were added to CN35 for all the experimental activities.

The low cetane number, the low density and the higher LHV of the naphtha fuel lead to different combustion settings and optimizations. Two types of molecules are considered the main precursors of soot in flames: unsaturated hydrocarbons and polycyclic aromatics [7]. Less aromatic content in the Naphtha composition will allow to reduce soot precursors.

Fuel Borne Catalyst (FBC)

To ensure a quick and complete regeneration of the soot contained in the DPF, a fuel additive containing an oxidation catalyst can be used. In this study, this additive is directly introduced in the fuel in the bench cell tank.

The fuel borne catalyst is the Solvay (formerly Rhodia) latest product, named Eolys Powerflex [8]. It includes a highly active Iron (Fe) catalyst, allowing active regeneration from 450°C even with a low dosing rate (6 ppm of Fe in the fuel in this study). From literature [9], nanoparticles of metal introduced in the fuel are very tightly included in the particulates matter, as shown in Figure 4.

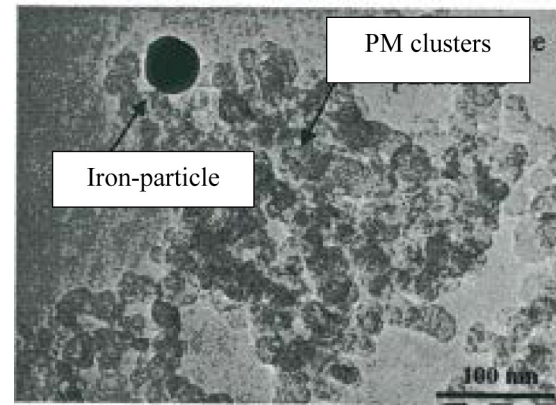


Figure 4. Iron nanoparticles introduction into a Diesel soot structure [9]

The size of the iron particulate (20-50nm) and their inclusion lead to a quicker soot light-off at lower temperature and to more complete soot combustion with lower carbon monoxide emissions.

The impact of FBC was checked for Diesel fuel and investigated with naphtha fuel during this study.

Engine and Test Bench

The experiments were performed on a 1.6L 4 cylinder conventional engine, with a 16:1 compression ratio. The engine utilizes a waste-gate turbocharger to boost the intake air to the desired pressure and also includes a self-contained HP EGR (High Pressure Exhaust Gas Recirculation) system. The engine is controlled by an open ECU. The ECU is capable of handling the fuel loop and the air loop parameters from maps or specific user dictated values. The engine provides a full power rate of 68 kW at 4000 rpm and a maximum torque of 230 Nm at 1750 rpm. Table 2 lists the specifications of the engine.

Table 2. Specifications of the multi-cylinder engine

Engine	4 cylinder engine
Displacement volume [L]	1.560
Bore [mm] x stroke [mm]	75.0 x 88.3
Valves [number /cylinder]	2
Compression ratio [-]	16:1
Swirl number [-]	2.0
Injector	Bosch, CRI 2.2
Hydraulic flow [cc/30s x100bar]	280
Nozzle holes [number]	7
Fuel pump [-]	Bosch, CP4

The exhaust line is a standard Euro5 aftertreatment system composed of a Close Coupled (CC) 1.6L DOC (Diesel Oxidation Catalyst) and a bare 2.5L DPF (dedicated to FBC use). The line has been fully equipped with:

- Temperature and pressure measurements upstream and downstream of DOC and DPF
- One internal temperature for the DOC (mid-length on longitudinal)
- 13 internal temperatures for the DPF (Figure 5) thanks to thin 0.5mm thermocouples introduced by the outlet channels
- Gas sampling upstream DOC and downstream DPF for the gas bench

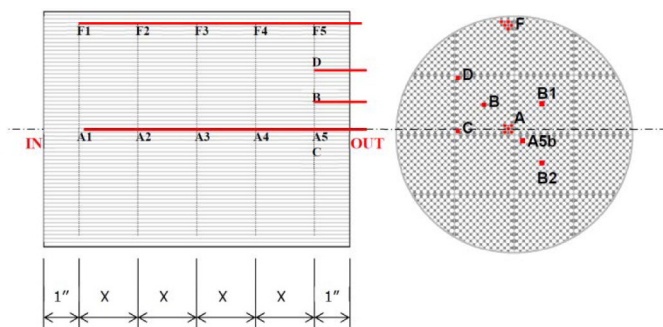


Figure 5. DPF instrumentation with 13 thermocouples (A5b, B1 and B2 have not been used). X is equal to 1 inch.

