Future Exhaust-Gas Aftertreatment Technologies for Spark-Ignition Engines; The Next Generation of Super Ultra Low Emission Vehicles

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1) Introduction

In the course of recent years, limiting values for emissions from motor vehicles have been continuously tightened up even further all over the world. The most stringent values at the moment are achieved with the SULEV (Super Ultra Low Emission Vehicle) legislation in California.

Table 1: Limiting values for exhaust gases

	HC	CO	NOx
ULEV*	0.04 g/m	1.7 g/m	0.2 g/m
SULEV (2003)*	0.01 g/m	1.0 g/m	0.02 g/m
EG III (2000)**	0.2 g/km	2.3 g/km	0.15 g/km
EG IV (2005)**	0.1 g/km	1.0 g/km	0.08 g/km

* FTP test; ** EC III test cycle

Originally, the Californian environment authority CARB planned the mandatory use of electric vehicles for the model year of 2003 with the aim of reducing the environmental impact, above all, in the Los Angeles basin. International studies and fleet tests with electric vehicles showed that, although electric vehicles may be regarded as "zero emission vehicles" locally, emissions arise globally during their manufacture and during the electricity generation [1]. Furthermore, published exhaust-gas results of modern vehicles with optimised exhaust-gas aftertreatment systems exhibit ever more frequently negative exhaust-gas results in the condition warm from operation, even in the case of comparatively low ambient immissions. This means that, with an active catalyst system, such vehicles are in a position, primarily in urban areas with relatively high immissions, to contribute a positive (i.e. cleaning) effect to the problems associated with emissions. In parallel, the catalytic coating of vehicle radiators ensures the reduction in the ozone values close to the ground [2]. Subject to the proviso that the entire vehicle fleet is, in the course of the next 10 - 20 years, equipped with engines and catalyst systems which can satisfy the SULEV or similar limiting values, the emission problem may be considered to be solved with regard to hydrocarbons, carbon monoxide and nitrogen oxides.

Not only the consumption of resources for the manufacture of fuel but also the CO_2 emissions as greenhouse gas and thus as a possible cause of the "greenhouse effect" are still the subject of discussion. If the natural CO_2 emissions and those produced by human beings are considered in a worldwide context, it is established that the proportion accounted for by private traffic amounts to just 0.2 % [3]. Whether the demonstrably rising CO_2 values of the Earth's atmosphere produce the global warming or the warming is caused, for example, by the extension of the cycle duration of the sunspots (thus resulting in higher CO_2 emissions of the seas) has still not been clarified unambiguously at the moment.

In future, it will be possible to achieve the reduction in the fleet consumption, as demanded by the legislators, without any loss of comfort by means of new engine and transmission technologies in conjunction with lightweight construction. In this connection, even greater emphasis is being placed on the discussion about the optimum drive. However, all the environmentally relevant aspects must also be taken into consideration when new drive technologies are compared.

2) Drive Technologies

Not only the well-known spark-ignition and diesel engines but also lean-running direct-injection spark-ignition engines, hybrid vehicles, fuel cells and (a continuing subject) the electric motor are under discussion as drive concepts.

In the comparison of emissions, it must be possible to gauge all the drives according to the ZLEV vehicle already presented in the USA today [4]. Compared with the spark-ignition engine, diesel engines exhibit a clear advantage with regard to consumption. The particle emissions and a further reduction in the nitrogen oxides must be designated as problematical. Lean-running direct-injection spark-ignition engines are based on the deliberation (similar to the diesel engine) of minimising the throttling losses, above all in the low-load range. However, continuous lean operation across all the load points (including the cold start) still cannot be constituted at the current point in time from the viewpoint of engine technology and the catalyst. The sulphur content in the fuel, the regeneration and the maximum temperature loadability of the NOx adsorbers currently used may even lead to consumption disadvantages in certain operating points. Similar to the diesel engine, the reduction in the emissions of nitrogen oxides constitutes the main problem since there are still no sufficiently effective "lean NOx catalysts" available at the moment.

Hybrid vehicles (e.g. the Toyota Prius [5]) represent a compromise between the electric vehicle and the diesel or spark-ignition engine. In addition to the great complexity of this system, the higher weight is one of the principal disadvantages. Consumption advantages can only be achieved in fields where vehicles predominantly run in traffic jams or in stop-and-go operation. The additional weight has a detrimental effect in the case of continuous operation, primarily at higher speeds.

The weak point in the case of the electric vehicles continues to be the on-board energy supply. Even ultramodern battery technologies do not have an adequate storage capacity in order to permit driving operation comparable with that of the internal-combustion engine. The mass of the batteries has an additional negative effect with regard to the energy balance.

The fuel cell would constitute a solution which could be used in order to generate power on board. In this respect, a distinction must be made between fuel cells with a direct hydrogen supply (hydrogen tanks in the vehicle) and fuel cells with upstream reformers. A reformer produces hydrogen from methanol and/or petrol. The present petrol infrastructure could be used with the aid of this system. However, today's reformers and fuel-cell stacks have a respectable weight and need a very large construction space. In the energy balance, it is only possible, with the current state of the art, to push forward into the field of the diesel engines. Since the final customer is not willing to accept any loss of comfort (e.g. long waiting times prior to starting-up), it appears to be doubtful whether the system will be introduced in large quantities within the next 10 years.

