

Application Guideline to Define a Catalyst Layout for Maximum Catalytic Efficiency

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ABSTRACT

The influence of physical parameters of the catalyst's substrate such as thermal mass, hydraulic diameter and geometric surface area on catalyst's efficiency is well known as published in numerous works. This paper will show interactions of these parameters and will provide a guideline on how to design the optimum system for a specific application, taking into account system's back pressure and system costs. Based on engine test bench results that show the influence of the physical parameters, the results for the optimized design regarding emission tests and maximum conversion rate at higher loads will be demonstrated.

1. INTRODUCTION

The tightening of emission legislation and the compulsion to guarantee of maintaining to these limits even after 80,000 miles, for example, led to an ongoing reduction of environmental pollution caused by motor vehicles in conjunction with On Board Diagnosis OBD.

The demands on reduced engine-out emissions and the quality of exhaust gas aftertreatment systems grew accordingly. Figure 1 shows the required Hydrocarbon (HC) conversion rates depending on the level of untreated engine-out emissions and the respective HC-limits of Californian exhaust gas legislation.

The conversion rates are based on HC engine-out emission levels of 2.0 g/mile resp. 1.5 g/mile in the Federal Test Procedure (FTP). It is striking that even in

the case of reduced engine-out emissions, catalyst efficiencies increases continuously starting from "Transient Low Emission Vehicles" (TLEV) through to the "Super Ultra Low Emission Vehicles" (SULEV).

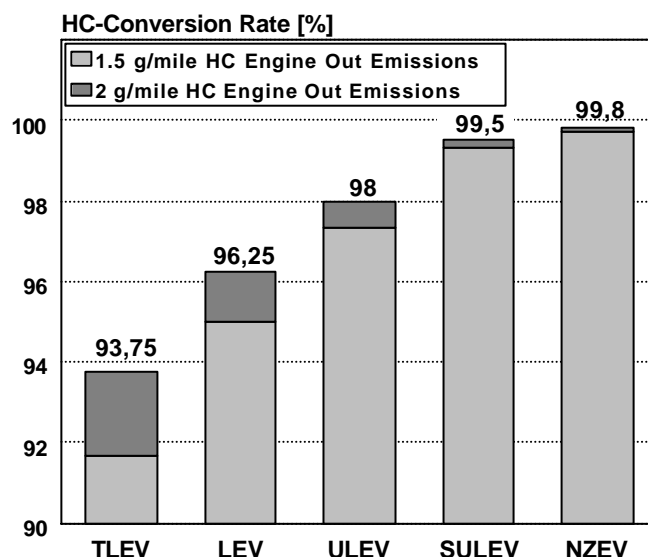


Figure 1: Required HC conversion rates depend on Californian emission legislation and on different levels of HC engine out emissions

It has been shown in various studies [1, 2, 3], that cold start emissions in particular can be significantly reduced by moving the catalyst in a position closer to the engine because of the more rapid heating of the catalyst system. In addition, an increase in cell density led to an increase

in efficiency at operating temperature and a reduction in the catalyst volume [4, 5, 6]. Due to existing conditions such as lambda control or system pressure-loss requirements it is, however, necessary to make compromises which are influencing both catalyst efficiency as well as the costs of the exhaust gas aftertreatment system.

However, the objective for the future has to be the development - without compromise - of the most cost effective exhaust gas aftertreatment system with a conversion rate as close as possible to 100% for all pollutants, so that motor vehicles do not place any additional burden on the environment - even in areas with low background emissions. This new generation of cars will help to clean up even these areas with regard to the limited pollutants.

2. CATALYST EFFICIENCY

Overall catalyst efficiency depends on cold start behaviour and the efficiency at operating temperature. Since in the past cold start emissions represented approximately 80% of the total emissions, development was focussed mainly on improving light-off behaviour. In summary it can be said that primarily due to a reduction in thermal mass [7, 8, 9] and in catalyst diameter [10] assisted by higher exhaust gas temperatures in the front of the catalyst it has been possible to significantly improve cold start efficiency. Today the development of close-coupled catalyst systems is almost the norm and more and more vehicles have appropriate passive Ultra Low Emission Vehicle (ULEV) aftertreatment systems as standard. The parallel development of engine management systems as well as an increase in cell density from 400 cpsi to 800 or up to 1200 cpsi made it possible to further reduce emissions at operating temperature as well.

2.1 EXHAUST GAS TEST ANALYSIS

Analysing the HC emissions of an ULEV vehicle with a close couples catalyst system (Figure 2), it can be noticed that even a larger proportion of total emissions is produced during the cold start phase, as compared to catalyst systems designed for e.g. the TLEV standard. The problem that at the beginning of the test before light-off the catalyst has an effect of a heat sink, has become even greater regarding the ever increasing exhaust gas standards.

