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Potential strukturierter Metallträger und fortschrittlicher Beschichtungen, getestet an einem EU IV 2,2 l Dieselfahrzeug

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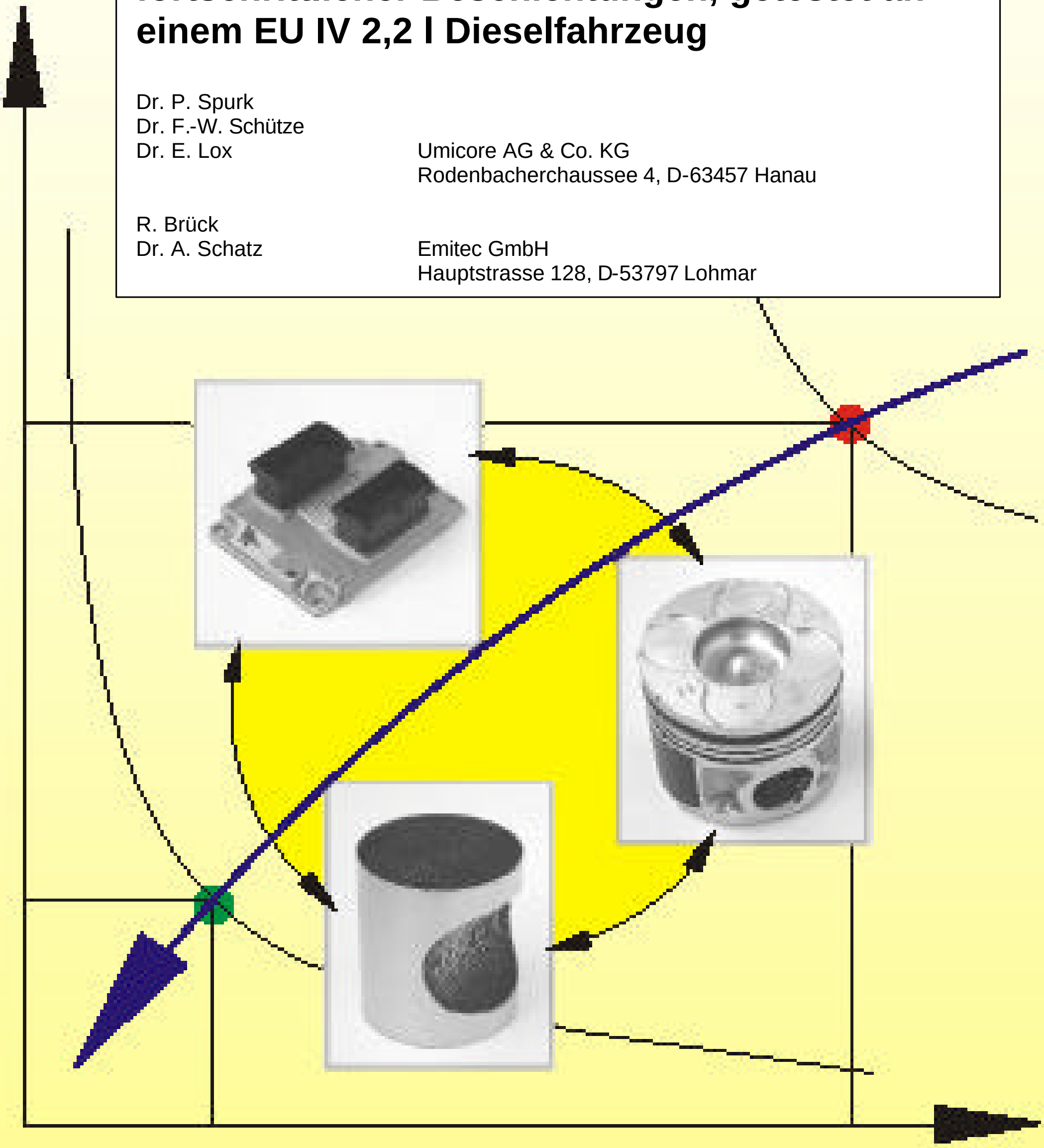

Lehrstuhl Verbrennungsmotoren

Potential of Structured Metal Substrates and Optimised Catalysts, tested on an EU IV 2.2 l Diesel passenger car

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Kurzfassung

Der steigende Anteil an Dieselfahrzeugen in Europa in Verbindung mit sich verschärfenden Abgasgrenzwerten steigert die Nachfrage nach hocheffizienten aber kostengünstigen Katalysatorsystemen, um die Wettbewerbsfähigkeit des Diesels gegenüber dem Ottomotor und den Hybridantrieben zu gewährleisten.
Im Folgenden wird die schrittweise Optimierung eines bestehenden EU IV Serien Katalysatorsystems in Richtung Effektivität und Kosten aufgezeigt. Innovative metallische Trägerstrukturen in Verbindung mit neuen fortschrittlichen Beschichtungen gewährleisten eine hohe volumenspezifische, katalytische Effektivität.

Abstract

The growing number of Diesel cars in Europe in combination with more stringent emission legislation increase the demand for high efficient, but cost-effective catalyst systems to guarantee the competition of diesel engines compared to SI-engines and hybrid vehicles.
In the following paper the stepwise optimization of a production EU IV catalyst system will be demonstrated. Innovative metallic substrates in combination with advanced catalytic coatings ensures a high volume specific efficiency.

1. Introduction

An analysis of new car registrations in Germany shows that the number of vehicles with diesel engines is increasing constantly. While over 10 years ago performance-oriented drivers would not even consider a diesel, these vehicles stand out today with performance data that are capable of measuring up to gasoline-powered vehicles. Additional benefits are the much lower fuel consumption and thus also reduced CO2 emissions of diesel vehicles. From driving comfort to „driving fun," diesel vehicles are fast approaching equivalency with gasoline-powered vehicles, which explains the increase in the number of registrations.

Beginning in 2005 all passenger cars and light utility vehicles in Europe must meet the EU IV limits. For vehicles with SI and diesel engines, this means a reduction in the controlled emissions of up to 50%. Many models are already being offered today as EU IV versions to take advantage of the tax benefits in Germany. Especially in diesel engines, a substantial reduction in raw emissions can be achieved with measures such as improved injection systems, controlled and cooled EGR systems, and optimization of the combustion process. Figure 1 shows an overview of currently certified diesel vehicles [14].

