An optimal NO_x assisted abatement of diesel soot in an advanced catalytic filter design

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Abstract

Two standard soot catalytic filter configurations and a so-called TU Delft (TUD) catalytic filter have been investigated in laboratory equipment to evaluate its potential as diesel soot exhaust catalytic filter. The first configuration, where Pt/SiC foam catalyst was placed upstream of soot loaded on SiC foam, showed the lowest NO_2-slip and the lowest soot oxidation rate. In the second configuration, soot was loaded on Pt/SiC foam. This gives rise to a higher soot oxidation rate at the expense of a higher NO_2-slip. An improvement on the utilisation of NO_2 with lower NO_2-slip has been observed in the TUD catalytic filter in addition with the highest soot oxidation rate. As with all diesel particulates filter TUD catalytic filter up to 10% CO (around 50 ppm) is formed as soot combustion product, whereas for the miniaturisation of the commercially available continuously regeneration trap (CRT) system at the same diesel soot oxidation rate 120–140 ppm CO is observed. Calculations to assess the potential application of the system showed that the system is promising.
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1. Introduction

The oxidation of diesel soot with NO_2 is the basic reaction in the application of the so-called continuously regeneration trap (CRT) technology. Without any doubt, this is a breakthrough in the abatement of diesel soot emission. In the CRT technology, NO_2 is generated from the oxidation of NO over a platinum catalyst supported in a flow-through monolith [1]. NO_2 generated is subsequently used to oxidise the downstream soot trapped on a wall-flow monolith. This technology showed an excellent performance during a road test over 600,000 km in the presence of ultra-low sulphur diesel fuel [2].

Driven by legislation, however, a trend in the production of new vehicle powered by diesel engine is to decrease NO_x emission by the improvement in diesel engine combustion technology and at the same time to improve the fuel efficiency of the engine. Other so-called primary measures in fulfilling the future more stringent emissions standards are fuel modifications and intake air modifications that will result in lower NO_x formation (up to 30–60%) [3].

These improvements are probably not sufficient to comply to the NO_x standard as defined in Euro IV and V. Nevertheless, the trend can have an impact on the performance of the soot abatement technology relying on exhaust NO_x due to limited NO_x/soot ratio. The following calculation deals with this critical NO_x/soot

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ratio for the CRT system to operate. If it is assumed that the reaction stoichiometry is:

$$NO_2 + C \rightarrow CO + NO \tag{1}$$

the mass ratio of NO_2 to C for reaction (1) is around 4. Because almost all NO_2 is produced from the oxidation of engine out NO and if one assumes that 50% of NO_x will be oxidised to NO_2, the critical engine out-NO_x/C ratio will be 8 by mass. If, however, the reaction stoichiometry is:

$$2NO_2 + C \rightarrow CO_2 + 2NO \tag{2}$$

The NO_x/soot ratio even becomes 16. Moreover, some of NO_2 will pass the trap system without reacting. This phenomenon is called NO_2-slip.

Test on a truck equipped with exhaust gas re-circulation (EGR) showed that the NO_x/particulate matter ratio can be in a range from 5 to 50 [4]. Some typical 1998 new passenger cars have the NO_x/particulate matter ratio between 5 and 20 [5] and for diesel engines of tomorrow technology, the ratio might decrease even further. This trend might be a burden in the application of the CRT technology.

This NO_x/soot ratio problem can be solved by a better utilisation of NO_x. In principle, this can be realised by giving NO a catalytic role as follows:

$$NO + \tfrac{1}{2}O_2 \xrightleftharpoons{Pt} NO_2$$

$$NO_2 + C \rightarrow CO + NO \tag{3}$$
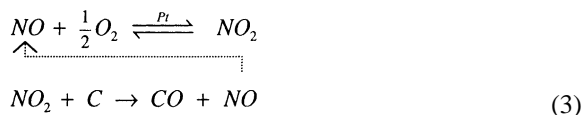
Recently, we published a paper describing that under laboratory conditions, this recycle reaction mechanism can be achieved. Two Pt–soot configurations were compared, *viz.* (A) a separated system consists of platinum catalyst upstream of a soot bed, and (B) a physical mixture of platinum and soot. We observed that a physical mixture of soot and platinum outperforms the separated system. The high oxidation rate of the physical mixture was due to the recycle reaction of NO–NO_2, that took place at least three times on average [6].

In this investigation, we explore the potential of this finding for a practical application. A new catalytic filter concept will be defined and consists of Pt/SiC foam and a SiC-membrane wall-flow monolith. The possibility of minimising NO_2-slip is explicitly taken into account.

Since the oxidation of soot with NO_2 and diesel particulates traps in general can yield secondary CO emissions (see Eq. (1)) the $CO/(CO + CO_2)$ is monitored to assess the potential of the investigated systems in CO emissions. At the end of the article an assessment on the practical application of the TUD system will be given.