The following table shows a summary of the assessment of the drives with regard to emissions, consumption (CO_2) and weight/costs.

Table 2: Assessment of different drive concepts with regard to emissions and weight/costs

	Spark-ignition engine	Diesel	DI spark-ignition, lean	Hybride	Fuel cell
HC	++	+	+	+	++
NOx	++	- (-)	- (-)	+	++
Particles	++	--	-	++	++
CO2	+	++	+ (+)	-	o / -
Weight/costs	+	+	+	--	-
Total	++	+	+	(+)	+

3) Assessment of Catalysts and Emissions

The portrayed assessment of the individual drive concepts shows clearly that the spark-ignition engine will remain the preferred drive concept with the lowest risks, at least for the next 10 - 15 years. Even today, the spark-ignition engine alone and in conjunction with new transmission variations still exhibits great potential for improvement with regard to power, emissions and consumption. Therefore, the requirements on new drive concepts will become ever more stringent.

Now, the objective must be to provide the vehicles with emission-technology equipment in such a way that it no longer makes any sense, even politically, to portray the motor vehicle as the number-one environmental offender and to expose it to more or less arbitrary political decisions, as is shown by the current discussion about the diesel taxation in Germany. The Californian SULEV limiting value is defined as the aim first of all.

The course in principle of the cumulative HC emissions of a production engine and the demanded SULEV values via the American FTP test are shown on Fig. 1.

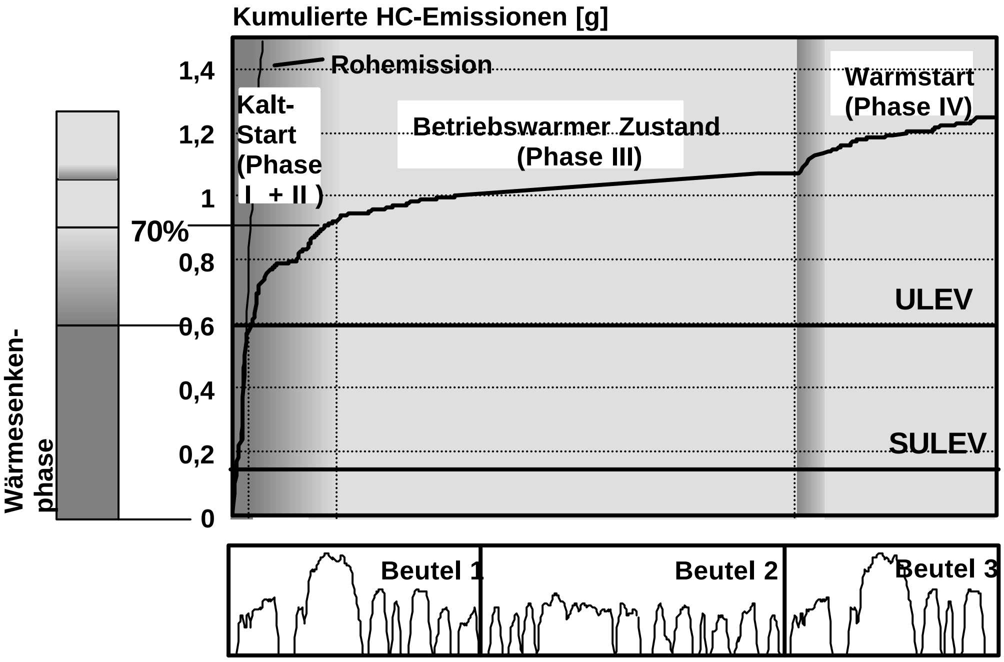

Fig. 1: Cumulative HC emissions in the FTP test

In addition to an almost 100 % conversion rate in the condition warm from operation, it is primarily an extremely short "starting time" of the catalyst after the engine start which is necessary. The maximum starting duration is directly dependent on the engine-out emissions in the cold start and, according to the engine, lasts between 3 and 15 seconds [6]. In order to satisfy this requirement, the gas temperature upstream of the catalyst must exceed the light-off temperature of 200 - 300°C a few seconds quicker. A catalyst position as close as possible to the engine and directly downstream of the manifold is suitable in order to avoid any unnecessary heat losses in the upstream tubes to the catalyst.

The cold-start behaviour of catalysts is determined not only by the gas temperature and the catalytic coating but also by the physical and thermodynamic data of the substrate. During the cold-start phase, the catalyst initially works as a pure heat exchanger with catalytic activity as from the time when the light-off temperature has been reached.

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For this reason, the objective is to provide the largest possible catalytic surface (GO) with the lowest possible heat capacity (cp). The so-called GO/cp cold-start factor is used in order to assess different systems [7].

Fig. 2 shows the cold-start factor of various metallic catalyst substrates.