On the other hand, a greater significance is placed on the emissions at normal operating temperature due to the weighting of "Bag 2" in the FTP-cycle. Due to the weighting (Equation 1), emissions in FTP bag 2 have

approximately 2,5 times greater an effect on the overall result than the cold start emissions.

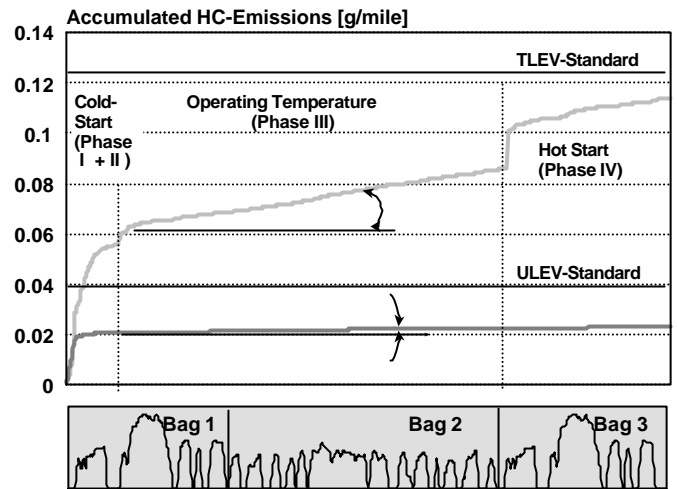


Figure 2: Analysis of accumulated HC emissions in the FTP-cycle for a TLEV- and a ULEV-vehicle

$$E_{\text{gew}} = \frac{(E_{\text{Bag 1}} + E_{\text{Bag 2}})/(D_{\text{Bag 1}} + D_{\text{Bag 2}}) \times 0.43 + (E_{\text{Bag 2}} + E_{\text{Bag 3}})/(D_{\text{Bag 2}} + D_{\text{Bag 3}}) \times 0.57}{1} \quad (1)$$

with $E_{\text{Bag 1}}$ - Emissions in Bag 1, and accordingly in Bag 2 and 3
 $D_{\text{Bag 1}}$ - Distance driven in Bag 1, and accordingly in Bag 2 and 3

Therefore its obvious that almost 100 percent conversion of emissions are necessary at operating temperature since the emission standard has to be adhered to even in aged condition, when significant increases in cold start emissions occur.

For the next generation of exhaust gas emission standards such as SULEV, "Near Zero Emission Vehicle" (NZEV) or the European standard "Enhanced Environmentally Friendly Vehicles" (EEV), however, catalytic efficiency has to be increased again by factor of 4 to 10. Expressed in other words, the decisive number is not the increase in the absolute conversion rate from 98 to 99.6% (ULEV => SULEV), but the reduction of tailpipe emissions from 0.04 to 0.01 g/m. Due to the weighting of the individual exhaust gas bag results in the FTP-cycle, now the catalyst efficiency at operating condition (Bag 2) gains increased importance, too. In the following section primarily the optimum catalyst layout for normal operating temperature will be discussed.

Studying the limiting factors for catalyst efficiency in a heterogenous catalyst (Figure 3), we find there the lambda control to be of great importance as well as the mass-transfer of the pollutants from the channel center to the walls.

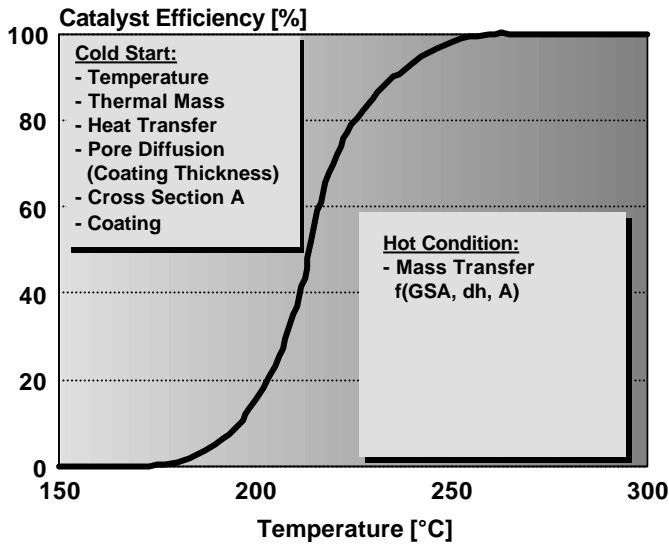


Figure 3: Limiting factors for catalyst efficiency in the heterogenous catalysis [11, 12]