2. Experiments

The catalytic filter configurations used in this investigation are Pt impregnated ceramic foam and a combination of Pt impregnated ceramic foam with ceramic membrane monolith. Pt impregnated ceramic foam was prepared by incipient wetness impregnation of SiC foam (20 ppi, Porvair Advanced Material). A 240 mg $Pt(NH_3)_4 \cdot Cl_2 \cdot H_2O$ (Engelhard) was dissolved in 2.5 cm^3 de-mineralised water and used as catalyst precursor solution. The impregnated foam was dried in 355 K in an oven for 2 h, and subsequently, calcined at 775 K for 2 h. This preparation method resulted in about 1.5% platinum content, as confirmed by XRF analysis.

A combination of SiC foam and a SiC membrane was made to prepare a so-called *TUD catalytic filter* configuration. A 30 mm length SiC 100 cpsi flow-through monolith (NOTOX) with a mean pore size of 55 μm and 2 mm wall thickness is used as the second compartment of the system. The alternate monolith channels are blocked by ceramic glue (Ceramabond 503 Aremco product), that is compatible for SiC material.

Soot sample used in this investigation is Printex-u, a model soot (Degussa). Soot was deposited on Pt/SiC foam and SiC monolith by dipping it into the soot *n*-heptane dispersion followed by evaporating at 353 K. This dipping procedure was repeated until the desired amount of soot was obtained.

The soot oxidation experiments were performed in a flow-reactor equipment. The simplified flow scheme is shown in Fig. 1. The feed gasses used can be carbon monoxide, nitrogen monoxide, SO_2, propene as hydrocarbon simulator, oxygen, and argon as a balance gas. With the exception of oxygen and water all gasses are mixed in the mixing section. After mixing the gas mixture is fed to the vaporiser, where water is injected. Oxygen is introduced just in front of the
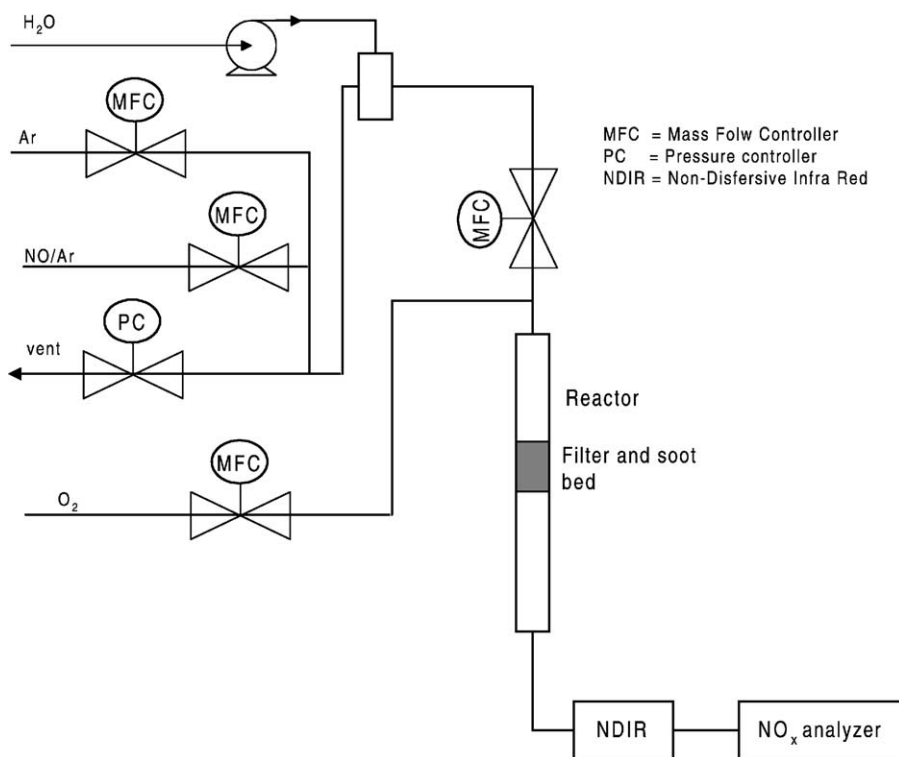
Fig. 1. Flow scheme of reactor set-up.

reactor to minimise the oxidation of NO to NO_2 before the reactant gas enters the reactor. Only up to 3% NO to NO_2 conversion is observed.

A Hartman & Braun Uras 10E Non-Dispersive Infra-Red analyser (NDIR) is used to analyse CO, CO_2, NO, and SO_2. A NO_x analyser, CLD 700 EL ht from Eco physics, is placed downstream of the NDIR to analyse NO and NO_2. For the temperature programmed oxidation experiments, the sample was heated to 775 K with 0.2 K/min, followed by 1.5 K/min to 900 K and kept there for 1 h to verify the mass balance. The reactant gas, NO (typically 700 ppm) and O_2 (typically 10 vol.%) in Ar (gas balance) were fed starting from 375 K. The gas-flow rate is 950 ml/min resulting in around 65,000 l/(l h) gas hourly space velocity (GHSV). This value of GHSV is a typical value encountered in 'high speed' part of trucks and passenger cars regulation test driving cycles. At full load of the engines GHSV values of over 300.000 l/(l h) are encountered.