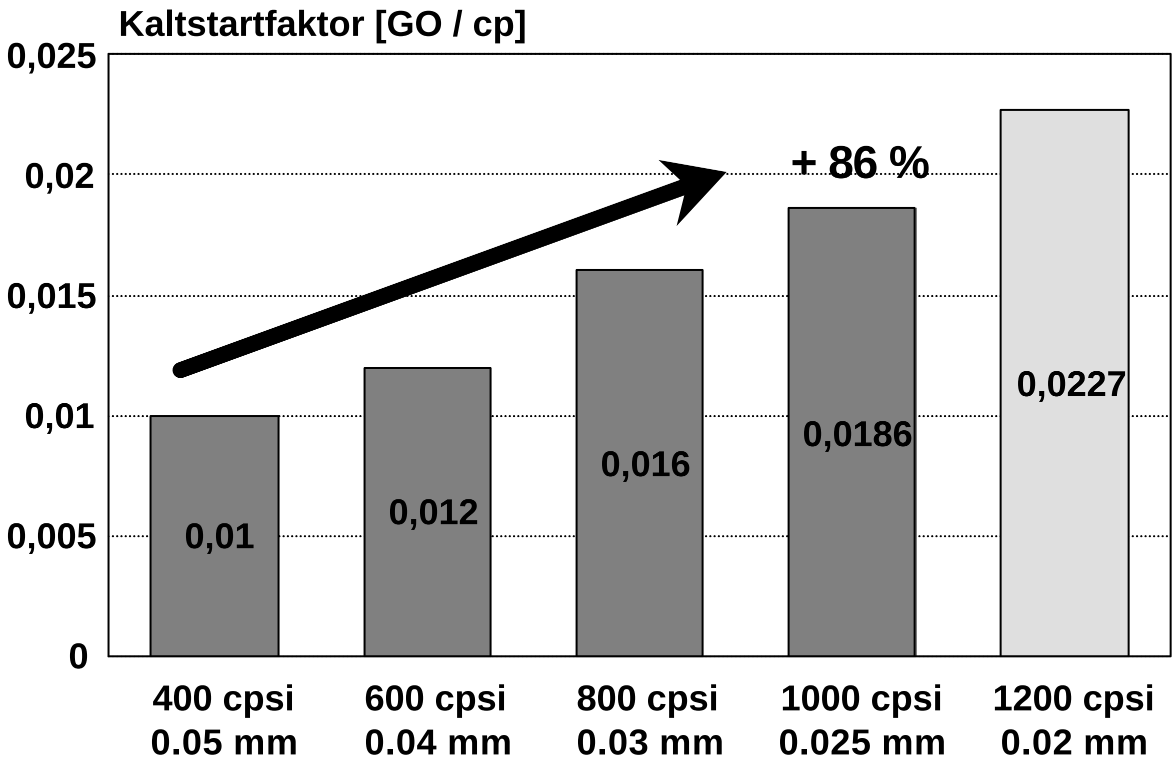

Fig. 2: Cold-start factor of various metallic catalyst substrates (400 - 1200)

It is possible to recognise that the cold-start factor, depending on the cell density, increases in connection with a reduction in the foil thickness. However, if the cell density is increased without reducing the foil thickness, it is possible to identify a deterioration.

Another important factor is the use of a heat cascade. The reduction in the entry cross-section of the first substrate has a positive influence on the cold-start behaviour and also on the flow distribution [8]. Fig. 3 shows the BMW six-cylinder engine as an example of a graduated catalyst installation close to the engine [9].

