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The Effect of NOx /Soot Ratio on the Regeneration Behaviour of Catalysed Diesel Particulate Filters for Heavy Duty Applications

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ABSTRACT

The control over particulate emissions is becoming increasingly important in modern diesel engines for Heavy Duty applications, that will comply to more and more stringent emissions norms. Use of particulate traps is an effective means of achieving this with the need to regenerate the particulate trap being imperative.

Passive regeneration using NO_2 by conversion of NO, as well as regeneration at lower temperatures with catalyzed DPF and the influence of NO_X to soot ratio on this, is the subject of the paper.

Both coated and uncoated filters in fresh and aged state are evaluated at temperatures typical of passive NO_2 and Oxygen-based soot regenerations and the results discussed.

INTRODUCTION

Since the middle of the 1980s, exhaust emission regulations for commercial vehicles have been updated and tightened. In Europe Euro 4 is introduced in 2005 and the further step will be introduced 2008 (EURO 5) using both transient and steady state certification tests, the ESC (European Stationary Cycle) and ETC (European Transient Cycle). Significant the legal limits for 2005 and 2008 include important reductions in NO_X and Particulate Mass (PM).

Similarly, regulations in Japan and the United States have been established which enforce even more severe reductions in NO_X and PM. India is also having Euro 3 equivalent norms in 2005 and Euro 4 is proposed by 2008 or 2010 (Fig. 1).

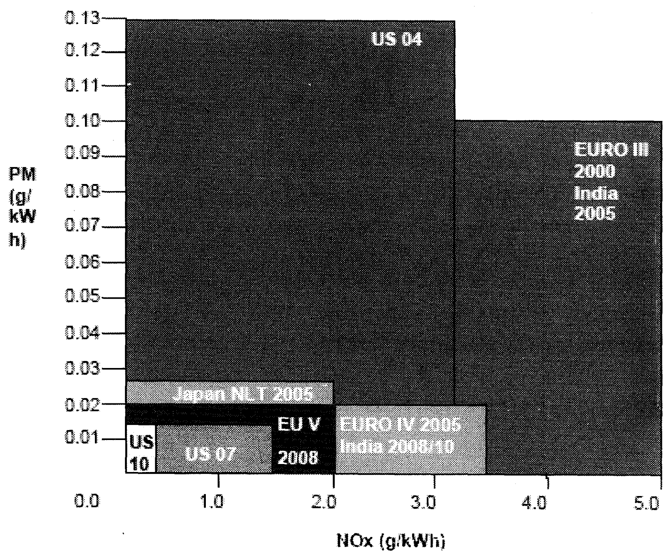

Fig.1 Future Emission Regulation Limits for Commercial Vehicles

As a result, heavy-duty (HD) engine manufacturers may be required to use advanced exhaust aftertreatment systems to meet these limits in the future.

While the control over CO and HC conversions by use of Diesel Oxidation Catalysts is well known, the trade off between particulate matter and NOx emissions may be addressed by:

- Particle reduction by optimizing engine control parameters combined with NO_X reduction by appropriate aftertreatment devices (e. g. Selective Catalyst Reduction or Lean NO_X Traps)
- NO_X reduction via cooled EGR combined with

A variety of filter materials is available that provide filtration efficiencies of 95 %. This level of particle reduction appears to be sufficient to meet the strictest regulations, however, the accumulation of soot on the filter leads to higher back pressure and corresponding increases in fuel consumption. Today's development therefore concentrates on the reliable regeneration of filters by soot combustion. Ideally, this regeneration should take place with a minimal input of energy or additives, yet be quick and reliable. This demand can be achieved by using catalysed filters [1–6].

By converting NO to NO_2, catalytic coatings significantly decrease filter regeneration temperatures. NO_2 is known to support soot combustion at temperatures of 300 °C. For the complete combustion of a given amount of soot, a stoichiometric mass of NO_2 is required. In future applications, the amount of NO_2 available for soot combustion may be limited due to two significant restrictions:

- Decrease of NO_X emissions by engine measures such as EGR or other advanced combustion methods
- Diminishing activity of the catalytic coating due to aging mechanisms [5]

Hence, a major task of catalyst development is to help ensure an adequate supply of NO_2 necessary for regeneration of the soot-loaded filter. This goal is even more challenging because of the expected lowering in NO_X raw emissions by future engine concepts [4].