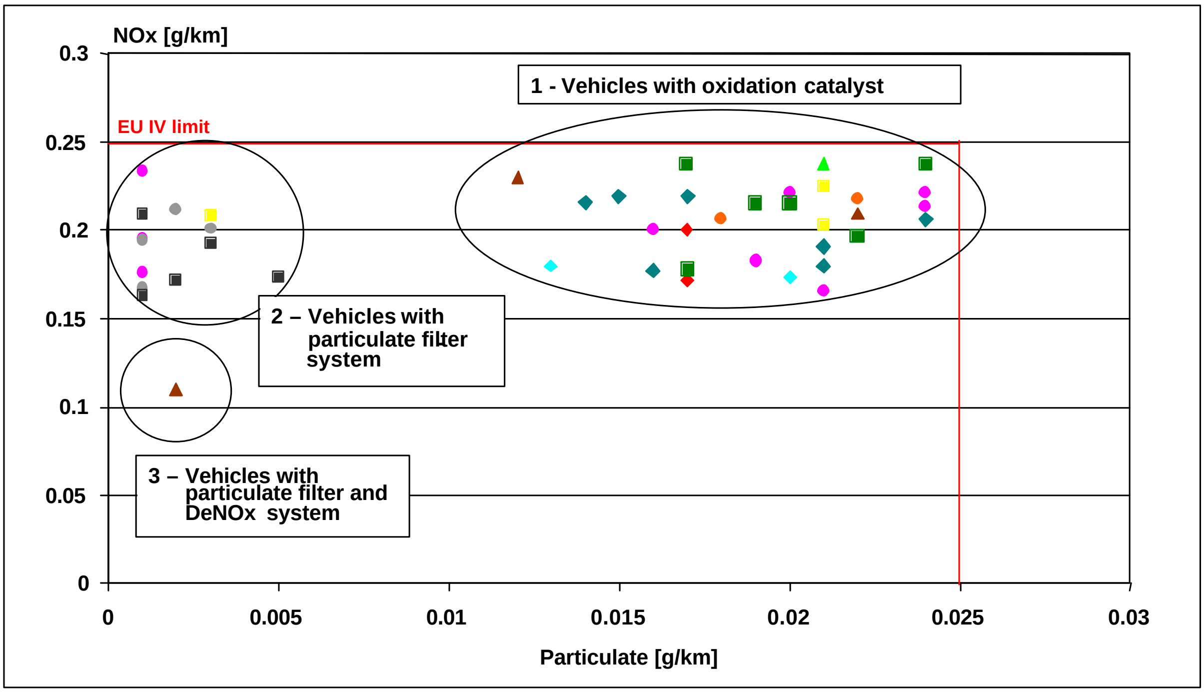

Figure 1: VCA certified EU IV vehicles; status: 6/2004

An analysis of the certification data shows in dependence on the raw engine emissions and the marketing of different exhaust gas aftertreatment concepts. In all vehicles an oxidation catalyst was used to reduce carbon monoxide (CO) and hydrocarbon (HC) emissions. Because of the low raw engine emissions, an additional filter was not installed (Group 1). A particulate filter was used, on the one hand, in engines with high particulate emissions and, on the other hand, for political, marketing-strategy reasons (Group 2). Identified as a third group are vehicles that use a particulate filter as well as a concept for reducing nitrogen-oxide (NOx) emissions. Here a distinction must be made between systems that require active regeneration measures such as NOx absorbers and systems with passive NOx reduction („lean NOx conversion“). The passive systems use a modified oxidation catalyst that achieves a limited NOx conversion by means of the CO and HC contained in the exhaust gas. Due to the reciprocal reactions of CO and HC oxidation with oxygen and the NOx reduction with HC, this reaction functions only within a restricted temperature range.

Even vehicles in the upper and luxury classes such as the Audi A8 3.0 l TDI and the Honda Accord 2.2l i-CTDi undershoot the EU IV threshold values by up to 50% using purely passive systems without particulate filters. Based on an analysis of the current literature and the anticipated technological steps in the development of diesel engines, further reductions, particularly of NOx and particulate emissions (PM), in the magnitude of 30% can be expected over the next few years [1, 2, 3]. In direct comparison to catalyst systems of SI and diesel engines, it was noted that most of the catalysts (oxidation) in diesel vehicles are larger and coated with a higher precious metal content. This results in increased cost pressure in diesel vehicles even without particulate filters.

It is unclear which systems should be used in the future, as the EU V threshold values are still in controversy at the present time. In particular the question as to whether an active nitrogen oxide aftertreatment will be necessary depends on whether the threshold values of 0.25 g/km (EU IV) will be lowered by 20% to 0.2 g/km or by 68% to 0.08 g/km. The same applies for particulate emissions, where threshold values between 0.0125 (50% lower than EU IV) and 0.0025 g/km (10% lower than EU IV) are in discussion.
The increase in the number of diesel vehicles of currently almost 45% in Germany reinforces the need to develop affordable catalyst systems to ensure the competitiveness of diesel engines also in the future.
The aim of the studies described below was to identify lower-cost alternatives to the series-produced system (precious metal loading, substrate costs). For this reason, new metal substrates and adapted oxidation catalysts were used and measured in the new European driving cycle (NEDC) in new condition and after aging.

2. Planning of the test

The new Honda Accord was selected as the test vehicle because it is equipped serially with metal substrates. The 2.2l I-CDTi Honda Accord available in Europe uses a newly developed 4-cylinder diesel engine with 2.2-l engine displacement and a power output of 103 kW at 4000 rpm. The engine has a common rail injection system, exhaust-gas turbocharging with charge-air cooling and a cooled exhaust gas recirculation system. The compression ratio is 16.7. The serial catalyst system consists of a total of three catalysts. Upstream a catalyst with a capacity of 0.7-l (Ø 98.4 x 95) and a precious metal loading of 90 g/ft^3 is used. Two catalysts are installed downstream (1.0 l and 1.2 l) with a precious metal loading of 90 g/ft^3 and 54 g/ft^3. Honda already demonstrated the potential for optimization through innovative metal substrates in 2003 [4]. Based on these results, a test program was created that contained both specific substrate designs and different coatings. In addition to the optimization of costs, the emissions potential was also to be examined with respect to future threshold values.

2.1 Measuring the serial condition

The test program was matched to the temperature boundary conditions. Figure 2 shows the temperature of the gas before the upstream catalyst (CCC) and before the downstream catalyst (UFC).f

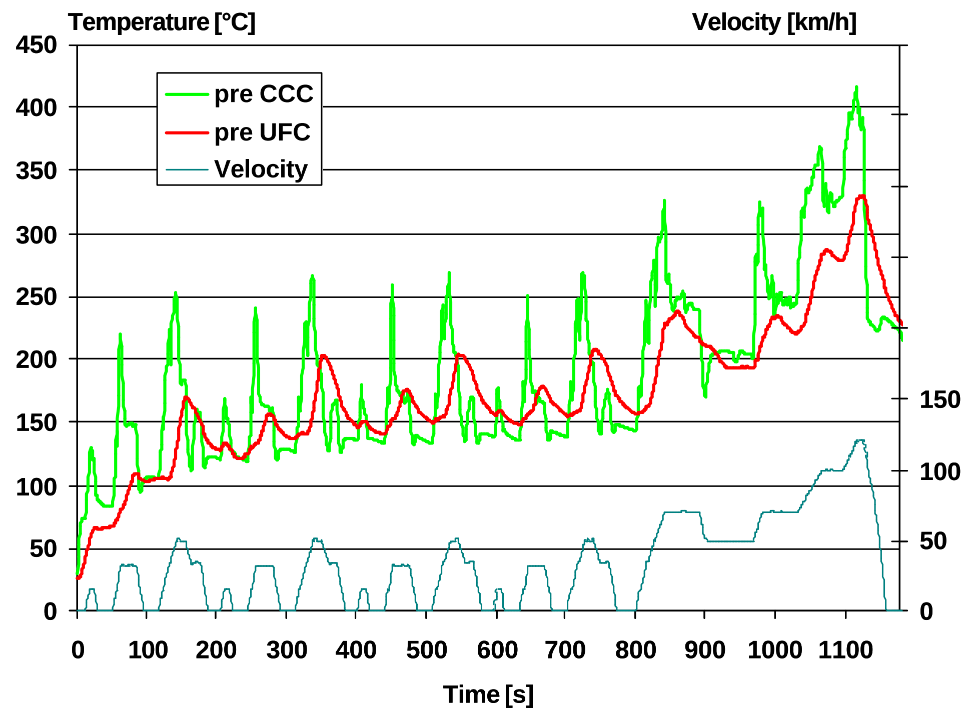

Figure 2: Gas temperature before upstream catalyst (CCC) and before downstream catalyst (UFC) in the NEDC of the 2.2l i-CDTi Honda Accord

Even though the exhaust gas temperature was higher compared to other vehicles in the same performance class, one of the greatest challenges in the exhaust gas aftertreatment of modern diesel vehicles was also shown here. Due to the very good engine efficiency, the exhaust gas temperature is at a low level. As a result, the exhaust gas temperature, for example in the NEDC cycle, is around the light-off temperature of the catalyst. This requires very careful design of the catalyst technology to be used to achieve the desired emissions even after aging.

Figure 3 shows the HC, CO, and NOx emissions of the vehicle measured in the raw exhaust gas and after the serial catalyst system. While the raw emissions of HC and CO, at 0.520 and 1.784 g/km respectively, tended to be on the high side, the NOx emissions, at 0,158 g/km, were well below the EU IV threshold value.