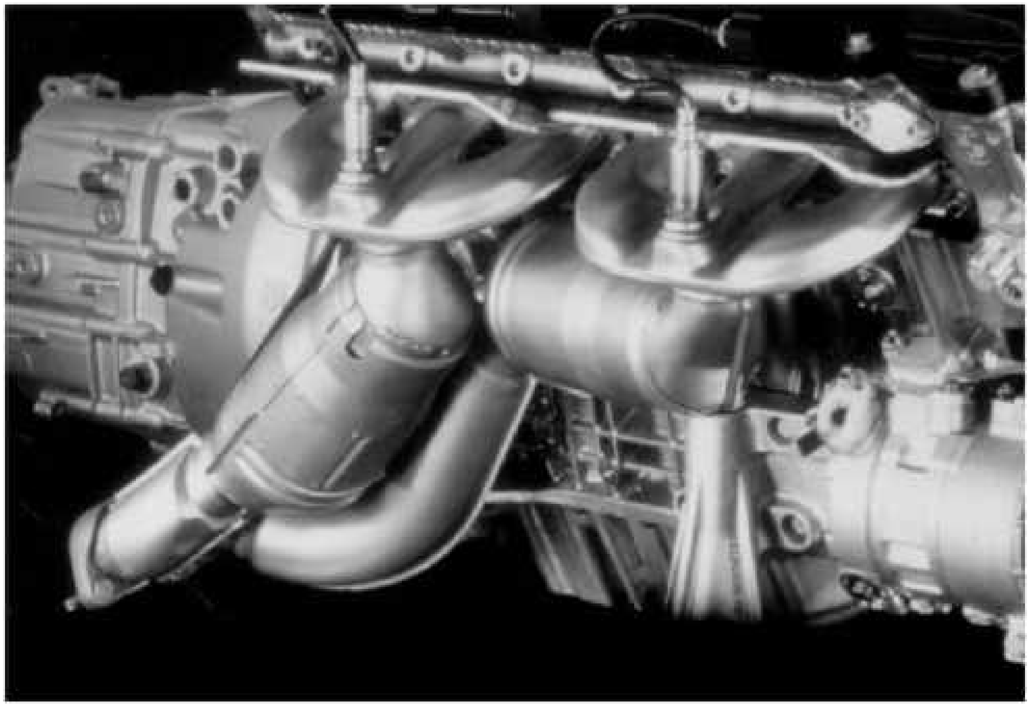

Fig. 3: Graduated catalyst installation close to the BMW six-cylinder engine

Due to the continuous reduction in the heat capacity of the catalyst substrates, ever greater significance is being attached to the proportion of the wash-coat in the total heat capacity. Wash-coat quantities of 200 - 250 g/l are absolutely usual for modern three-way catalysts. In the case of a 400 cpsi catalyst with a foil thickness of 0.05 mm, a reduction in this mass to 150 g or 100 g entails a decrease in the wash-coat proportion from 63 % to 25 %. With a 1000 cpsi substrate and a foil thickness of 0.025 mm, the improvement amounts to 53 %. The following figure illustrates the dependence of the wash-coat mass on the total heat capacity.

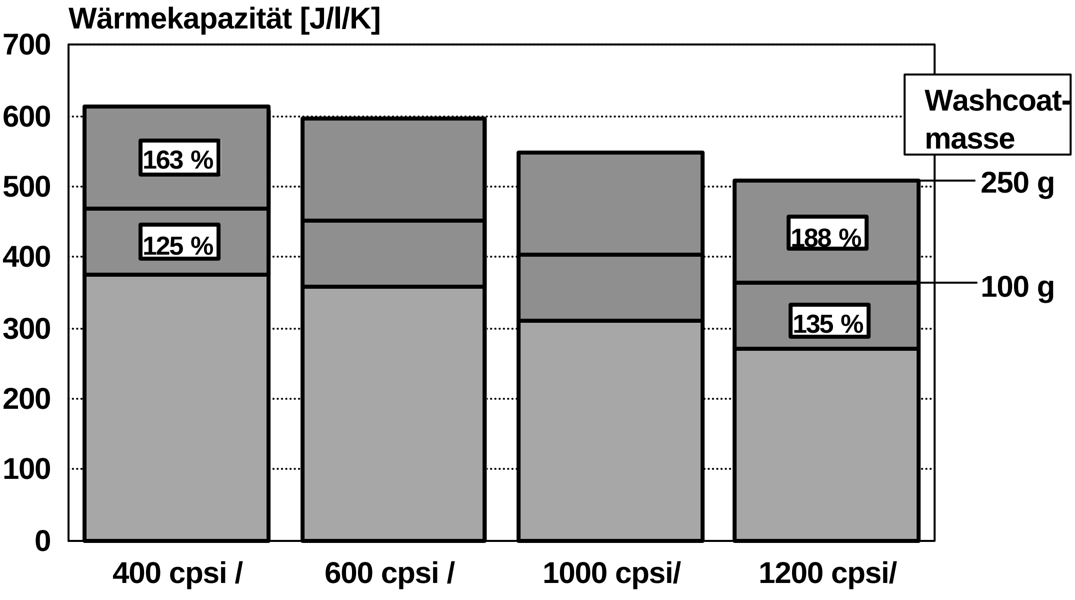

Fig. 4: Influence of the wash-coat mass on the total heat capacity of the catalyst

In order to make optimum use of the substrate and of the noble metals contained in the coating, uniform distribution of the wash coat around the circumference of the cell and along the axis of the catalyst is imperative. Fig. 5 shows the example of one thick and one thin wash coat within the cell cross-section.

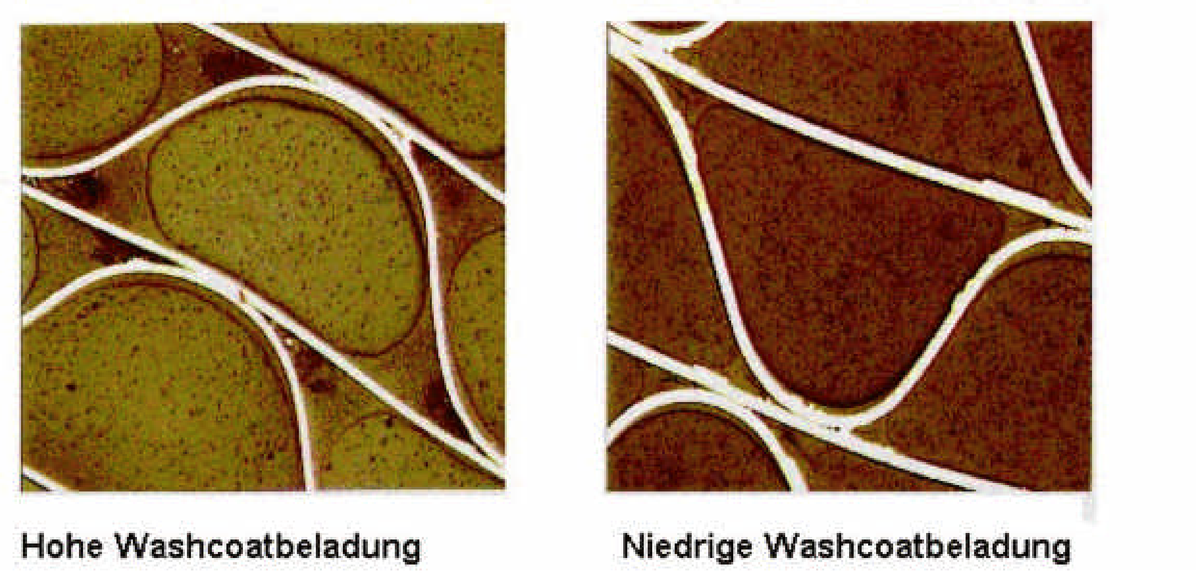

Fig. 5: Wash-coat distribution depending on the wash-coat mass in the cell cross-section

In the condition warm from operation, the catalytic effectiveness is only limited by the material transport, subject to the prerequisite of optimum lambda control. The material transport within the laminar flow in the cell channels is primarily influenced by the hydraulic diameter (dh) of the channel. GO/dh is regarded as the effectiveness factor [10]. Fig. 6 shows the effectiveness factor GO/dh of different cell densities.