In this paper a study will be presented showing the influence of NO_X concentration and exhaust flow rate on the regeneration rate of soot-loaded filters.

EXPERIMENTAL SETUP

The experiments described below were performed on a heavy-duty bench equipped with a turbocharged, intercooled in-line 6-cylinder diesel engine with specifications shown in Table 1. Fuel with a sulfur concentration of 30 ppm was used.

Table 1 Engine Data

Displacement	6.37 liter
Rated power	205 kW @ 2300 rpm
Peak torque	1100 Nm @ 1200-1600 rpm
Fuel Injection System	Pump – Line – Nozzle
Certification	EURO 3

The test cell's dynamometer allowed the engine to operate across its entire speed-load map, including fully transient test cycles like the ETC and HD FTP. Data were recorded at a frequency of 1 Hz.

Figure 2 shows the configuration of the test equipment (SCR) system was placed in front of the Catalysed Diesel Particulate Filter (CDPF) to lower the level of engine out NO_X emissions to simulate expected future emission limits and their impact on CDPF soot regeneration. The urea injection strategy was controlled by a NO_X emission map stored in the ECU of the dosing unit.

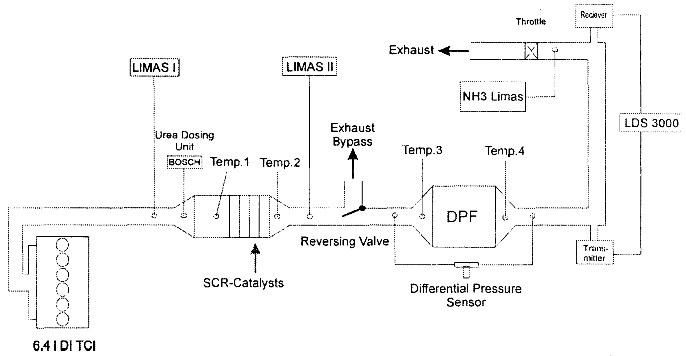

Fig.2 Schematic Representation of the Test Bench

A valve in front of the filter allowed exhaust flow to be bypassed around this device on demand. While being operated in this mode, a flow of inert gas was passed across the CDPF to flush the system. This configuration was designed to produce defined and reproducible soot burning by avoiding any uncontrolled chemical reactions which might otherwise occur within the CDPF during this operation.

The test bench was equipped with standard exhaust gas measuring systems. The focus of these tests was measuring the concentrations of NO and NO_2 at the following locations within the exhaust line:

- Upstream of the SCR catalyst (raw emission)
- Downstream of the SCR catalyst
- Downstream of the CDPF

Additionally, NH_3 concentrations were measured downstream of the SCR catalyst and CDPF by a Laser based SIEMENS LDS 3000 unit and an ABB LIMAS 11-UV instrument, respectively. Thermocouples were installed upstream and downstream of the SCR system and the CDPF. Additionally, a differential pressure sensor was employed to measure the pressure drop across the CDPF during soot loading and regeneration.

The test procedure to analyze soot burning rate as a function of NO_X concentration required referencing of the soot loading within the CDPF before each stationary and transient test. To do this, filters were first conditioned at 130°C in an oven and immediately weighed. They were then loaded with 50 grams of soot by operating the engine at constant speed (1400 rpm) and a load generating a filter inlet temperature of 240 °C [51. Subsequently, the filter was conditioned and reweighed. CDPFs were regenerated for 10 minutes, and again conditioned and weighed. This procedure helped ensure the reproducibility of filter state before each test.

The geometric dimensions (diameter x length) of all of 17 liters. Catalysed filters with 200 cells per in^2 (cpsi) were utilized throughout these studies. The catalysed filter is with 35 g/cft precious metal loading on a cordierite NGK DHC 611 substrate.

INVESTIGATION OF THE IMPACT OF NO_X/SOOT RATIO ON SOOT OXIDATION RATE

In order to study the soot-burning rate as a function of different NO_X/soot ratios, engine speed, and CDPF aging state, the setup shown in Fig. 2 was employed. The strategy was to utilize an SCR catalyst unit upstream of the CDPF to reduce NO_X concentrations within the exhaust. By doing this, it was possible to simulate different engine-out NO_X levels at selected load points and engine speeds without changing the corresponding soot production rate, or other raw emission levels. The level of NO_X reduction for a given load point was controlled by adjusting the rate of urea solution injection. Conversion levels were limited such that no ammonia slip was detected downstream of the SCR catalyst.