Tests were carried out with regulated coolant and oil temperatures (both 90°C) by standard control systems. The emissions and inlet air (CO_2 , O_2) were analyzed using a Horiba gas bench (MEXA-7100-DEGR). Raw soot emissions were measured using an AVL 415S smoke meter and fuel consumption with an AVL FuelExact. For steady state and transient tests, all parameters were logged once per second through the bench supervisor D2T MORPHEE 2.

Specific Measurements

Particles Size Distribution

To measure the particle size distribution, a Cambustion DMS500 system was used. DMS500 is designed to continuously measure the concentration of particulates with an electric diameter ranging from 5 to 1000 nm. The DMS500 uses its own dilution system. The sampling was done between the DOC and the DPF with a dedicated low pressure system (250 mbar absolute).

The DMS500 system is a non-certification system to measure particle concentration and size distribution, integrating a complex sampling system with an automatic double step dilution.

A unipolar corona charger places a positive charge on each particle. The charge received is approximately proportional to the particle's surface area. Charged particles are deviated in the classification section made of 22 electrometers allowing a measurement of the particle size distribution. Other parameters as PM and PN (Particulate Number) are calculated from this spectrum in real time.

That aerosol measurement apparatus is designed for engine application and engine test bench environment. Several studies are based on its usage [11, 12, 13].

Lab Tests

Thermo-Gravimetric Analysis (TGA)

Tests were performed with a Mettler-Toledo Thermobalance DSC-1 associated with IR (Infra Red) spectrometer. Feed gas is ambient air, the gas flow is 25 ml/min for a sample mass between 3.15 and 3.65 mg. The temperature increases from 20 to 800°C with a 10°C/min slope. The IR spectrometer allows a non-quantitative CO / CO_2 analysis.

Transmission Electron Microscopy (TEM)

Microscopy pictures were realized with a JEOL JEM-2100F Transmission Electron Microscope equipped with EDX (Energy Dispersive X-ray Spectrometry). EDX allows a local material analysis of points of interest.

METHODOLOGIES AND CONDITIONS

Soot Loading

The target is to have a high level of reproducibility between the different soot loadings along the project for both Diesel and Naphtha fuel combustions. The loading operating point must be carefully chosen:

- To be similar for both fuels, with representative settings
- With a high soot emission level, to maximize the number of tests in a working day
- With a good combustion stability, to ensure a constant emission level during the loading

- To ensure a similar level of particulate flow rate, BMEP (Brake Mean Effective Pressure) is adjusted to higher value for naphtha.
- Finally, with an exhaust temperature lower than 300°C upstream of the DPF to avoid any continuous regeneration with NO₂

The selected operating points corresponding to the listed criteria are presented in Table 3.

Table 3. Selected operating points for DPF soot loading

Fuel		Diesel	Naphtha
Engine speed	rpm	1500	1500
BMEP	bar	3	3.6
Injection pressure	bar	450	300
Main SOI	CA BTDC	1.2	11
EGR	%	30.5	21.4
Smoke	FSN	4.3	2.61
Part AVL	g/h	5.96	4.2
Ds DOC HC	ppmC	265	273

Once the loading OP (Operating Point) were chosen, the complete loading procedure must be defined and automated, including DPF characterization (pressure loss measurement) and DPF weighing. To fulfill all the requirements, the procedure presented in Figure 6 was designed and coded.