The soot-catalytic filter arrangements are schematically described in Table 1.
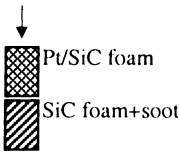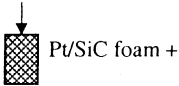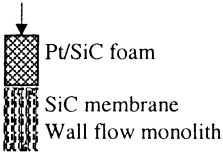
3. Result

3.1. Basic utilisation of NO_2 in Pt/SiC foam system in the oxidation of soot

The mass balances of soot were calculated by integrating the amounts of CO and CO_2 released during reaction. The typical mass balances obtained were about 90–105%.

Fig. 2 compares the oxidation profile of the separated system with the integrated system. In this figure the oxidation profile of Pt/SiC foam catalysed soot oxidation with O_2 is also displayed. The oxidation rate is expressed by normalising to its initial mass to allow reliable comparison. Moreover, it can be used to predict the oxidation rate at different soot initial mass that

Table 1
Experimental schemes and gas compositions

Catalyst configurations	Schematic figures	Gas compositions
(1) Separated system Pt/SiC foam upstream of ceramic foam loaded soot	Pt/SiC foam SiC foam+soot 200 mg Soot was loaded only on SiC foam	(1) 700 ppm NO + 10% O_2
(2) Integrated system	Pt/SiC foam +soot 200 mg Soot was loaded only on SiC foam	(1) 10% O_2, (2) 700 ppm NO + 10% O_2, (3) 700 ppm NO + 10% O_2 + 3% H_2O
(3) TUD filter system	Pt/SiC foam SiC membrane Wall flow monolith 100 mg soot was loaded on Pt/SiC foam and 100 mg on SiC membrane	(1) 700 ppm NO + 10% O_2, (2) 700 ppm NO + 10% O_2 + 3% H_2O

will give the same profile in its normalised form. Compared to O_2, the NO/O_2 system shows much higher oxidation rates in both configurations. Nevertheless, the two configurations of NO/O_2 system are far from identical. Only at low temperatures the oxidation profiles of the two configurations are similar. At higher temperatures the separated system shows a moderate increase in soot oxidation rate with an increase of temperatures. This configuration has a maximum oxidation rate of 17 $\mu g/(g_{initial}$ s) at 625 K. At increasing temperatures, the integrated configuration shows a tremendous increase in oxidation rate. The maximum rate is 36 $\mu g/(g_{initial}$ s) at a temperature around 600 K. For practical application the presence of 3% water in the NO/O_2 gas stream is beneficial. Due to the presence of water the soot oxidation rate is significantly increased, especially at low temperature. At for example 550 K, the rate is about 16 $\mu g/(g_{initial}$ s) for the water free system and 27 $\mu g/(g_{initial}$ s) for the system with water.

Fig. 3 represents NO_2-slip and $CO + CO_2$ produced from the reaction systems. The NO_2, that is generated under these conditions, is included in this figure. These data have been measured in a separate experiment in the absence of soot and are referred to as the 'base case NO_2'. The difference between the base case NO_2 and NO_2-slip at any particular temperature is NO_2 that has reacted with soot at least one time. In the separated system, as shown in Fig. 3a, it is clear that at low temperatures the increase in the base case NO_2 is accompanied by an increase in $CO + CO_2$ production. At higher temperatures, an enhancement in $CO + CO_2$ production is accompanied by a decrease in NO_2-slip. At the temperatures around 625 K, the amount of NO_2-slip reaches its lowest value (around 50 ppm).

In the integrated system, as illustrated in Fig. 3b, the production of $CO + CO_2$ is significantly higher than that of the separated system. Between 525 and 625 K the amount of $CO + CO_2$ is also higher than the difference between the base case NO_2 and NO_2-slip. Nevertheless, the amount of NO_2-slip is also high. When H_2O is present in the NO/O_2 gas stream of the
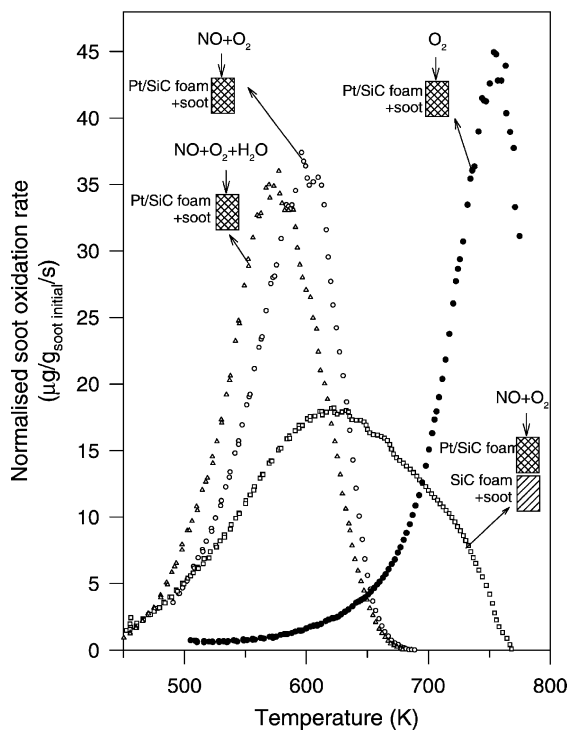
Fig. 2. Effect of catalyst configuration and gas composition on soot oxidation rate; temperature programmed reaction; heating rate of 0.2 K/min with 10% O_2, 700 ppm NO, 3% H_2O with Ar as gas balance.

integrated system as shown in Fig. 3c, a similar result was observed as in the water free system. The major difference is NO_2-slip between 500 and 550 K, which is around 50% lower than that in the integrated system in the absence of water. Furthermore, the temperature of the observed highest oxidation rate in the presence of water shifts to around 30 K lower than that in the case in the absence of water.