To study the soot oxidation rate of the catalytic soot filters on this engine bench setup, a specific test procedure shown in Fig. 3 was established.

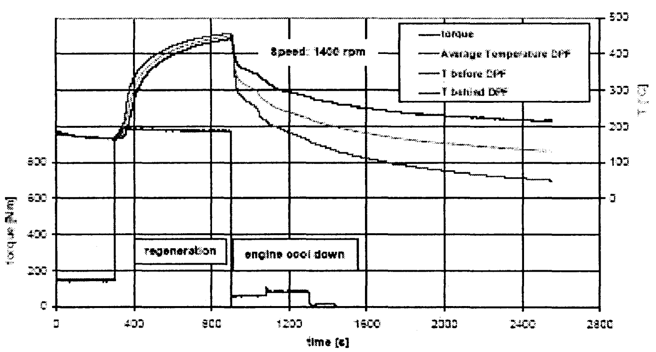

Fig. 3 Overview of Soot Regeneration Test Conditions

The general procedure starts with a conditioning phase of low torque for 5 minutes. The regeneration of the soot is initiated by a load jump at constant engine speed which is maintained for 10 minutes before the cool down phase begins. During the cool down phase, engine exhaust is bypassed around the CDPF. At the same time, the filter is cooled in a nitrogen atmosphere to quench soot burning after the 10 minutes soot regeneration period. To determine soot burning rate, each filter was weighed before and after the test sequence.

To generate reproducible results, this procedure required a well-defined initial state of loading within the CDPF before and after testing. The test filters were loaded with 3 g/L soot at an engine speed of 1400 rpm using a specially designed soot loading procedure [1]. The soot-loaded filter was conditioned in an oven at 130 °C and weighed. Afterwards the CDPF was installed in the test equipment shown in Fig. 2 and regenerated according to the procedure outlined in Fig. 3. The filter was then removed, conditioned in an oven and reweighed to

For the evaluation of the soot burning rate as a function of average CDPF temperature in the range of 300 – 450 °C, torque was adjusted at engine speeds of 1000, 1400 and 1800 rpm. The nominal temperatures and engine-out NO_X emissions for the selected engine operating points are shown in Fig. 4.

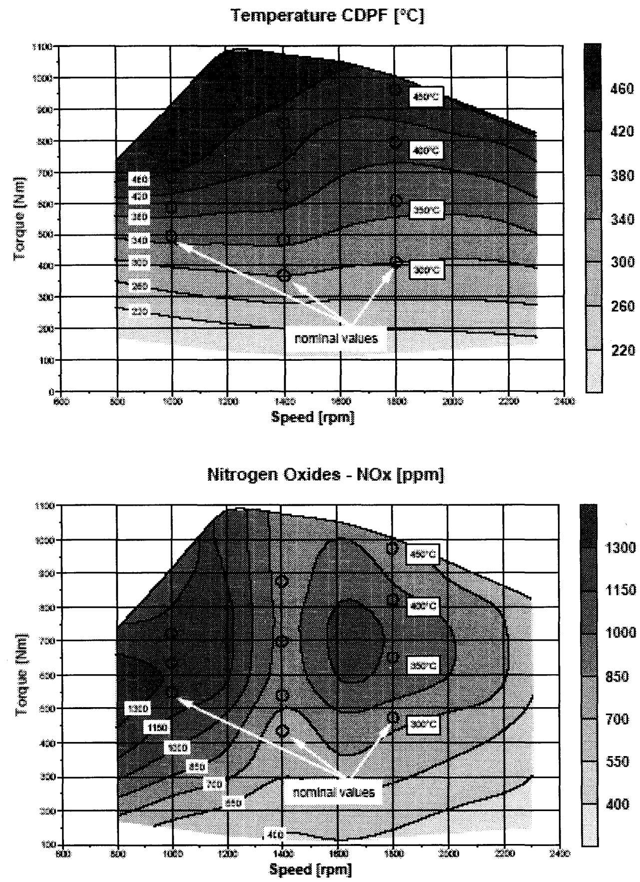

Fig. 4 Average Temperatures in CDPF and Engine-Out NO_X Emissions for Selected Evaluation Points Used in the Soot Regeneration Test

For the experiments with modified NO_X/soot ratio, the same engine operating points as shown above were selected, however, NO_X reductions of 50 and 70 % were targeted by operating the SCR unit upstream of the filter during the corresponding soot regeneration test. The specific NO_X output (i. e. NO_X input to the CDPF) ranged from < 1.5 g/kWh for the 70 % NO_X reduction case to 2.0 – 2.4 g/kWh for the 50 % NO_X reduction case, based on an engine-out NO_X level of 4.2 – 5.5 g/kWh.