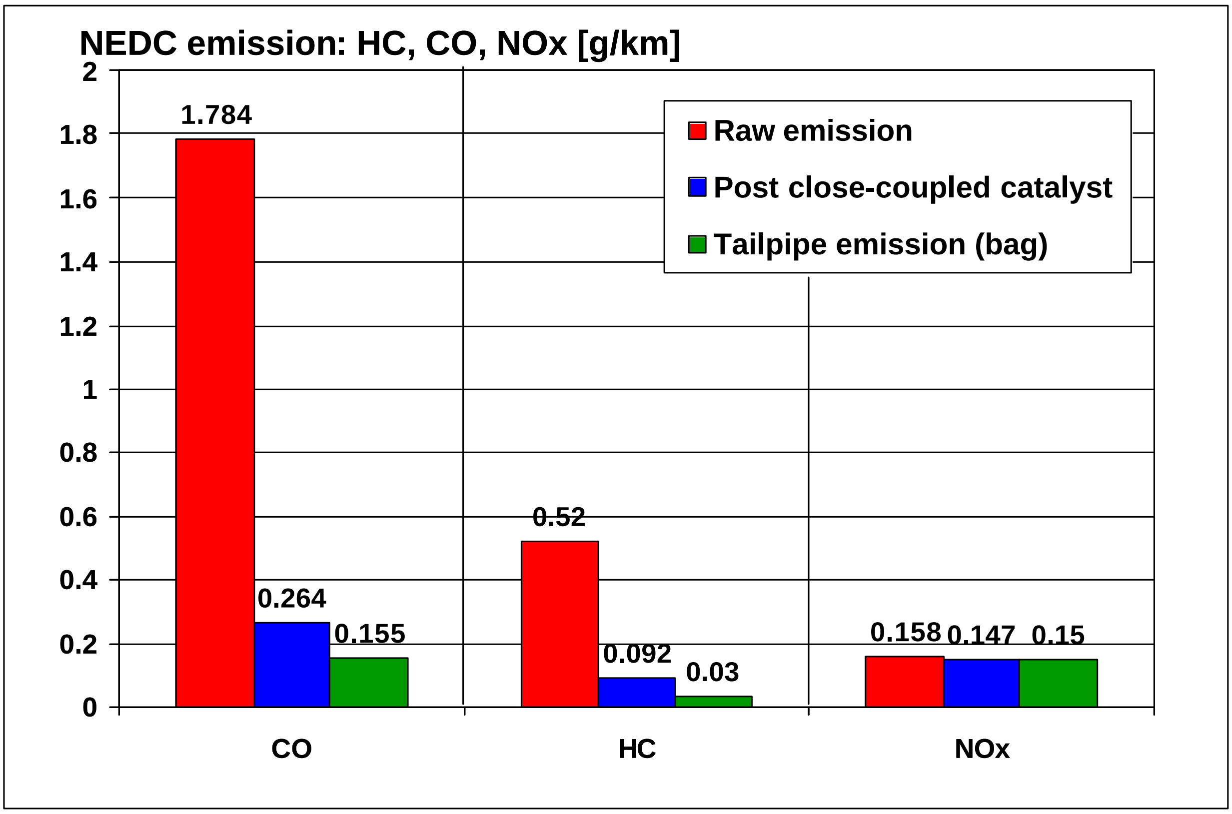

Figure 3: HC, CO, NOx raw emissions in the NEDC; modal values

The new serial system demonstrated very good efficiency, with conversion rates of over 90% for CO and HC. Even the particulate emissions of 0.015 g/km, which are not shown here, were well under the EU IV threshold value.

2.2 Evaluated samples

2.2.1 Tested metal substrates

In correspondence with the boundary conditions and the developmental goal, the following metal substrate variants were used:

Hybrid catalyst:

Due to the above-described lower exhaust gas temperatures, the goal of the catalyst development must be rapid heating up and slow cooling-down of the catalyst. These contradictory requirements can be fulfilled by a catalyst substrate that has two different and separate structures. It consists in the front of a short disk with a small foil thickness and thus lower thermal capacity to ensure fast light-off. On the other hand, the matrix features a large foil thickness in the rear (high thermal capacity) for storing as much heat as possible and thus preventing the monolith from cooling down too quickly [5].

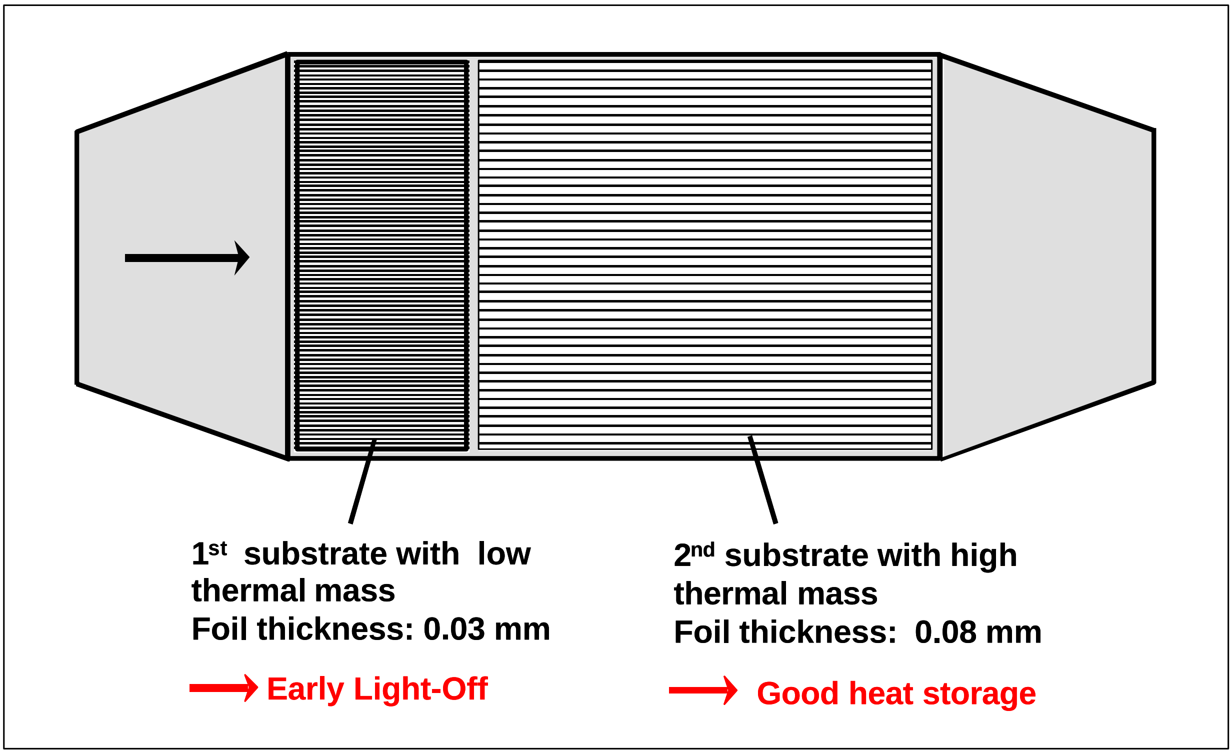

Figure 4: Operating principle of the hybrid catalyst

Pre-turbocharger catalyst (PTC):

Due to the energy conversion in the turbocharger, the position upstream of the turbocharger offers advantages with respect to the catalyst temperature in the test cycle. In a diesel engine, with its relatively low CO and HC concentrations, the oxidation processes are limited at an adequate temperature by the mass transport; i.e., by the effort of bringing the pollutant molecule out of the gas and to the catalyst (precious metal). The cross-section and therefore the capacity of pre-turbocharger catalysts are limited by the design space. The high flow rates in the channels result in a turbulent flow and thus an optimum mass transport. Relative to the position downstream of the turbine, the specific effectiveness of a PTC is approximately 10 times greater [6]. However, the fact that the PTC may influence the exhaust gas recirculation due to its backpressure behavior should be taken into consideration.

PM filter catalyst:

While the technologies described above may help improve the specific efficiency of the oxidation catalyst with respect to CO and HC emissions, they do not reduce particulate emissions. The so-called PM filter catalyst is an open separation system in which a part of the gas stream and therefore the particulate is routed by deflection shovels through metal-fiber fleece and filtered [8] [9]. The particulate deposited in the fleece can be oxidized in a pseudo-continuous process with the help of NO_2 and O_2.[11].

2.2.2 Tested catalysts

From the catalyst side, the use of metal substrates requires coatings that have been optimized specifically for these substrates. This is especially true for metal substrates that differ from „state of the art“ substrates with respect to their specification (PM filter catalyst) and their mounting position (PTC).

Particularly with PM filter catalysts, in addition to optimizing the catalyst the coating process must also be adapted to prevent clogging of the fleece or deflection shovels. The advantage of a coated PM filter catalyst - aside from reducing the required number of substrates – lies in the fact that regeneration with the catalytically coated filter can be effected at much lower exhaust gas temperatures and/or higher burn-off rates [12].