3.2. Improved utilisation of NO_2 by means of filter configuration

Attempt to reduce the NO_2-slip from the integrated system has been done by employing a configuration consisting of integrated system and a wall-flow monolith filter. For this filter configuration two experiments were performed. The first experiments used only NO + O_2 as feed gas (no H_2O vapour), while in the second experiment 3 vol.% H_2O was added into the feed.

The emissions of CO + CO_2 of these experiments are presented in Fig. 4a. Data from the integrated system and separated system are incorporated to make an honest comparison. Meanwhile the NO_2 concentration as well as the base case NO_2 and thermodynamic equilibrium are plotted in Fig. 4b. Experiments with TUD filter simulates the combination of integrated and separated system.

Among these three systems, the integrated system shows the highest rate of CO + CO_2 emission, while the TUD filter without H_2O has the same temperature at the maximal oxidation rate (around 10% lower than that in the integrated system), only the shape of the oxidation profile is somewhat less sharp. The lowest oxidation rate is observed for the separated system which is about 55% of the TUD filter system. Unfortunately, high CO + CO_2 emission rate of the integrated system is accompanied by the highest NO_2-slip, while the separated system emits the lowest NO_2-slip but also lowest CO + CO_2 emission. The TUD filter system has the same NO_2-slip as in the separated system when the major part of the soot is oxidised. In the TUD filter system, a tremendous effect of water is, once again, observed. The effect starts already at 475 K. The lowest NO_2-slip is observed at 550 K which corresponds to the highest CO + CO_2 production.

3.3. CO emission

The possible CO emission by all systems has been evaluated as illustrated in Fig. 5. In the integrated systems, both with and without water, the production of CO is hardly observed. For the TUD filter around 5–10% of soot oxidation products is CO, whereas for the separated system (miniaturisation of the CRT) this CO value was around 20%. The CO/(CO + CO_2) ratio decreases at medium temperature window (500–600 K) and this ratio increases when almost all the soot is oxidised.

4. Discussion

4.1. Basic utilisation of NO_2 in Pt/SiC foam system in the oxidation of soot

Fig. 2 demonstrates that soot oxidation can take place either with O_2 or with NO_2. Significant soot
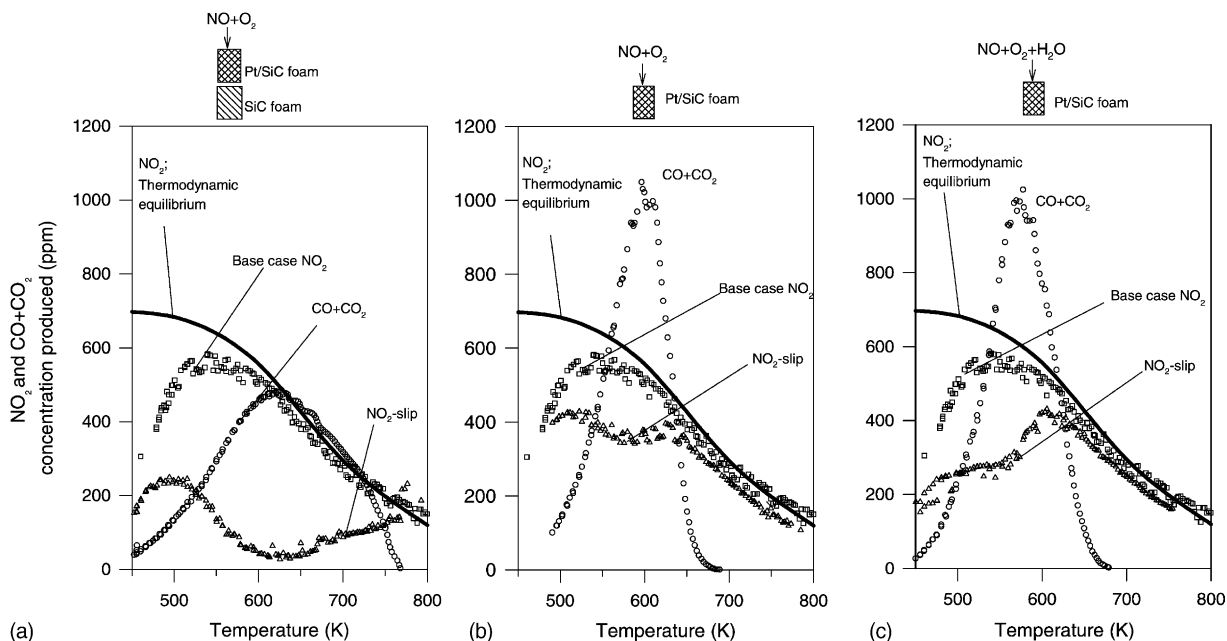
Fig. 3. $CO + CO_2$ and NO_2-slip production in different catalyst configurations and gas composition; temperature programmed reaction; heating rate of 0.2 K/min Ar as gas balance; (a) separated system with 10% $O_2 + 700$ ppm NO, (b) integrated system with 10% $O_2 + 700$ ppm NO, (c) integrated system with 10% $O_2 + 700$ ppm NO + 3% H_2O.