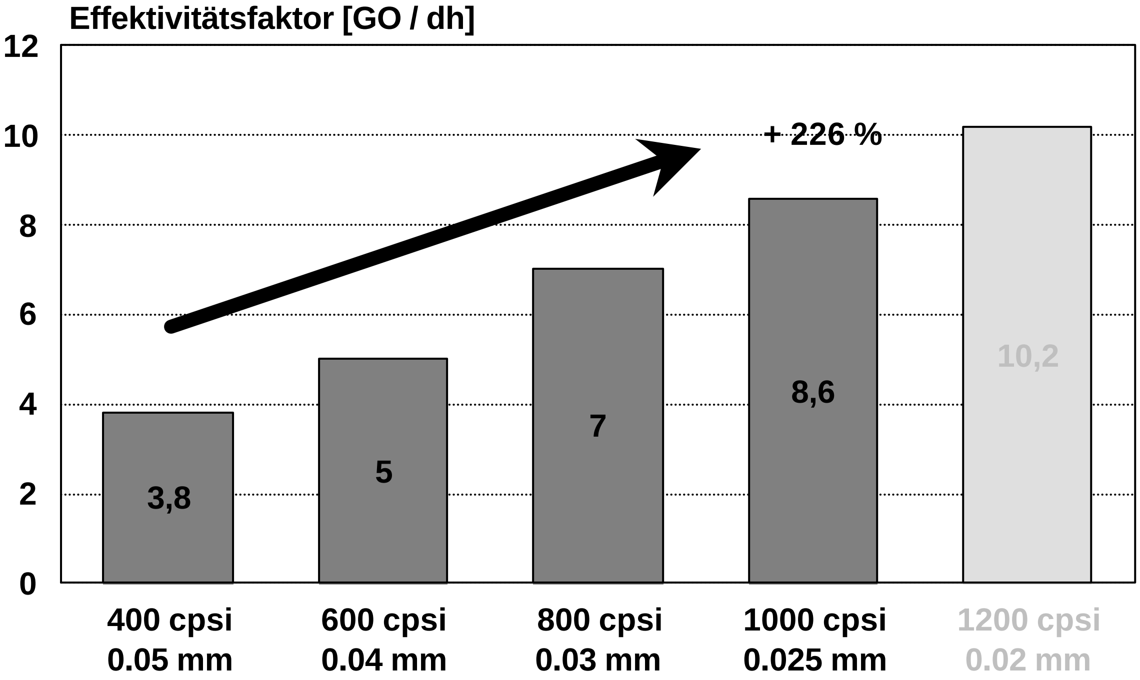

Fig. 6: Effectiveness factor GO/dh of different cell densities

Higher cell densities entail improved effectiveness for all pollutants [6, 11, 12]. Even in the case of greater catalytic effectiveness, this will make it possible to reduce the volume of the catalyst. Therefore, no additional underfloor catalysts will be necessary. In this connection, NOx reacts most critically to a reduction in the volume of the catalyst if the engine control is not optimum.

If the cumulative NOx emissions within an exhaust-gas test are analysed, it is established that the total emissions may, in most cases, be attributed solely to just a few "individual events" within the test. These short NOx leaps shown on Fig. 7 are not dependent on the spatial loading of the catalyst but are instead triggered by non-optimised lambda control in the transient range. E-Gas and quicker engine-control devices with which it will also be possible to control the individual cylinders in a targeted way will guarantee a substantial improvement.