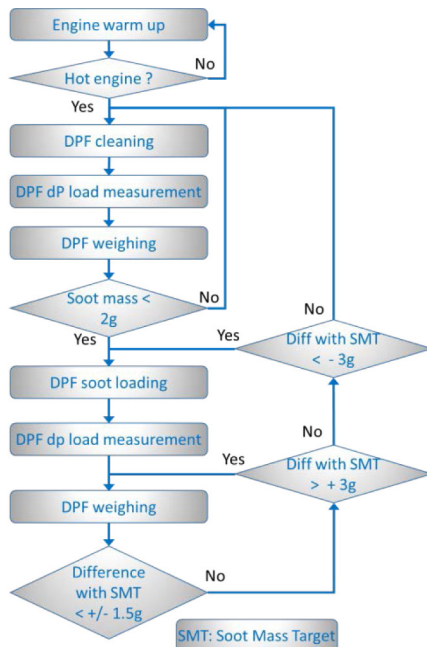


Figure 6. DPF soot loading procedure

To continuously estimate the mass of soot in the DPF during the loading, two empiric calculations based on the AVL 415S smoke number were done. Their accuracy will be tested for both Diesel and naphtha fuel later in this paper.

The first calculation is the “AVL” one [14]. Hereafter is proposed the calculation for Smoke Number (SN) lower than 8.08:

$$\text{PART}_{\text{g/h}}^{\text{AVL}} = \frac{5.32}{405} \times \text{SN}_{\text{FSN}} \times \exp(0.3062 \times \text{SN}_{\text{FSN}}) \times Q_{\text{v}^{\text{exh}}}^{\text{m}^3/\text{h}}$$

The second calculation is the “MIRA” one, used in some laboratories:

$$\text{PART}_{\text{g/h}}^{\text{MIRA}} = \text{Poly} \times Q_{\text{v}^{\text{exh}}}^{\text{m}^3/\text{h}}$$

with $\text{Poly} = 0.016 \times \text{SN}^3 + 0.0049 \times \text{SN}^2 + 0.0244 \times \text{SN}$

SN : Bosch Smoke Number from smoke meter AVL415S [FSN]

$Q_{\text{v}^{\text{exh}}}$: Volume exhaust flow [m³/h]

Pressure Loss Characterization

To have a clear follow-up of the DPF state, regular pressure loss tests were carried out for both empty and loaded cases. A specific method was developed on the engine test bench to be integrated to soot loading test. It consists of an exhaust temperature stabilization by scavenging, followed by an engine operating point's sweep. Thereby, a large range of exhaust flow was covered, allowing to plot the DPF backpressure for different exhaust flows. For a loaded DPF, only the operating points without load (engine motored by the dyno) were used. See Table 4.

Table 4. Pressure loss selected operating points

Engine Speed	BMEP	Exhaust MFR		
[rpm]	[bar]	[kg/h]		
1000	motored	49	Loaded DPF	Empty DPF
1750	motored	92		
2500	motored	136		
3250	motored	171		
4000	motored	188		
3500	0	220		
3750	0	240		
4000	0	259		
4000	4.7	290		
4000	9.74	330		
4000	13.08	350		

Active Regeneration for a Steady Temperature

After DPF loading, with a soot mass objective typically equal to 15g (6.3 g/L for this 2.4L DPF), regeneration was carried out. To observe the kinetic of the soot combustion, a specific methodology was developed to make an active regeneration with a temperature target between 450 and 650°C.

The regeneration operating point, 2000 rpm - 4.5 bar BMEP, was chosen to be achievable on a vehicle: it corresponds to a steady 90 km/h on a chassis dyno. The settings were similar between Diesel and naphtha combustion, only the post-injection rates were adapted to reach the temperature targets.

The regeneration procedure was coded to ensure a good test accuracy. It consisted in a warm-up (DOC temperature = 260°C) prior to a “regeneration” mode with a post-injection quantity chosen to reach the temperature target. That regeneration mode was maintained during 15 minutes, then the exhaust temperature was cooled down and the engine stopped.

An example is given in Figure 7 for a 15g DPF soot loading and a 600°C target temperature.

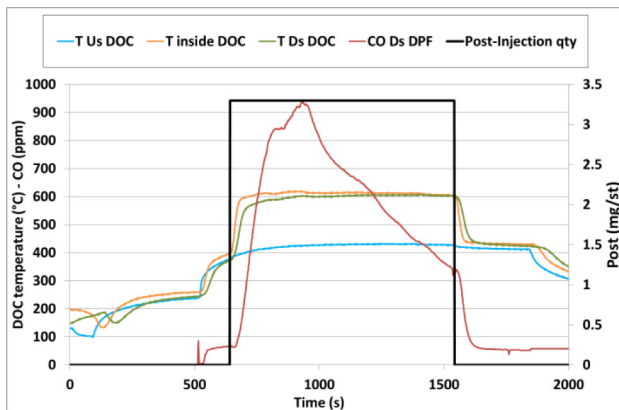


Figure 7. Active Regeneration example (Soot Mass = 15g, Target Temperature = 600°C)