oxidation rate with O_2 only takes place at relatively high temperature, at our experimental conditions ≥ 675 K. With NO/O_2 a significant reaction rate is observed at 500 K and higher.

By using $NO + O_2$ as feed gas, two configurations of platinum–soot give rise to a different oxidation profiles. In the separated system, Pt/SiC foam is placed upstream of the soot loaded on a wall-flow monolith and functions only as NO_2 generator. NO_2 produced then enters the wall-flow monolith loaded with soot. In this compartment soot oxidation with NO_2 is taken place. In these circumstances, NO is reproduced and not further utilised. This process is the miniaturisation of the commercially available CRT system.

In the integrated system, soot is loaded on Pt/SiC foam catalyst. By this configuration, soot particles are randomly distributed along the Pt/SiC foam. As a result, NO reproduced from soot oxidation with NO_2 can go through the reaction cycles, and thereby, oxidises soot multiple times. This explains that the oxidation rate of soot loaded in in situ NO_2 generator is much higher than that in NO_2 generator placed upstream of soot configuration. This effect is similar

to the oxidation of soot with $NO + O_2$ gas mixture in a fixed bed containing a physical mixture of soot and Pt/alumina catalyst [6].

For soot oxidation utilising NO_x in the gas stream, the production rate of NO_2 is critical. The formation of NO_2, reaction (3), will initiate soot oxidation reaction, reaction (1). At our experimental conditions NO_2 production reaches the thermodynamic equilibrium above 600 K, see Fig. 3. At higher temperatures NO_2 production decreases due to the thermodynamic equilibrium.

Around 500 K the base case NO_2 is significantly high (about 550 ppm), but in the separated system, Fig. 3a, NO_2-slip amounts to half of this value (250 ppm). At temperatures up to 650 K the NO_2 reacts with increasing efficiency in the soot oxidation and a decreasing NO_2-slip is observed. The observation of NO_2-slip demonstrates that only a part of the generated NO_2 was used to oxidise soot. Above 675 K the NO_2-slip increases due to less soot available in the system. At this point the soot conversion is about 80%. It should be noted that the soot oxidation is a batch process as described in the experimental procedure.
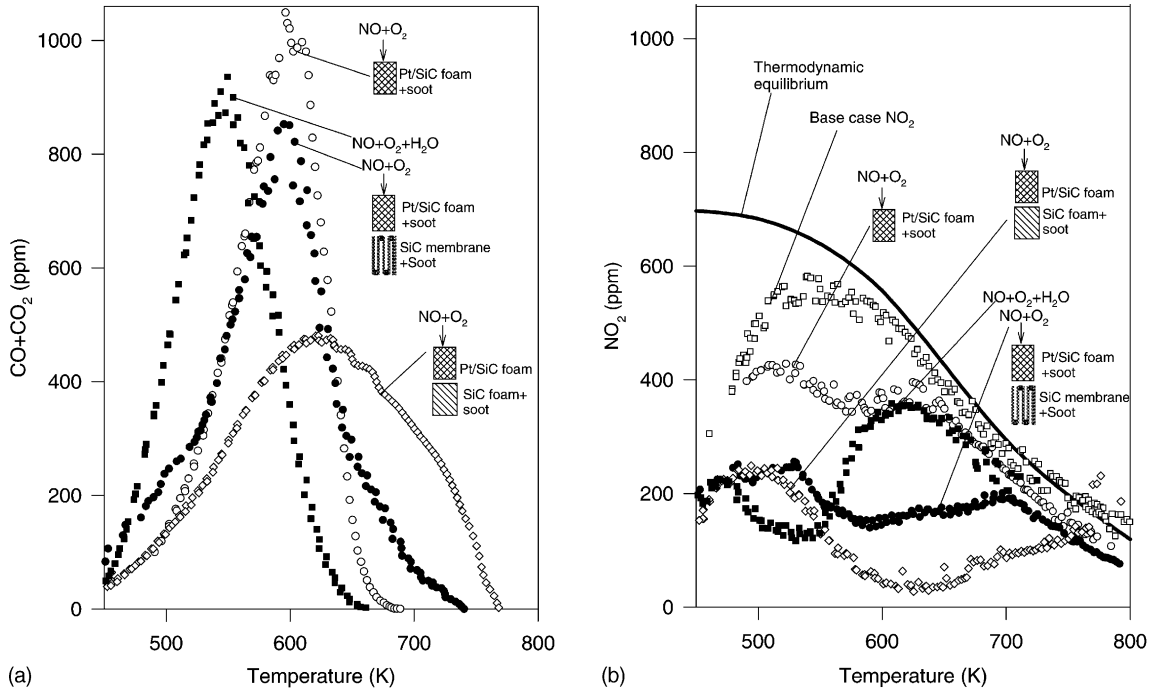
Fig. 4. Comparison between integrated system, separated system, and TUD catalytic filter performances; demonstration of an optimal usage of NO_2 in TUD catalytic filter; (a) $CO + CO_2$ production, (b) NO_2-slip.