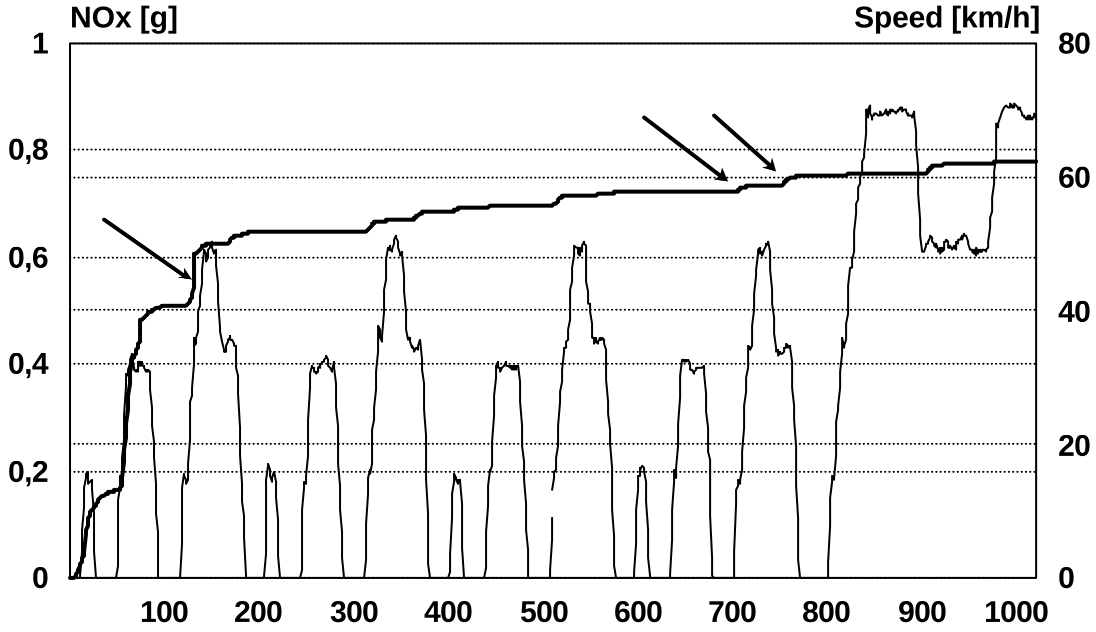

Fig. 7: Cumulative NOx emissions of a production vehicle in the exhaust-gas test

4) SULEV catalyst concept

An engine with low engine-out emissions and optimised lambda control is imperative in order to constitute a SULEV vehicle. For this reason, all the further deliberations are made on the assumption of a ULEV vehicle which is available on the market and has SULEV potential. The engine-out emissions of this vehicle in the FTP test are 1.6 g/m.

The exhaust-gas temperature and the cumulative HC emissions during the first 100 seconds in the FTP test are shown on Fig. 8.

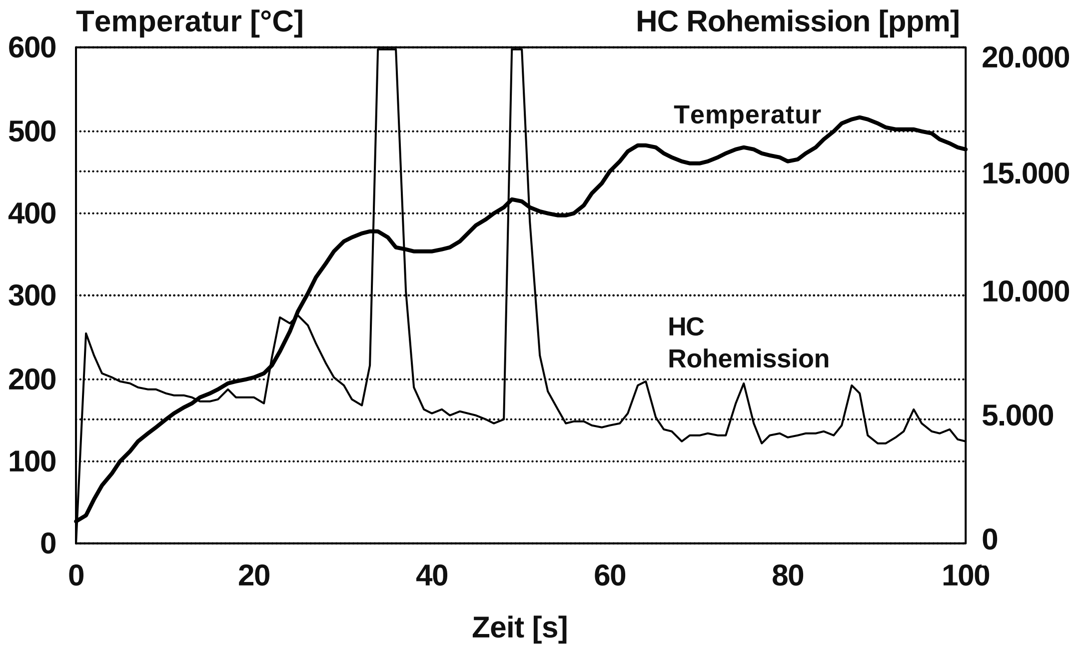

Fig. 8: Gas temperature upstream of the catalyst and HC engine-out emissions during the first 100 seconds of the FTP test

The catalyst system is designed according to the assessment factors described in Chapter 3. The total volume should correspond to approx. 0.6 - 0.8 times the cubic capacity. Reference was made to the KatProg catalyst simulation program for assistance purposes.

The basic idea is to develop a catalyst system which can be modified in a modular form to new legislation right up to SULEV. On the basis of the OBD requirements, the system should be a "two-brick" solution with the possibility of monitoring the first substrate separately. With regard to good flow distribution and to the improved cold-start effectiveness, the catalyst system is designed as a cascade. If the incoming flow is very poor due to the construction space, the first substrate is replaced by a conical catalyst.

The cell density and the foil thickness can be varied according to the effectiveness requirements in each case. For applications with higher engine-out emissions or low gas temperatures, it is possible, without altering the construction space, to replace the first substrate with an electrically heatable catalyst consisting of a heating disc and a back-up catalyst. Fig. 9 shows the principle of the system in different design variations.