2.2 MASS TRANSFER

Under normal operating conditions the reaction is limited by mass-transfer. Caused by the chemical reaction, the concentration of pollutants is decreased dramatically within the channel in flow direction towards the rear end of the catalyst. This results in decreasing difference of pollutant's concentration between center and wall and therefore in a decrease of the driving force for diffusion. Independently from that, due to the laminar flow, the mass transfer rate numbers are comparably poor. Mass-transfer usually is described using the mass transfer coefficient "Beta".

$$b = \frac{D_{12} \cdot Sh}{d_h} \quad (2)$$

with

- Sh = a Re^m Scⁿ - Sherwood number
- D₁₂ - Binary diffusion coefficient
- Re - Reynolds number
- Sc - Schmidt number
- d_h - Hydraulic diameter of channel

Equation (2) exhibits the dependency of the mass-transfer rate on both the velocity of flow in the channels (described with the Reynolds number), as well as on the channel's characteristic size described with the hydraulic diameter. Discussing now the influence of the catalyst diameter, its obvious to receive a significant gas velocity level, when the catalyst's diameter is reduced. According to equation (2) that results in increased numbers of the mass transfer coefficients, as plotted in figure 4. Therefore smaller catalyst diameters – for a given catalytic reaction, which is limited by mass transfer – reveal higher efficiency. Consequently, in figure 5 the

dependence of the catalyst's HC-conversion efficiency on catalyst's diameter and on the exhaust gas mass flow is displayed.

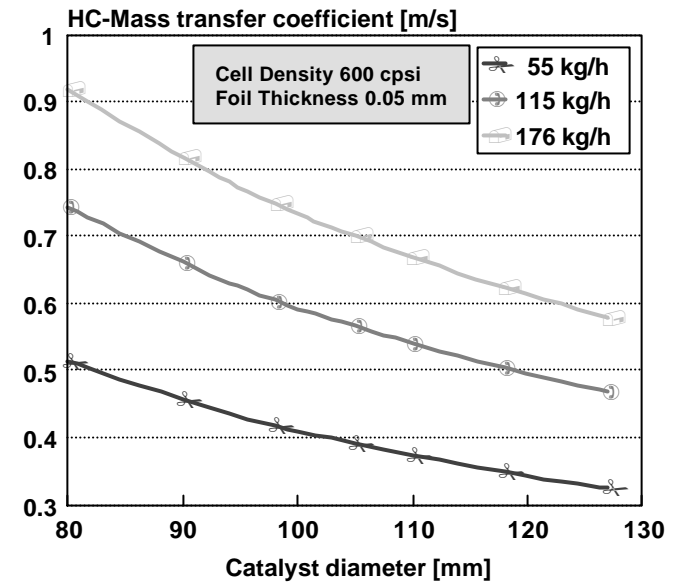


Figure 4: Mass transfer coefficients as a function of catalyst diameter and of exhaust gas mass flow (cell density 600 cps)

In these experiments, the catalyst length was kept constant. Accordingly, catalyst volume increases with increased diameter. The results plotted in figure 5 show, that for each mass flow level there exists an optimum catalyst diameter range, giving an maximum conversion rate result. It can furthermore be observed, that, at the low mass flow level of 50 kg/h the catalyst with 70 mm diameter reveals a higher conversion rate compared to the catalyst with 127 mm diameter, even though the former has a volume deficit of 70%.

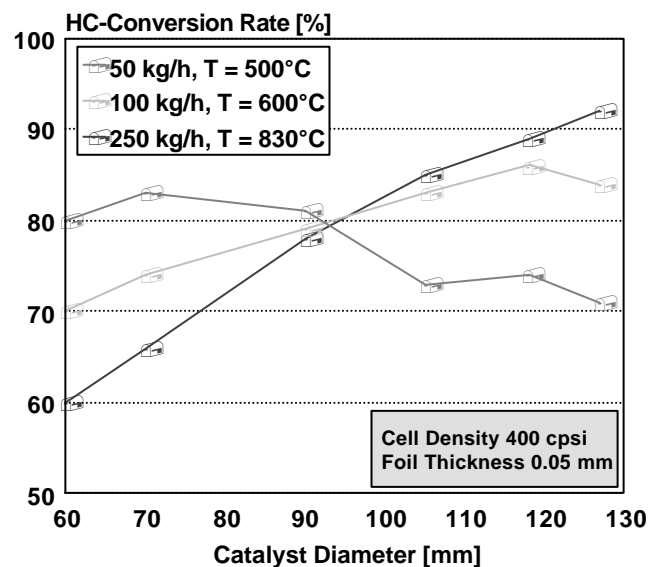


Figure 5: HC conversion rate as a function of catalyst diameter and mass flow (cell density 400 cps)

Since mass-transfer essentially increases with increasing gas velocities, the collapse in efficiency, which can be observed in figure 5 for catalysts with small diameters, would not occur for high mass flows if the length was also adapted (to maintain the catalyst volume). A thin, but long shaped "catalyst-cigar" of this kind, however, would cause a considerable pressure loss, which would not be permissible in a real vehicle for performance and fuel consumption reasons.