The optimal NO_x assisted soot oxidation requires a defined temperature window. At low temperature the oxidation rate of soot with NO_2 is relatively low, while at higher temperature the production of NO_2 is limited by thermodynamic equilibrium. At intermediate temperature, more NO_2 is formed and the reaction rate of soot with NO_2 is high. At higher temperature, above 750 K, O_2 is the main reactant for diesel soot oxidation. For practical application, this catalyst temperature window is very critical and needs to be in the range of the average exhaust temperatures.

In Fig. 3b, the performance of the integrated system is displayed. The system simulates a catalytic trap, performing two functions at the same time viz. the generation of NO_2 as well as serving as reactor for soot combustion. It is clear that starting at 520 K, the $CO + CO_2$ released is much higher than the difference between the base case NO_2 and NO_2-slip. Clearly, NO is used multiple times. This explains the significantly higher oxidation rate of soot loaded on Pt/SiC foam compared to the rate of Pt/SiC foam upstream of the soot-trapping configuration.

In the integrated system, the random distribution of soot and platinum particles as well as the SiC foam structure are favourable for the multiple usage of NO. The foam texture, consisting of large cell, enhances turbulence and produces good mixing. These features suggest significant advantages for the process limited by mass or heat transfer like soot oxidation [7,8].

The oxidation rate becomes even higher when 3% H_2O is injected into the gas stream, as illustrated in Fig. 2. Cooper and Thoss [9] observed the same phenomena and proposed that the increase in soot oxidation rate by water addition might be due to the facilitation of NO_2 adsorption on the active site of soot particles. In agreement with their proposal, in the low temperature regime the amount of NO_2-slip is less than the system without water, if one compares Fig. 3b and c. The presence of water, therefore, might help the adsorption of NO_2 on the soot surface. Adsorbed NO_2 can then initiate soot oxidation at lower temperature. As a result, in the presence of water the soot oxidation onset temperature is lower. Other possible explanation of increasing soot oxidation rate by the presence of
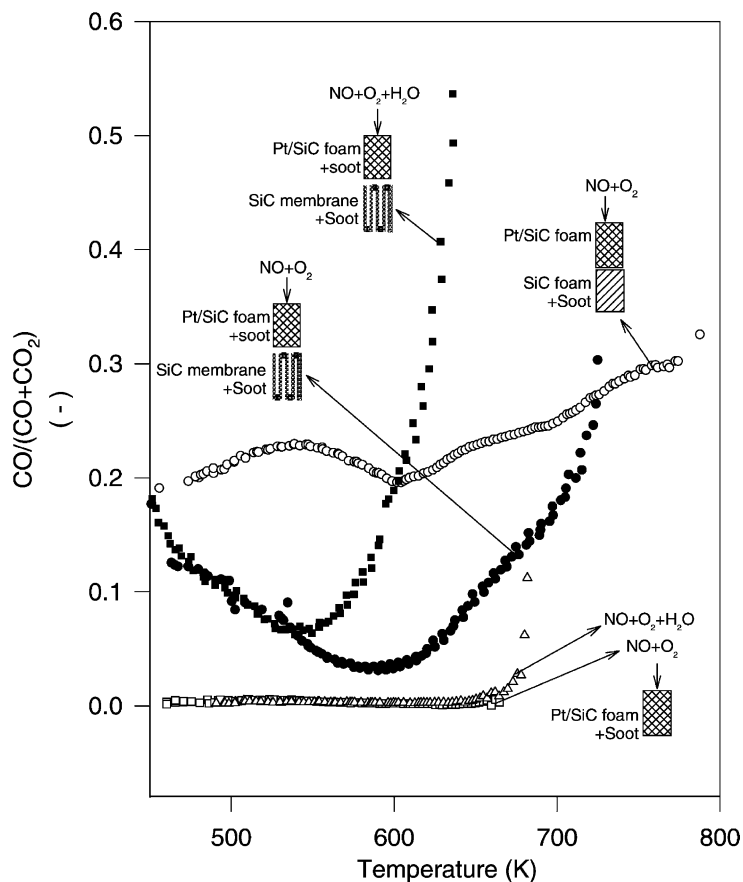
Fig. 5. $CO/(CO + CO_2)$ ratio of different catalytic filter configurations as observed in the temperature programmed reaction experiments. For details see Table 1.

water is that water takes a role in the decomposition of surface oxygen complexes (SOCs), presumed as intermediate in the oxidation of diesel soot [10]. The fact that H_2O in the absence of NO/NO_2 also increases the oxidation of soot with oxygen [11] supports the explanation of SOC's decomposition mechanism.