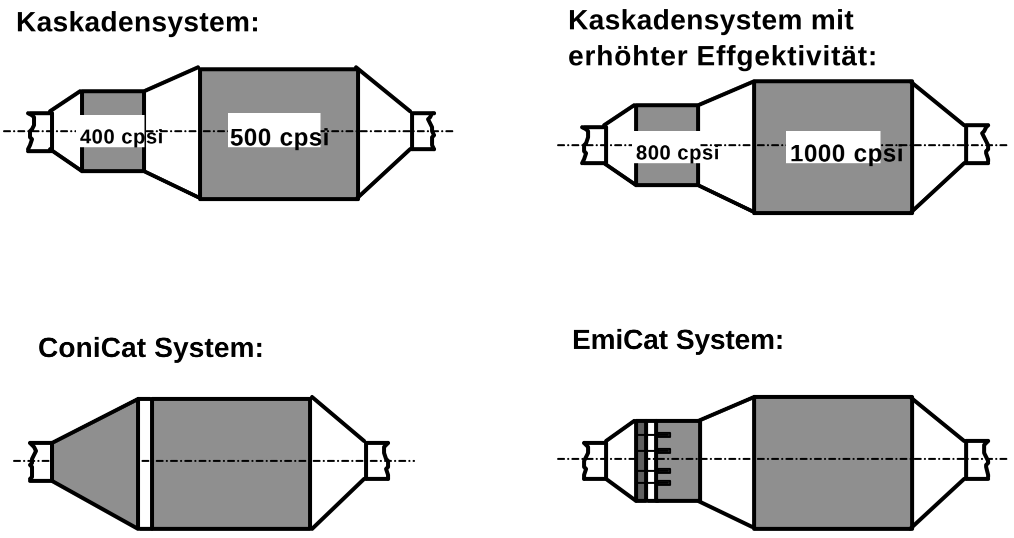

Fig. 9: Design variations of a catalyst system with a modular structure

5) Emission and calculation results

The catalyst system with a modular structure provides the possibility of adapting an application to different limiting values for exhaust gases without altering the canning. Of course, this is only possible with parallel further development of the engine and of the engine management. Catalysts with high cell densities and ultrathin foils can only be used in conjunction with an optimally acting engine-management system without HC peaks in the transient driving operation. In the case of these very active catalysts with a low heat capacity, HC peaks lead, in comparison with substrates with thicker foils, to extreme increases in temperature and thus to the ageing of the catalyst. Variations of the cascade system depending on the legislation are shown in Table 4. The total catalyst volume is 1.4 l and thus accounts for 70 % of the cubic-capacity volume of the test vehicle.

Table 4: Variations of a catalyst system with a modular structure

	First substrate Ø 80 x 74.5 mm			*Second substrate Ø 110 x 110 mm*	
	Cell density / foil thickness		*Cold-start factor*	*Cell density / foil thickness*	*Effectiveness factor*
LEV / EG III	*300cpsi / 0,05*		*0.01*	*500cpsi / 0,05*	*4.6*
ULEV / EG IV	*600cpsi / 0,04*		*0.12*	*800cpsi/0,03*	*7.0*
SULEV	*800cpsi / 0,03*		*0.16*	*1000cpsi / 0,025*	*8.6*
EZEV	*Heating disc: 600cpsi / 0.04*	*Back-up catalyst 800cpsi / 0,03*	*0.12 / 0.16*	*1200 cpsi / 0,025*	*10.1*

The following illustration shows the HC conversion rates and the calculated cumulative HC emissions during the first 100 seconds of the FTP test. In order to validate the program and the catalyst effectiveness, comparative calculations were made in advance with measured results for the exhaust gas of the original catalyst system with deviations of < 5 %.

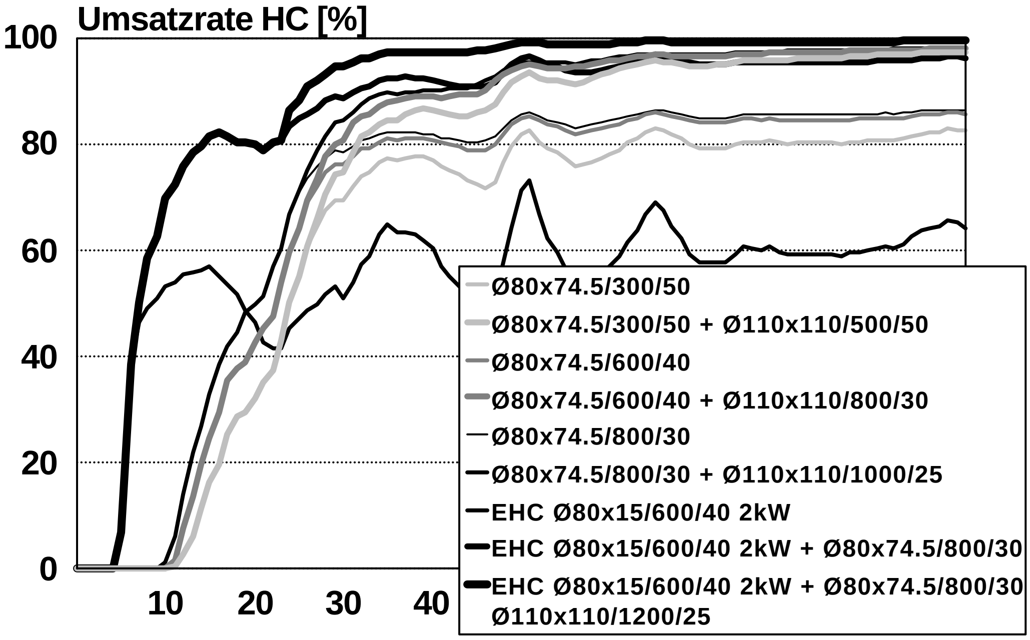

Fig. 10: HC conversion rates of different catalyst installations during the first 100 seconds of the FTP test