Figure 5 contains the results of a previous study that was performed on a 1.9-l direct-injection diesel engine. This study examined the filtration efficiency of uncoated and catalytically coated (precious metal loading: 50 g/ ft^3 Pt) filter catalysts in various stationary engine operating points.

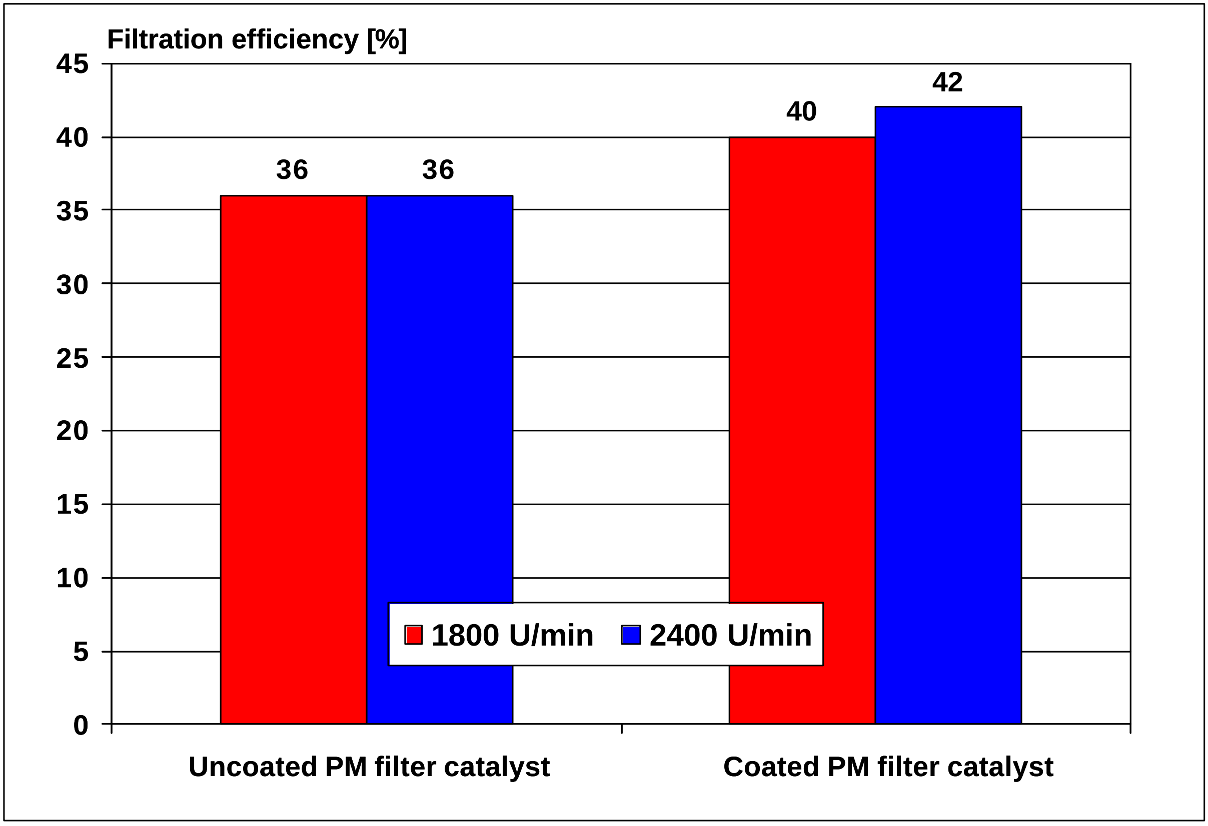

Figure 5: Comparison of filtration efficiency of an uncoated and a coated PM filter catalyst (AVL Smokemeter)

The results seen in Figure 5 show that the filtration efficiency can be retained with the appropriately optimized catalytic coating. Even from the catalytic viewpoint (light-off evaluation), the coated PM filter catalysts with standard oxidation catalysts demonstrated comparable results.

The systems used for this test series are listed in Table 1.

System	Pre-turbo catalyst		Upstream catalyst		Downstream catalyst 1		Downstream catalyst 2	
	Substrate	Coated	Substrate	Coated	Substrate	Coated	Substrate	Coated
1 Serial	-	-	Ø98,4x95mm; 400 cpsi	CCC 1 Serial (90g/ft^3)	Ø110x100mm; 400 cpsi	UFC 1 serial (90g/ft^3)	Ø110x130mm; 400 cpsi	UFC 2 serial (54g/ft^3)

2 Hybrid +UFC, serial	-	-	Hybrid-cat Ø98,4x95mm; 400 cpsi	CC1 (70g/ft³)				
3 Hybrid +UFC mod.	-	-			Ø110x130mm; 400 cpsi	UC1(90g/ft³)	-	-
4 PTC + Hybrid + UFC mod.	Ø36x42mm; 200 cpsi	PTC1 (100g/ft³)						
5 PTC + Hybrid + PM filter cat					PM filter cat. Ø110x130mm	PM1(90g/ft³)	-	-

Table 1: Test matrix

2.3. Conducting the Test

For the tests on the vehicle a 48“ compact roller dynamometer was available with a maximum braking power of 153 kW (sustained) or 258 kW (peak). The test rig allowed testing speeds of up to 250 km/h. Aerodynamic drag was simulated with a blower with the flow controlled in proportion to the speed of the vehicle. For control, monitoring, and data recording of the exhaust gas texts, the automation system PELE (PEUS) was used in combination with an INCA PC data recorder.

The emission tests were performed with the help of a variable Venturi CVS system (CVS 7200 S). Three modal measuring lines (MEXA 7500-D) were used, allowing a test of raw emissions, as well as of the emissions between two catalysts and after the entire catalyst system. At the same time a dilution tunnel was used to measure total emissions and particulate emissions during a test run. The gas emissions that were analyzed were: HC, CH_4, NOx, CO, CO_2, and O_2. Using CVS allowed a comparison between the integrated modal result and the total result of the bag measuring line and thus a check of the results. The particulate was sampled with a PMU 7000 from Horiba. Sulfur-free fuel (< 10 ppm) was used.

The vehicle emissions of all tested systems were evaluated over the standardized new European driving cycle (NEDC). In addition to being measured in new condition, the systems shown in Table 1 were also measured after aging. The systems were aged according to a procedure used by several OEM’s: the catalysts are aged hydrothermally in a continuous furnace with a gas flow of 0.3 Nm³/h and a relative humidity of 10 Vol.-% in air. In this case aging was carried out for a total of 70 h at a temperature of 550 °C.

3. Test results

The first results with a hybrid catalyst [4] were shown in comparison to the serial system already in 2003. The catalyst capacity and the catalytic coating were held constant. The improved thermal management led to a reduction of HC and CO emissions of 33% and 35%, respectively (Figure 6).