4.2. Advanced utilisation of NO_2 in the TU Delft filter system

From the Section 4.1, it is clear that NO_x assisted soot oxidation can operate in two basic modes. Firstly, it works as the CRT system and, secondly, it gives NO a catalytic role, so that the recycle reaction, reactions (1) and (3), can take place as in Pt/ceramics foam. Nevertheless, if only Pt/ceramic foam catalyst

is applied as catalytic filter, there will be two major disadvantages. The first disadvantage is the limitation in filtering efficiency and the second one is the occurrence of NO_2-slip. Depending on the geometry and test conditions, the filtering efficiency of ceramic foam will only be in the range of about 40–70% [12]. NO_2-slip might be considered as secondary emission. As demonstrated in the separated system (CRT system) has a low NO_2-slip at the expense of the low oxidation rate. In an application test of CRT technology in the city buses, a 20% NO_2-slip was observed [13].

To circumvent these limitations, the so-called TUD catalytic filter was designed and developed. As explained in the experimental part, the filter candidate consists of cylindrical Pt/SiC foam that is placed upstream of SiC membrane wall-flow monolith. By

design, the filter should capture diesel particulate matter in two stages. Firstly, the particulates are partly trapped in Pt/SiC foam and, secondly, the rest is trapped on SiC membrane monolith. In SiC membrane the total filtration will take place. In the Pt/SiC foam compartment, NO_2 production and the recycle reaction of NO to NO_2 coupled with soot oxidation are taken place. In the SiC membrane, the rest of trapped soot is oxidised by NO_2-slip released from the Pt/SiC foam.

The results, as can be seen in Fig. 4b, show that in the absence of water lower NO_2-slip can be observed than the one of the integrated system when soot is loaded on both Pt/SiC foam and SiC membrane in the TUD filter system. Roughly, the system reduces 50% of NO_2-slip emitted from the integrated system and the slip is at the same level as that in the separated system. It is clear that in Pt/SiC foam compartment, NO is used several times while the downstream SiC membrane functions as a CRT system, effectively reducing the NO_2-slip from Pt/SiC foam. Although, the total 200 mg soot is equally distributed in two compartments with two kinds of reaction modes, the overall oxidation rate lies rather close to the rate of the integrated system.

In the presence of water a lowest NO_2-slip is observed at low temperatures, this temperature corresponds to the highest oxidation rate indicated by highest $CO + CO_2$ as shown in Fig. 4a. At higher temperature the amount of NO_x-slip increases due to high conversion levels of soot (e.g. almost no soot will be available).

4.3. CO emission

Fig. 5 illustrates that Pt-soot reactor configurations have a large impact on the composition of CO and CO_2. It is no surprise to observe a 100% CO_2 yield in the integrated system. In this system, once a CO molecule is formed, it will be followed by CO oxidation to CO_2 catalysed by platinum sites distributed along the reacting soot. This so-called secondary emission of CO will occur in the separated system. In the mechanism of soot oxidation, which is initiated by the formation of surface oxygen complexes (SOCs) as intermediates as proposed in [10,14], CO and CO_2 can be formed from thermal decomposition of SOCs, or the reaction of SOCs with oxygen [15]. In the separated system CO_2 can only be formed from this

so-called primary and secondary emission, while CO from secondary emission could not oxidised further to CO_2 as in the integrated system. The high abundance of O_2 in the system, 10%, helps the formation of CO_2 in its reactions with SOCs.

Considering CO emissions, the TUD system has a better performance than the separated system (the CRT system), where the CO emissions are around 20–25% of the formed soot oxidation products. Nevertheless, compared to the integrated system, the TUD filter produces more CO, about 5–10% of soot oxidation product. It should be noted that the calculated soot amounts to filter volume ratio for the simulated ECE15 + EUDC (see section d) reaches a steady-state value of 0.7 g soot/l filter. This ratio is about 10 times lower than that the value of 7 g soot/l substrate in these laboratory experiments. This can lead to higher NO_2/soot ratio, expectedly more favourable to form CO_2 from the intermediates (e.g. CO). Since the CO-production is also a function of the amounts of soot deposited it is anticipated that under practical conditions CO concentration in exhaust stream will be around 10 ppm, whereas in the laboratory experiment the observed levels are around 50 ppm. In conclusion, secondary CO emissions can be generated during the oxidation of trapped soot in a diesel particulates filter. The integrated system is in that respect preferred (no CO emissions). With TUD catalytic filter up to 50 ppm CO is formed as soot combustion product, whereas for the miniaturisation of the commercially available CRT system at the same diesel soot oxidation rate 120–140 ppm CO is observed.