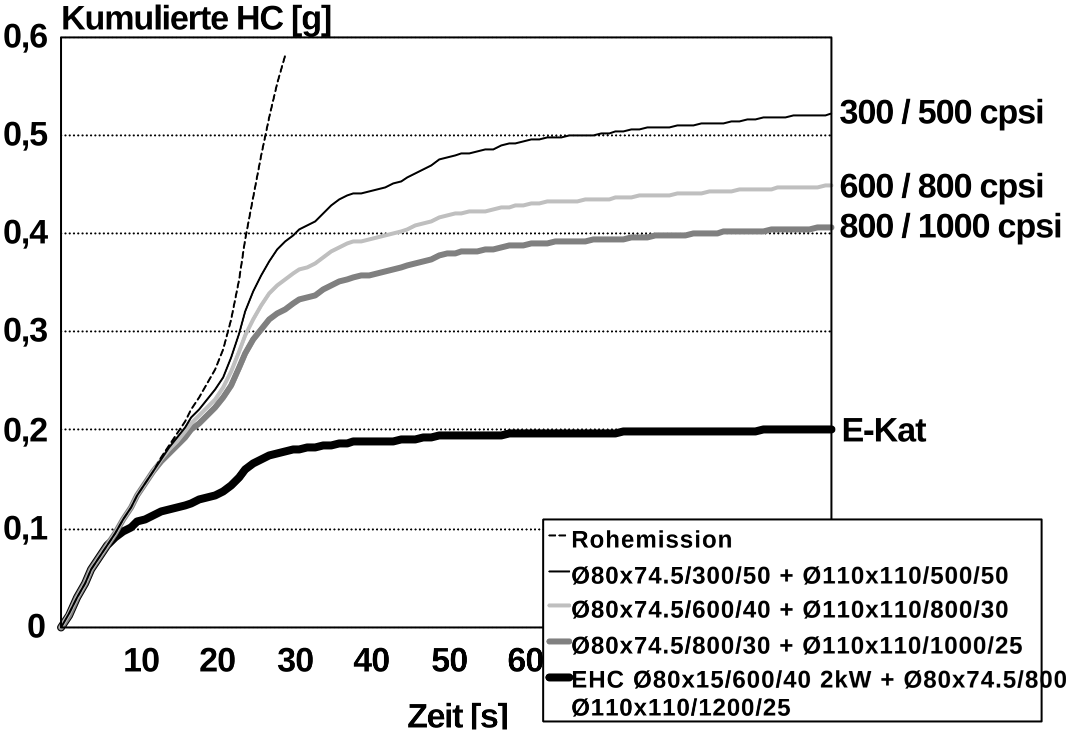

Fig. 11: Cumulative HC emissions of different catalyst variations during the first 100 seconds of the FTP test

The calculation results indicate the advantage of the higher cell densities in conjunction with the thinner foil thicknesses. In the identical canning, the cold-start emissions can be reduced by 23 % by changing from 300 cpsi / 0.05 mm to 800 cpsi / 0.03 mm. The additional use of an electrically heatable catalyst entails an improvement potential of 60 % in the cold start. Experience has shown that, in the condition warm from operation, the increase in the cell density also leads to a substantial improvement in the catalyst effectiveness due to better material transport in the channels.

The possibility of achieving the catalyst effectiveness in an identical canning is suitable as an optimum-cost solution for vehicles which must comply with different limiting values for exhaust gases over the production period.

6) Summary

Those limiting values for emissions achieved by vehicles published in America as prototypes illustrate that, with regard to the limited emissions, it will be possible in future to build vehicles whose exhaust gas in urban areas will be cleaner than the intake air. Thus, the car will clean the environment.

A comparison of different drive concepts under discussion supports, apart from the CO_2 *emissions, the leading role of the spark-ignition engine for the next 10 to 15 years. The weighting of the* CO_2 *in this connection certainly depends on the question of the extent to which the carbon dioxide emitted by the private traffic also shares the blame for the global warming or is only being used as a means of exerting political pressure.*

In order to comply with future limiting values for exhaust gases with the spark-ignition engine, it is imperative to optimise not only the catalyst but also the engine and the engine management. The overall system consisting of the engine, the management, the catalyst substrate and the catalytic coating must be coordinated. Subject to this proviso, it is possible to design catalyst systems with a modular structure in accordance with the future requirements.

Modular catalyst systems permit the cost-favourable adaptation of the catalyst effectiveness by increasing the cell density while simultaneously reducing the foil thickness (heat capacity) or by using conical or electrically heated catalysts. Thus, no additional catalyst volume is necessary while the overall system is improved at the same time.

Work is currently being performed on the validation of the calculated emission results and on an examination of the long-term mechanical and thermal durability.