Oxidation rate is calculated from the DPF weighing before and after the regeneration and regeneration duration. For uncomplete regenerations, total duration (900 seconds) is considered. For complete regeneration (>90% of the loaded soot mass), regeneration duration is determined with CO emission. Difference between tailpipe CO emission and engine CO emission gives the CO emitted from the soot oxidation. Regeneration is considered over when a threshold of 99% of the cumulated CO emission is reached. Duration of the regeneration is calculated with the difference of the timing of the threshold and the beginning of the post-injection activation.

Uncontrolled Regeneration Test

Another way to test the reactivity potential of the soot is to carry out a Soot Mass Limit (or SML) test. That test was designed to define the maximum soot mass admissible in a DPF. For that, the DPF was loaded to a soot mass target (typically 7 g/L) and an uncontrolled regeneration process was launched. That severe regeneration is based on the field experience showing that the worst case, for a highly loaded DPF, is a drop-to-idle occurring after a regeneration start [15].

That worst case was reproduced on the engine test bench by an active regeneration (target 600°C) interrupted with a drop-to-idle as soon as a negative pressure drop gradient was observed.

This procedure leads to high internal temperature and thermal gradients, as illustrated in Figure 8. Upstream (Us), inside (different locations, Figure 5) or downstream temperatures were measured.

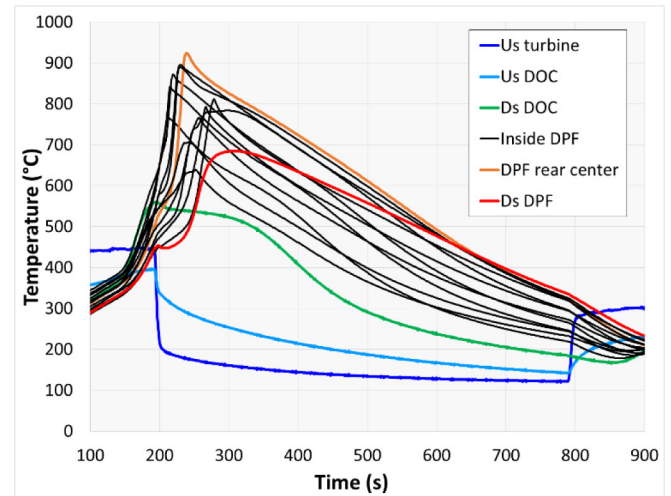


Figure 8. Illustration of internal DPF temperatures evolutions during a MSL test

Particle Size Distribution

To compare the particle size distribution between a naphtha and a Diesel combustion, 6 operating points representative of a WLTC (Worldwide harmonized Light vehicles Test Cycle) cycle as well as 2 operating points at full load were optimized for both fuels based on previous works [16]. They are listed here and illustrated in Figure 9:

- 2000 rpm / 2 bar BMEP
- 1500 rpm / 3 bar BMEP
- 1500 rpm / 6 bar BMEP
- 1250 rpm / 8 bar BMEP
- 1750 rpm / 17 bar BMEP
- 1750 rpm / Full Load
- 2650 rpm / 10 bar BMEP
- 3500 rpm / Full Load

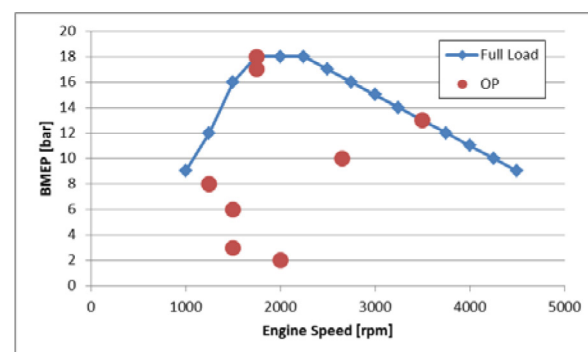


Figure 9. Operating points for particle size distribution study