Impact of NO_X/Soot Ratio at Constant Engine Speed

At a constant engine speed of 1400 rpm the average soot burning rate (SBR) was determined at several temperatures, as well as at varying NO_X emission levels at the inlet to the CDPF. The results are summarized in Fig. 5.

At a constant NO_X mass flow (flux), the soot burning rate (SBR) rises with increasing exhaust gas temperature. Similarly, with increasing NO_X mass flow the soot burning rate increases as well.

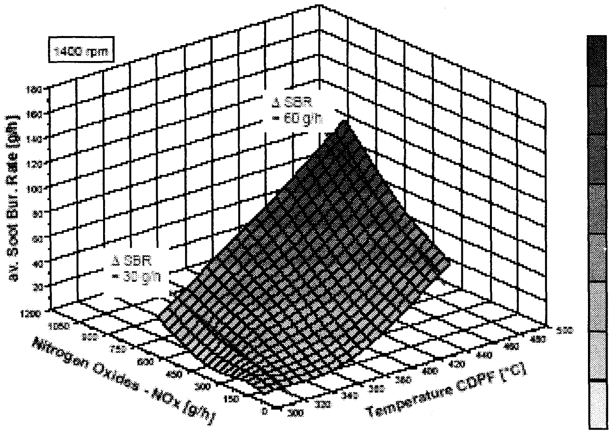

Fig. 5: Average Soot Burning Rate (SBR) as a Function of Temperature and NO_X Mass Flow at 1400 rpm for CDPF (Conditioned).

The change in soot burning rate with change in NO_X flux (Δ SBR) also strongly depends on temperature. For the same increase in NO_X mass flow (150 – 750 g/h) the change in soot burning rate at 440 °C is twice that observed at 320 °C.

This effect is also observed when the amount of soot emitted by the engine is taken into consideration. In Fig.6, the average soot-burning rate is presented as a function of NO_X/soot ratio.

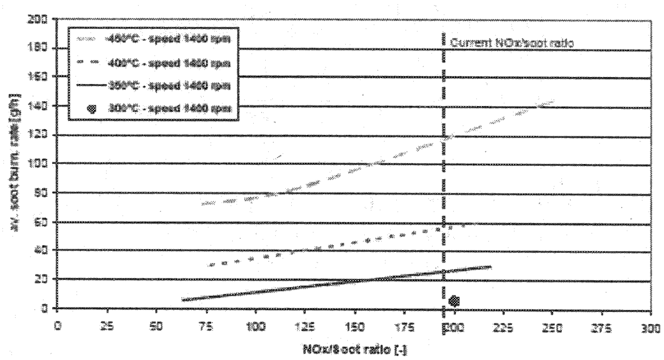

Fig. 6 Average Soot Burning Rate as a Function of NO_X/Soot Ratio at Constant Speed and Different Temperatures

The balance point of the investigated CDPF was earlier determined to be about 300 °C. Accordingly, the soot-burning rate is low even at a high NO_X/soot ratio such as that indicated by the solid black circle in Fig. 6. Fig. 6 suggests that CDPF can be regenerated at all investigated temperatures, even at NO_X/soot ratios as low as projected for future heavy duty engines.

At temperatures up to 350 °C, passive filter regeneration is already limited at today's NO_X/soot levels. Yet, lowering the NO_X/soot ratio only results in a slight decrease in soot burning rate. The lower the temperature of the investigated engine operating point, the less the impact a decrease in NO_X/soot ratio has on soot burning rate.

Evaluation of the Influence of Engine Speed on Soot Burning Rate

To investigate the impact of different mass flow rates on filter regeneration behavior, experiments have been performed at engine speeds of 1000, 1400, 1800 rpm. For this set of experiments engine out NO_X levels were not reduced by the use of the SCR unit upstream of the CDPF. These tests also utilized the conditioned CDPF. In Fig. 7, the average soot-burning rate is plotted versus temperature and the NO_X flux at the inlet to the CDPF.