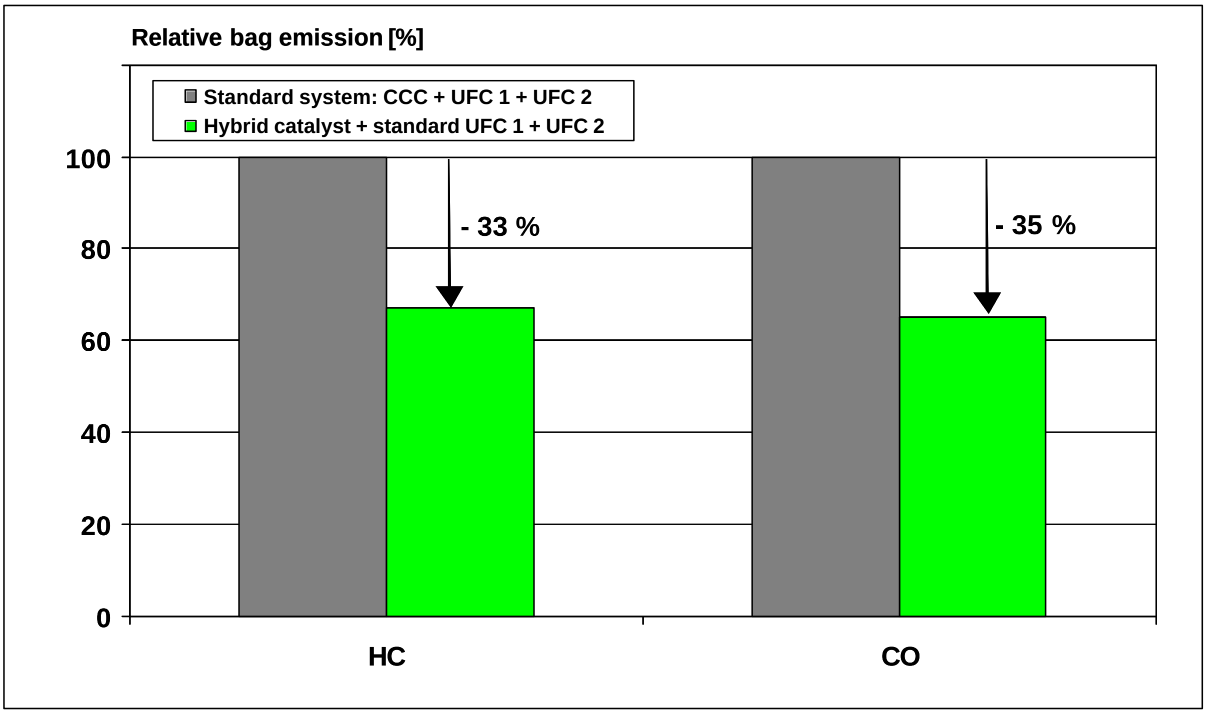

Figure 6: NEDC test results of the serial catalyst compared to the hybrid catalyst [4]

Based on these experiences the tests were designed specifically with a view toward cost savings potential and reductions in emissions.

3.1 Results of measurements in new condition

3.1.1. Hybrid catalyst with optimized coating CC1 (System 2)

As described in Table 1, the upstream serial catalyst was replaced with a hybrid catalyst CC1 in a first test series. Figure 7 shows a comparison of the exhaust gas temperature histogram after the serial upstream oxidation catalyst and after CC1. Due to the higher thermal mass, CC1 requires somewhat longer to heat up, which can be seen in Figure 7 in an increased portion in the temperature range below 100°C. As soon as the system is heated up, the hybrid catalyst CC1 allows better storage of the heat. This is manifested in the higher exhaust gas temperature portion in the range between 200°C and 250 °C.

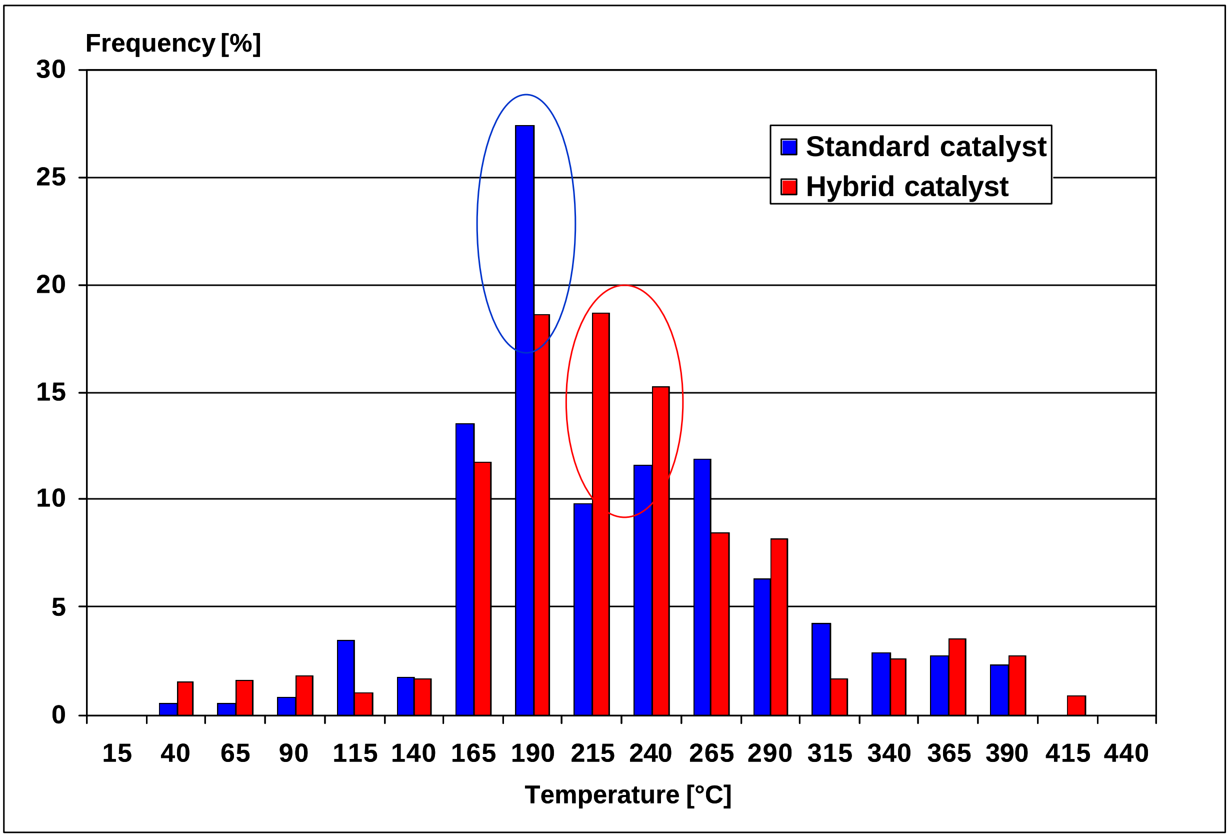

Figure 7: Temperature histogram determined after serial CCC and hybrid cat CC1

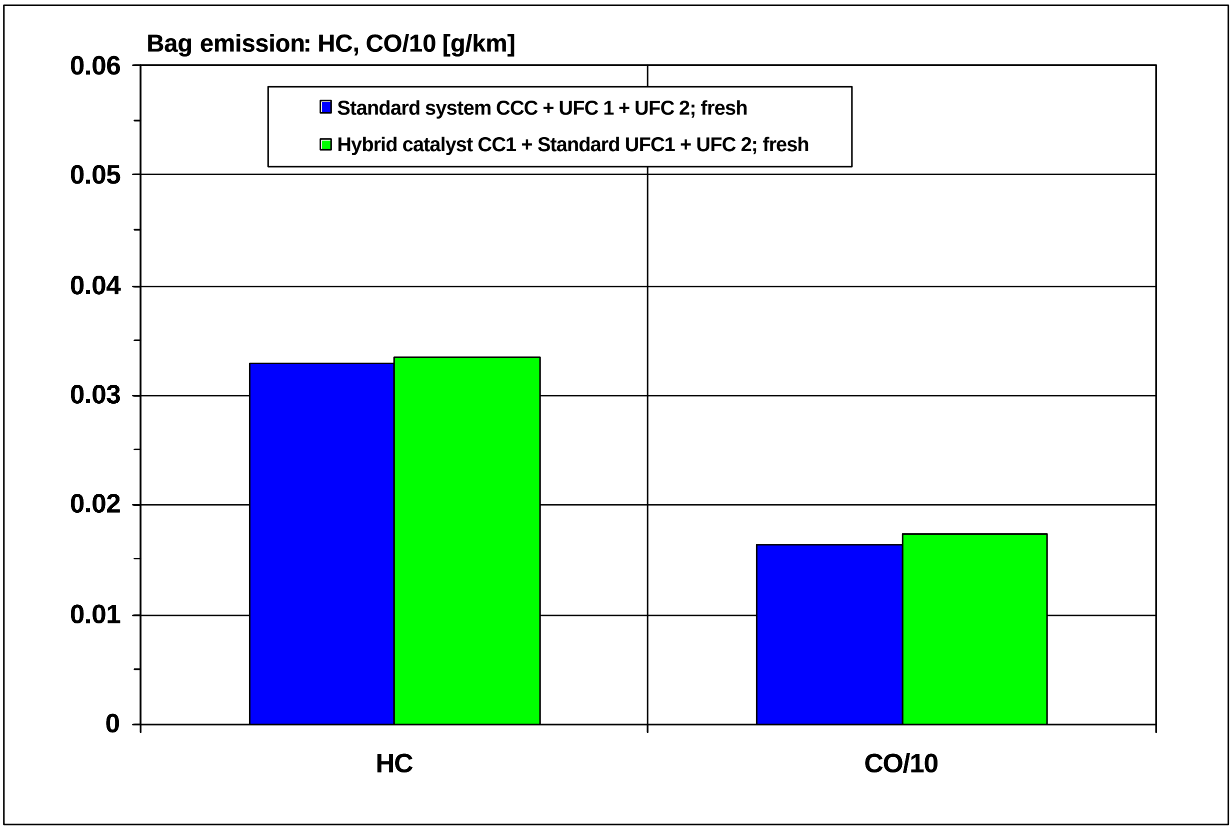

Figure 8: Comparison of HC and CO emissions of the serial system (System 1) to the hybrid catalyst with optimized coating (System 2)

Figure 8 shows the HC and CO bag results obtained in the NEDC with the hybrid catalyst CC1 (System 2) compared to the serial system (System 1).