4.4. Assessment of the TU Delft catalytic filter system application

For a passenger car in the European countries, the ECE15 + EUDC test cycle is applied for the certification of the Euro IV standard. The test cycle is the

Table 2
Summary of ECE15 + EUDC test cycle

Test cycle	ECE15	EUDC
Maximum speed (km/h)	50	120
Average speed with idling (km/h)	18.7	62.6
Distance (km)	4.052	6.955
Duration (s)	780	400

Table 3
Simplified ECE15 + EUDC test cycle temperature results and soot loading calculation in one cycle of ECE15 + EUDC test

Step (cycle)	Duration (s)	Distance (km)	Average Temperature (K)	Oxidation rate (μg/($g_{initial}$ s))	Estimated process	Soot deposited (g)	Soot burnt (g)	Remaining soot (g)
A (ECE15)	780	4.052	<500	Negligible	Soot deposition	0.2067[a]		
B (EUDC)	40	0.696	<500	Negligible	Soot deposition	0.0355		0.2422
C (EUDC)	160	2.782	500	18	Soot oxidation		0.0282[b]	
					Soot deposition	0.1419		0.3558
D (EUDC)	160	2.782	550	36	Soot oxidation		0.0835	
					Soot deposition	0.1419		0.4143
E (EUDC)	40	0.696	<500	Negligible	Soot deposition	0.0355		0.4498

[a] Soot deposited (g) = distance (km) × 0.051 (g/km).
[b] Soot burnt (g) = remaining soot from previous step (g) × oxidation rate (μg/($g_{initial}$ s)) × 60% × duration (s) × projected filter volume (2 l)/experimental filter volume (0.0294 l).

simplified representation of a typical European urban centre and a drive at the country side/high way and is summarised in Table 2.

The introduction of wall-flow monolith as diesel particulate filter significantly suppresses diesel particulate emission to a very low level. Filtration efficiency up to 97% are reported in [16,17]. The problem is whether the regeneration can take place in the exhaust condition in order to keep the back pressure at an acceptable level. One of the difficulties is the exhaust temperatures becomes lower and lower due to the advances made in diesel engine combustion technology. Analysing the data of ECE15 + EUDC tests shows that the exhaust gas temperature downstream of the turbine during the ECE15, and the first 40 s of the EUDC is mostly lower than 500 K [18,19]. In the last part of the test the temperature is mostly higher than 500 K. Table 3 summarises the conditions during the test. This information is then adopted to calculate the soot deposited and burnt during a series of the test if the TUD filter would be applied in a passenger car. The calculations are done as follow.

As a case, a passenger car specified having a particulate emission of 0.051 g/km has been chosen. This is an emission from a typical new passenger car [5]. The filter volume is estimated to be 2 l. It is assumed that 50% soot is deposited on Pt/ceramic foam and the rest is deposited on SiC membrane. The oxidation rates are based on Fig. 4a when water is present. Unfortunately, in the test cycle NO$_x$ emissions data are presented in gram per kilometre without explicit report as function of temperature, for example in [20]. Therefore, direct comparison with NO$_x$ data is not straightforward. However, information is available for

in-cylinder NO$_x$ emission of a passenger car measured in steady state. The NO$_x$ emission varies between 0 and 800 ppm [21]. Because the real exhaust conditions are most likely less favourable than our laboratory conditions, we estimate that the oxidation rate should be decreased to 60% to adjust for practical conditions. It should be noted that the GHSV in the ECE15 + EUDC is around 50.000 l/(l h), and in our laboratory experiments is around 65.000 l/(l h) and the applied catalytic efficiency of 60% for the TUD filter could be realistic.

The calculation was continued for the 2nd until the 25th series of the cycle, the soot remaining in the previous series is accumulated and added as deposited soot in the next series. At the first 20 series the soot loading increases. Starting at series 21st, equilibrium is achieved at the soot loading of 0.7 g/l. This amount is acceptable in relation to the back pressure to be expected, as shown in [22]. This could be an indication that the system is promising. It should be noted that sulphate formation from exhaust SO$_2$ could worsen the catalyst performance. However, tomorrow sulphur free diesel fuel will lead to essentially SO$_2$ free diesel exhaust.

5. Conclusions

1. The use of Pt/SiC foam as NO$_2$ generator and reactor for the oxidation of soot tends to produce a secondary emission due to NO$_2$-slip.
2. Comparing the separated system with the integrated system, the former produces low amount of NO$_2$-slip with a low oxidation rate. The integrated

system can, however, lead to a high soot oxidation rate with at the same time a high NO_2-slip.

3. An improvement on the utilisation of NO_2 has been observed in the TUD catalytic filter. In this filter soot oxidation takes place in Pt/SiC foam as an integrated system and in SiC membrane as a CRT system. The oxidation rate of TUD catalytic filter is 90% of that of the integrated system with effectively lower NO_2-slip.

4. In TUD catalytic filter as in all diesel particulates filters secondary CO emissions take places (with the exception of the integrated catalytic system). The CO emission of the TUD filter is 2–3 times lower than the CO emissions of the commercially available CRT at identical diesel soot oxidation rates.

5. Calculation shows that steady-state diesel soot loading on the TUD catalytic filter during 25 series of ECE15 + EUDC cycle is reached and is low (0.7 g/l); this will lead to a low back pressure when applied in the passenger car. Therefore, the system might be promising for future application.

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