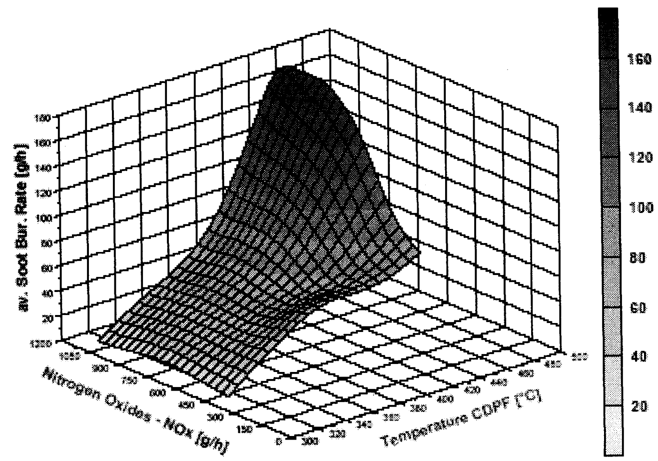

Fig.7 Average Soot Burning Rate as a Function of NO_X Flow Rate and Temperature at 1000, 1400 and 1800 rpm

In general, the impact of different NO_X and temperature levels as found at a constant engine speed (see Fig. 5) seems to be confirmed. Upon closer inspection, however, an interesting difference is revealed. At lower temperatures, an increasing NO_X mass flow does not necessarily result in a higher soot burning rate.

To understand this phenomenon a detailed consideration of the chemical processes associated with soot filter regeneration is necessary. The main chemical reactions that are involved in soot combustion are summarized in equation (1) and (2):

$$C + NO_2 \rightarrow CO + NO \qquad T > 200°C \qquad (1)$$

$$C + O_2 \rightarrow CO + \tfrac{1}{2} O_2 \qquad T > 450°C \qquad (2)$$

While diesel exhaust gas generally contains an excess of oxygen needed for the reaction described in equation (2), it contains only low levels of NO_2 which is required for reaction according to equation (1). An oxidation catalyst is needed to facilitate the conversion of NO to NO_2 according to equation (3), thereby enabling low temperature soot burning as described in equation (1).

$$NO + \tfrac{1}{2} O_2 \leftrightarrow NO_2 \qquad (3)$$

The NO oxidation given in equation (3) is subject to thermodynamic restrictions. At higher temperatures the equilibrium is shifted to the left side of reaction (3) resulting in a limited NO_2/NO ratio. Fig. 8 provides a comparison between the theoretical NO_2/NO limited ratio and the experimentally observed values, both as a function of exhaust gas temperature.

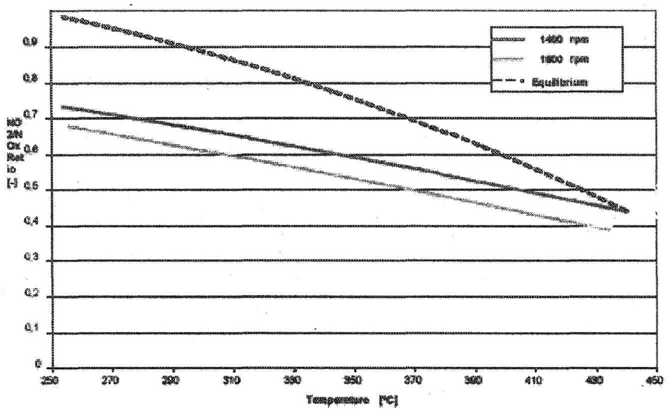

Fig.8 NO_2/NO Ratio as a Function of Temperature: Thermodynamic Equilibrium Compared to Experimentally Obtained Values in the Exhaust Gas

NO_2 formation in the experiment decreases with increasing temperature due to thermodynamic restriction as indicated by the dashed line. The lower the temperature the more the experimental values differ from the equilibrium. This means that in the lower temperature range the oxidation of NO to NO_2 is kinetically limited. This reaction rate is accelerated by the catalytic coating of the filter, however, the reaction rate is not only determined by NO_X exhaust gas concentration and temperature. There is also an impact of engine speed (i. e. space velocity over CDPF).

At 1400 rpm, NO_X concentration in the engine exhaust is lower than at 1800 rpm (see Fig. 4). Standard kinetic expressions for NO oxidation predict that this should lead to lower reaction rates at a given temperature [6]. Nevertheless, NO conversion at 1400 rpm is higher than at 1800 rpm as shown in Fig. 8. The reason for this phenomenon is the higher exhaust gas flow at 1800 rpm resulting in a lower residence time of the exhaust gas inside the CDPF. This in turn leads to decreased reaction rates associated with mass diffusion limitations at the catalytic sites.