Despite the approx. 30% reduction in the precious metal content, the results obtained with System 2 were comparable to those of the serial system. This applies equally to the results for NOx and particulate, which are not shown here.

3.1.2. Hybrid catalyst with optimized coating CC1 and new downstream catalyst UC1 (System 3)

For further optimization of the catalyst system, in an additional step the two serial underfloor catalysts UFC1 and UFC2 were replaced with the catalyst UC1. Special emphasis was placed in this case on the NOx emissions, since the catalyst UFC2 is designed as a so-called HC-DeNOx catalyst [13]. The NEDC results are summarized for the gaseous emissions HC, CO, and NOx in Figure 9. Despite a reduction of the catalyst capacity by approx. 45% and of the precious metal loading by approx. 30%, the results showed only a slightly increased value for the CO and HC emissions Figure (9). The particulate emissions, at 0.0146 g/km, were on a level comparable to that of System 1. For further optimization with respect to exhaust gas emissions, in the next step of the test the previously discussed pre-turbocharger catalyst PTC1 was installed and measured – under consideration of the fact that this vehicle was not calibrated for the use of a PTC.

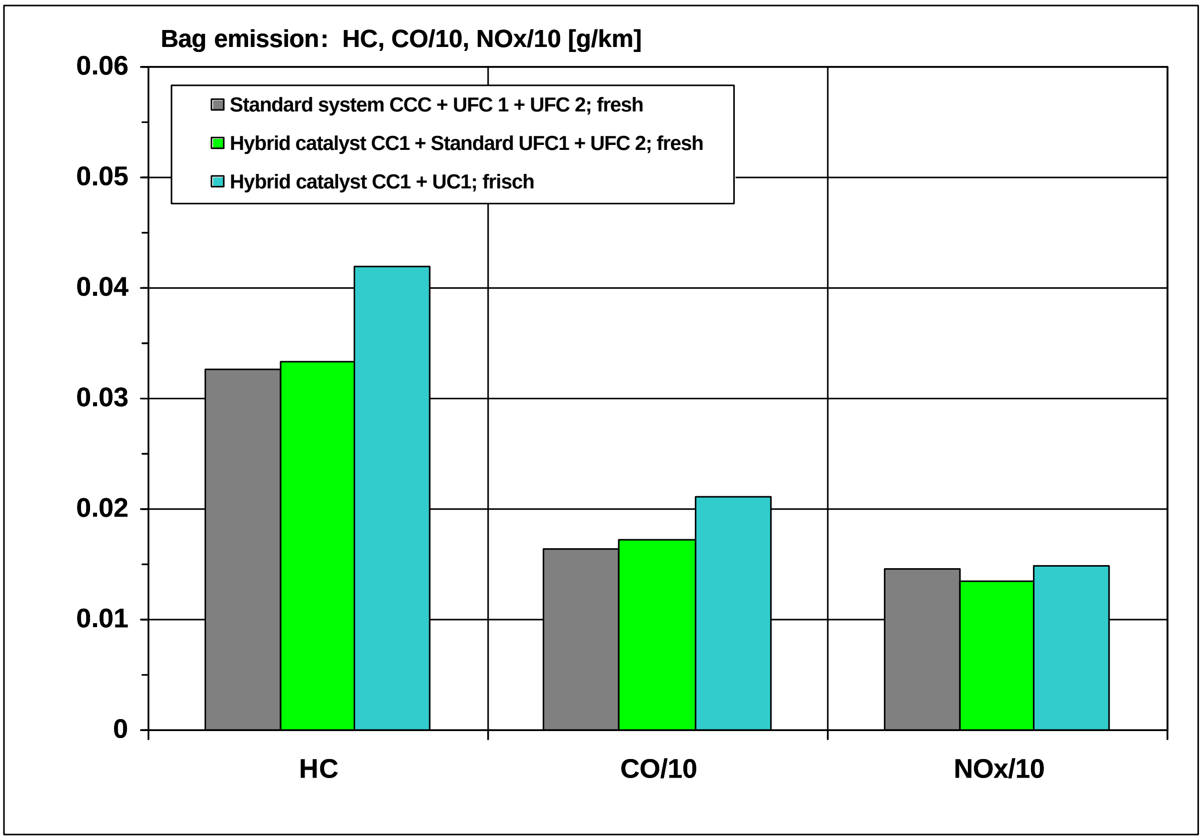

Figure 9: HC, CO, and NOx results for the different tested systems

3.1.3. Influence of the Pre-Turbo Catalyst (PTC)

Figure 10 shows a comparison of the cumulative HC and CO emissions in the raw exhaust gas to that after the hybrid catalyst CC1 and/or after PTC 1 and hybrid catalyst CC1.

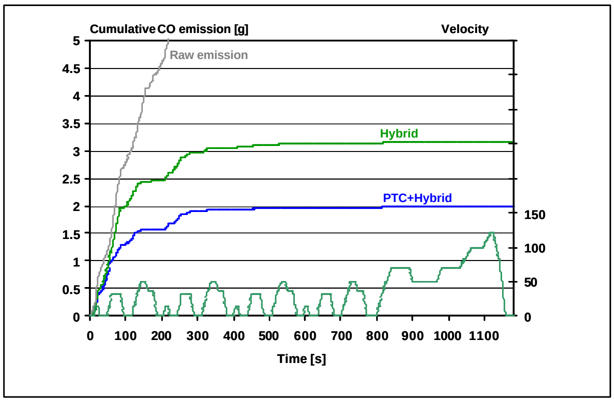

Figure 10a:
Cumulative CO emissions in the raw exhaust and emissions after PTC + hybrid catalyst and after hybrid catalyst alone

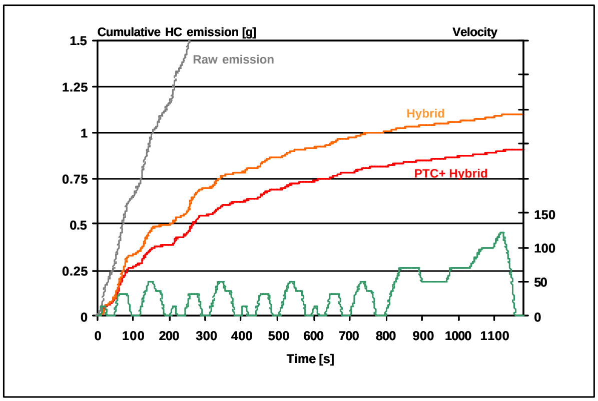

Figure 10b:
Cumulative HC emissions in the raw exhaust and emissions after PTC + hybrid catalyst and after hybrid catalyst alone

As shown in Figure 10, the pre-turbo catalyst PTC1 improves emission behavior – measured after hybrid catalyst CC1 – by approx. 35% for CO and 20% for HC.