Despite the decreased reaction rates at 1800 rpm, the NO_2 concentration is higher at 1800 rpm due to the higher NO_X raw emissions (see Fig. 9).

The highest NO_2 concentration within the exhaust gas was measured at 350 °C with the lowest exhaust gas flow. In this case all considered rate influencing factors are beneficial with regard to NO_2 formation: high NO_X concentration, high theoretical limit for NO_2/NO ratio, and long residence time in CDPF. However, for the determination of the final amount of NO_2 that is available for soot oxidation according to equation (1) the influence of the exhaust gas flow has to be included. The NO_2 mass flows generated at each of the investigated engine operating points are shown in Fig.10.

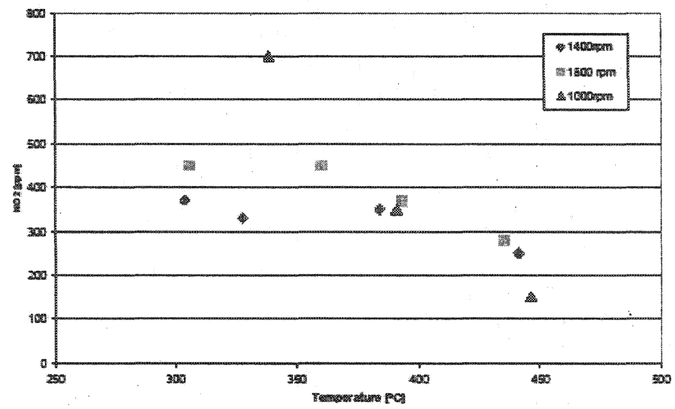

Fig.9 NO_2 Concentration as a Function of Temperature and Engine Speed

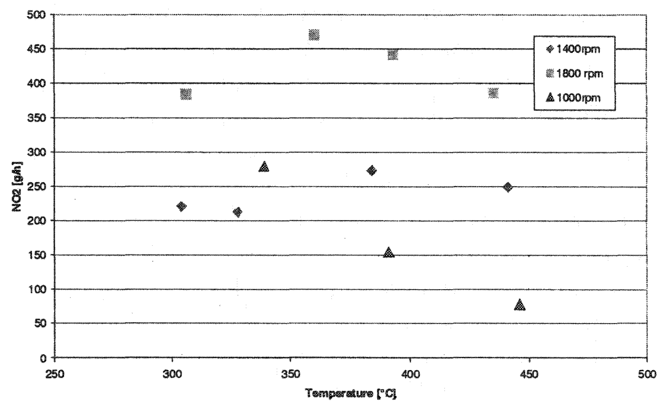

Fig. 10 NO_2 Flow as a Function of Temperature and Engine Speed

Although the highest NO_2 concentration was generated at 1000 rpm and 350 °C, the NO_2 mass flow is comparatively low. A higher NO_2 mass flow was generated at lower NO_2 concentrations but higher exhaust gas flow at 1800 rpm. Since the mass of soot being converted according to equation (1) depends on the mass flow of NO_2, this should result in higher soot burning rates for 1800 rpm as compared to 1000 rpm. Summary of these results are given in Fig.11.

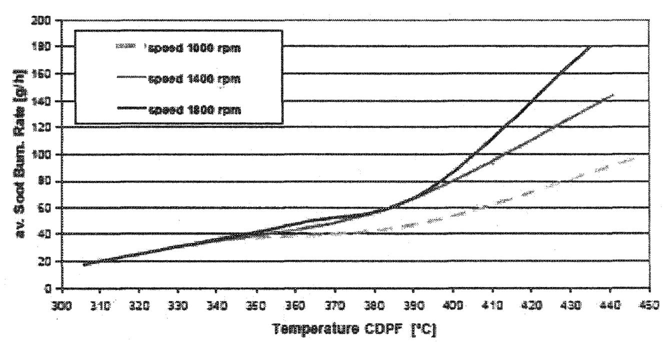

Fig. 11 Average Soot Burning Rate as a Function of Temperature and Engine Speed

Interestingly, the soot-burning rate for this experimental setup is independent of the engine speed up to a temperature of 380 °C. Although according to Fig. 10 almost twice as much NO_2 is available at 1800 rpm, this

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