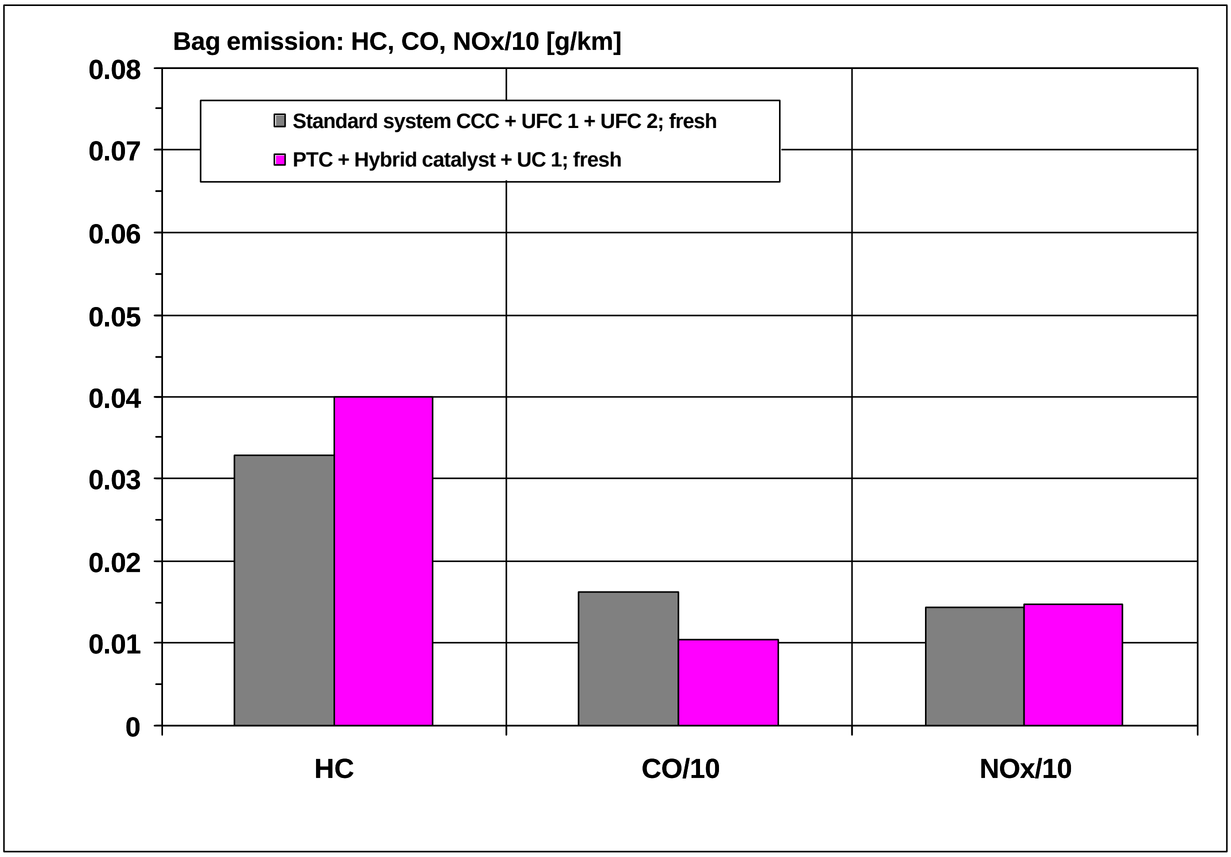

Figure 11: HC, CO, NOx emissions in the NEDC of the PTC catalyst system compared to the serial system

The emission results obtained with this System 4 – PTC1 with CC1 and UC1 – (bag results) are represented in Figure 11 in comparison to the serial system. Whereas a slight reduction of the efficiency in the HC emissions was identified (see also Figure 9), a substantial reduction in the CO emissions of approx. 45% was obtained with System 4. The NOx tailpipe emissions were comparable for both systems.
Thus with the additional PTC1 the decrease in turnover performance discussed in Section 3.1.2 was compensated in the CO emissions after the use of the UC1. The work performed up to this point demonstrated that the potential for downsizing of the catalyst system is given through the use of optimized coatings and advanced substrate structures. Through the above measures, a reduction in the total precious metal content in the range of 25% was achieved, with comparable and in some cases improved emission behavior.

3.1.4. Influence of the PM filter catalyst

To investigate the extent to which the already low particulate emissions could be reduced even further, the coated PM filter catalyst PM1 was used instead of the downstream catalyst UC1. Thus the coated PM filter catalyst had to take over the CO and HC oxidation of the downstream catalysts as well.

Figure 12 shows the gaseous HC and CO emissions and the particulate emissions compared to the serial system.

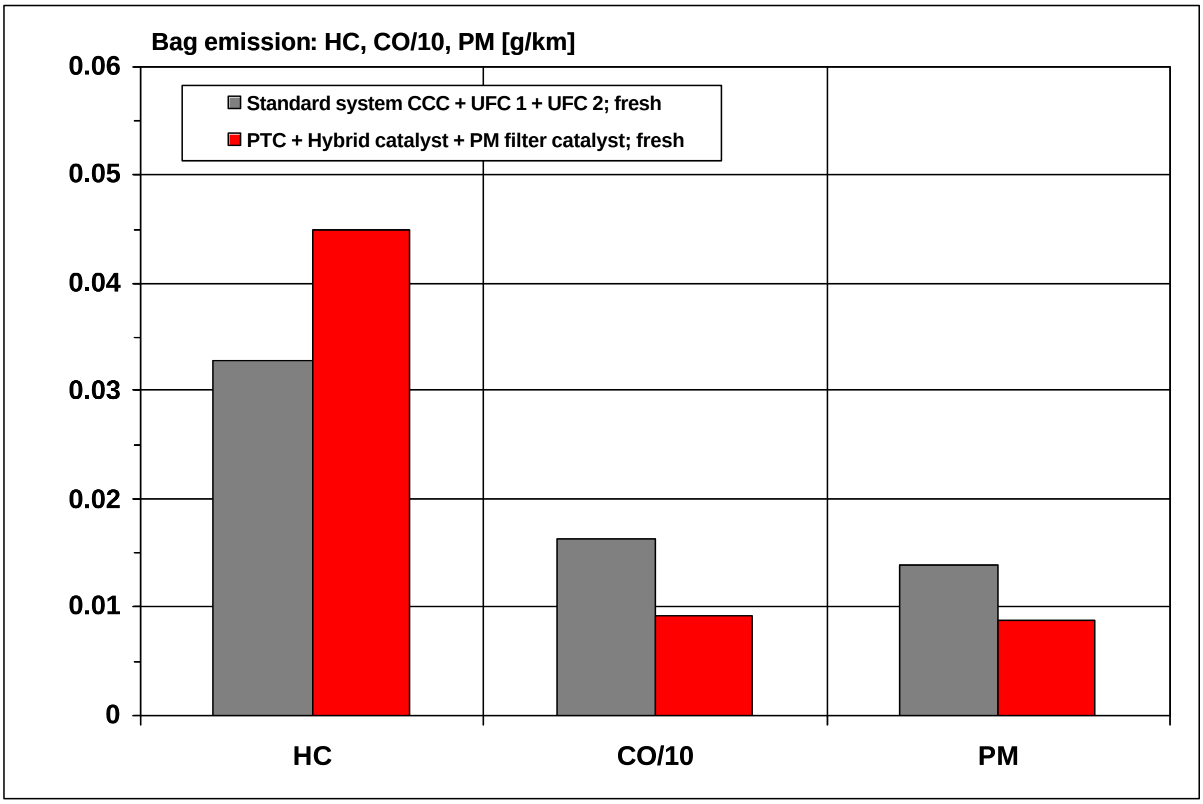

Figure 12: HC, CO, and particulate emissions of the serial system compared to the optimized system with PM filter catalyst

Compared to the results shown in Figure 12 that were obtained with System 4, the HC emissions of the system with coated PM filter catalyst (System 5) increased slightly. However, the CO emissions were comparable and much lower than the serial system. The filtration efficiency of the PM filter catalyst was retained also after coating; thus the coated PM filter catalyst reduced particulate emissions by 38%, whereby an overall test result of 0.0088 g/km PM was achieved.

3.2. Testing of the aged systems

The results discussed up to this point were based on new systems. However, because in realistic applications in particular the influence of aging on the conversion behavior is of interest, the systems tested in chapter 3.2 were aged according to the process described in chapter 2.3.
Figure 13 shows a comparison of the HC, CO, and PM emissions of the following systems:

System 1:	Serial system - new
System 2:	Hybrid catalyst CC1 with serial downstream catalyst UFC1 + UFC2 – new
System 3:	Hybrid catalyst CC1 with downstream catalyst UC1 – new
System 3:	Hybrid catalyst CC1 with downstream catalyst UC1 – aged
System 4:	Pre-turbo catalyst PTC1 with hybrid catalyst CC1 and downstream catalyst UC1 – aged

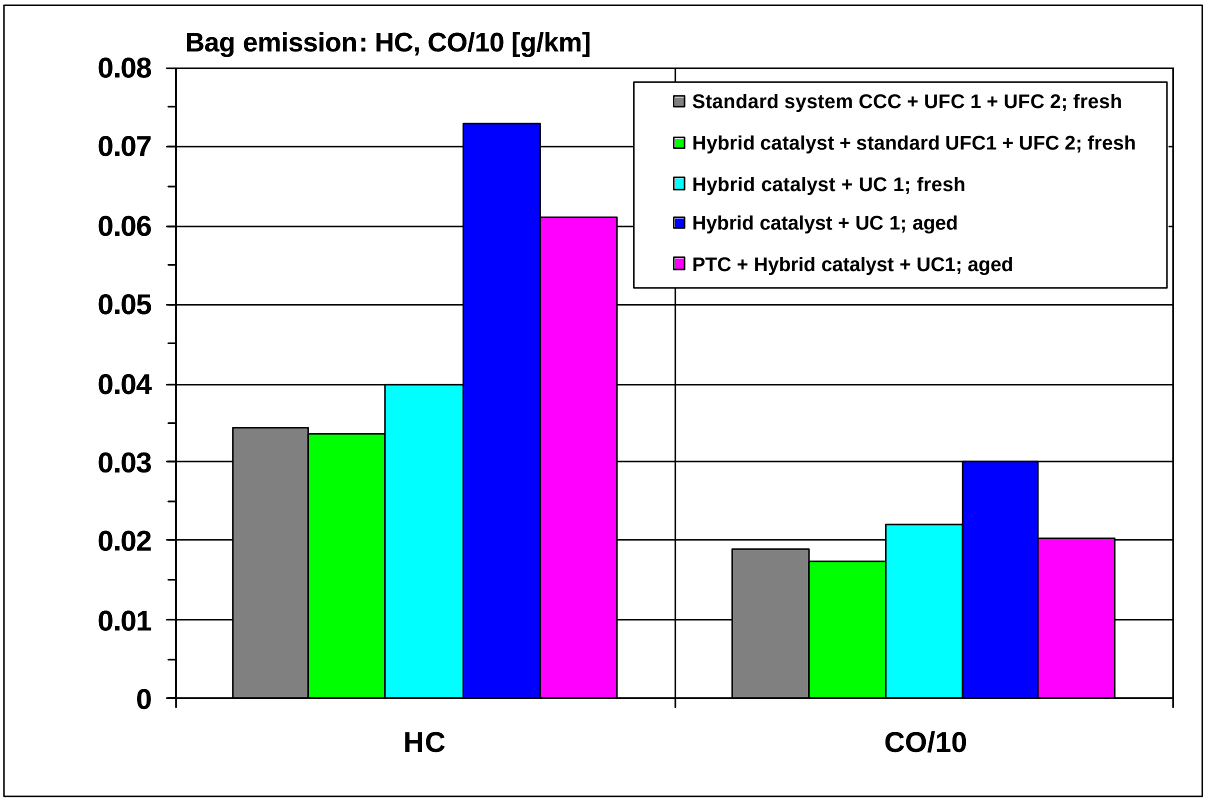

Figure 13a: Comparison of the HC and CO emissions (bag result) of the different systems – new and after aging

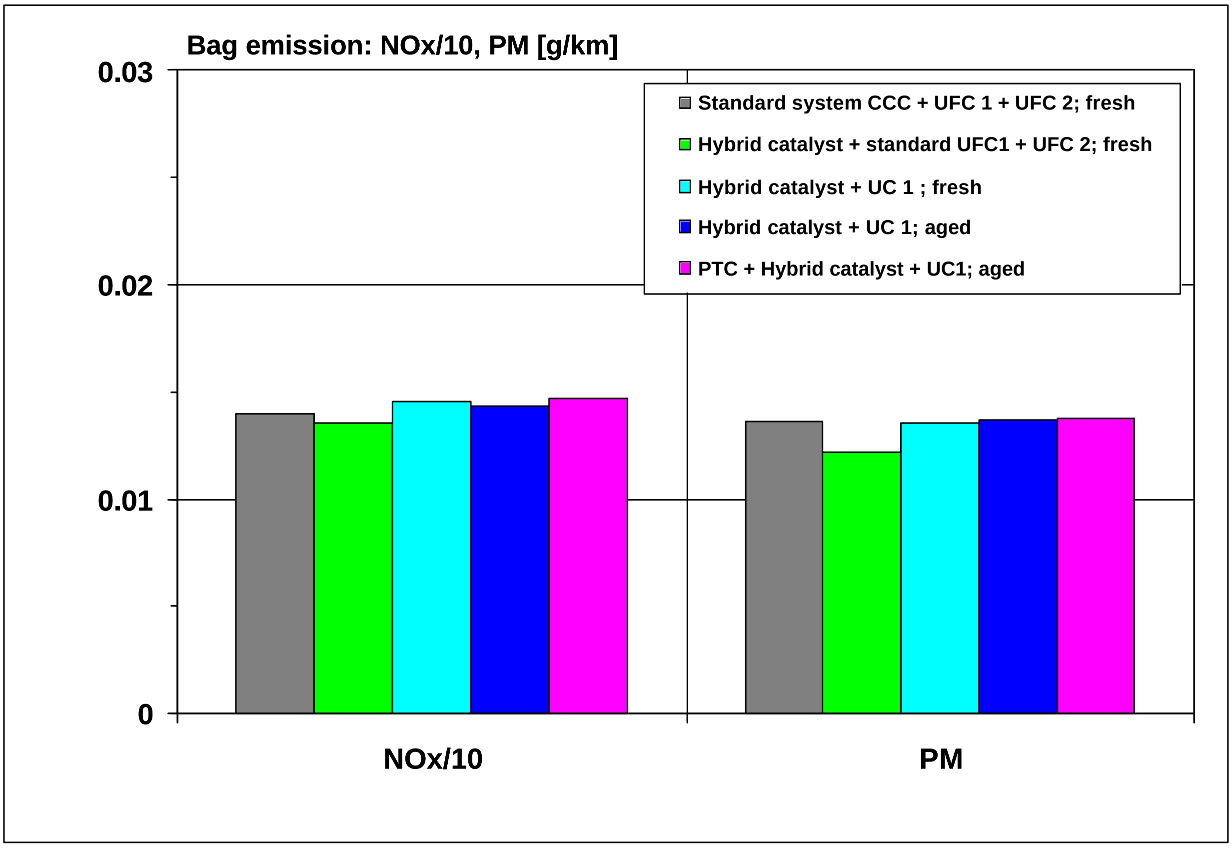

Figure 13b: Comparison of the NOx and PM emissions (bag result) of the different tested systems – new and after aging

As expected, catalyst efficiency decreases after aging. Accordingly, the HC emissions of System 3 – hybrid catalyst CC1 with downstream catalyst UC1 – increased to 0.072 g/km. However, this is still well below the EU IV threshold value for HC+NOx (0.30 g/km). The use of the PTC1 showed a positive effect after aging also, for both the HC and the CO emissions. Compared to the CC1/UC1 system, for example, the aged system consisting of PTC1, CC1, and UC1 achieved the same CO emissions.

4. Summary, Outlook

The described results demonstrate that through the use of optimized coatings and advanced substrate structures, additional potential exists with respect to cost savings and lower emissions.

Due to the improved thermal management with the hybrid structure used, as well as an appropriately adjusted catalyst formulation, the results that were obtained were comparable to those of the serial system despite a reduction in the precious metal content (by about 30%).

Even after aging, the use of the pre-turbo catalyst showed a positive tendency toward reduction, primarily of CO emissions. No negative influence on the emission of CO2 could be determined during the NEDC test cycle with the vehicle tested here.

New catalyst formulations and adjusted processes allow the catalytic activation of PM filter catalysts without decreasing filtration efficiency.

The coated PM filter catalyst used in this test series reduced the already very low particulate emissions by a further 38%.

The results continue to demonstrate that the key factors in a system development are the adjustment of the thermal behavior influenced essentially by the substrate and the temperature-dependent, kinetic characteristics of the catalyst.

Further potential is expected from the use of substrates with improved mass transport (LS design) and more advanced coatings. Additional studies on this subject are planned.

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