However, it can be derived from the results in figure 5, that in principal there is an ideal diameter for each level of exhaust gas mass flow. An evaluation of the mass flow classes occurring in the Federal Test Procedure makes it possible to establish from this the optimum catalytic diameter. The classified exhaust gas mass flow of a medium-class vehicle established via the FTP-cycle is plotted in figure 6. The frequency peak at 14 to 21 kg/h corresponds to the idling mass flow in warm condition. Mass flow when driving is relatively equally distributed within a middle flow range, but frequency falls down for mass flows higher than approximately 100 kg/h. The mass flows above this level occur only in the very short term in phases of stronger acceleration. As a result, an optimum catalyst design derived from this classification would need to have "n" numbers of diameter levels. Because that cannot be produced for reasons of costs and construction space, other parameters will have to be adapted for optimised mass transfer.

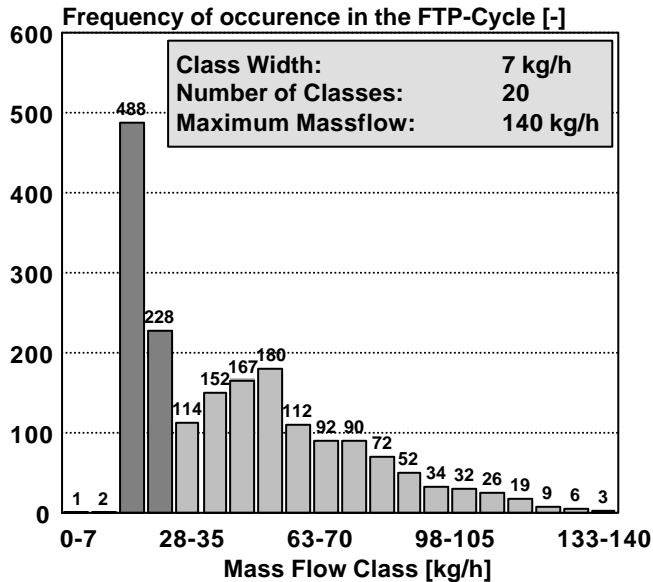


Figure 6: Classified exhaust gas mass flow in the FTP-cycle, tested on a medium-class vehicle

The diffusion path of the pollutants corresponds approximately to half of the hydraulic diameter of a cell within a catalyst monolith. An increase in cell density means both an enlargement of the catalytic surface and a reduction in hydraulic diameter. The positive influence of this measure has been proven already in the past [4, 5, 6].

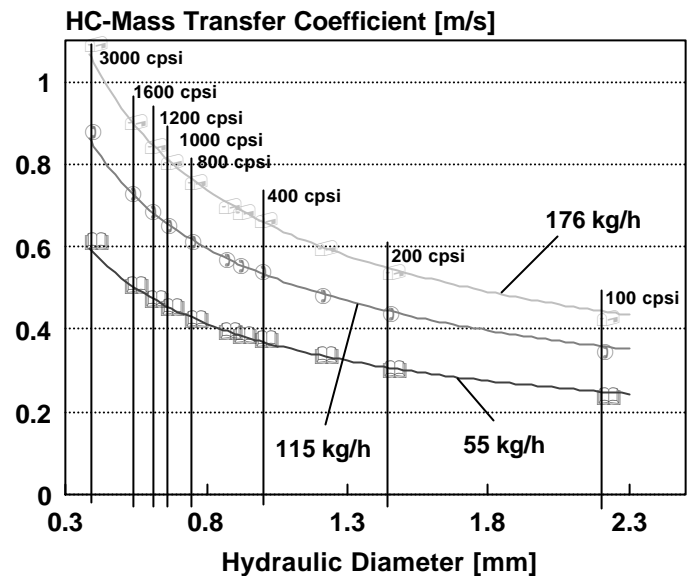


Figure 7: HC-mass transfer coefficients as a function of cell density and of hydraulic diameter at various mass flow rates

In figure 7 HC-mass transfer coefficients are displayed as a function of the cell density resp. the hydraulic diameter at various mass flow rates. The coefficients increase with an increase in cell density or a reduction in channel diameter. This effect is amplified exponentially in the range of cell densities > 1000 cpsi. Comparing the pressure loss of various catalyst designs (catalyst volume const. = 1.5 l) with identical values for mass transfer coefficients reveals a clear pressure loss disadvantage of "cigar-catalyst", which can be compensated by using simultaneously applying a higher cell density and a larger diameter (pancake-type), as shown in figure 8.

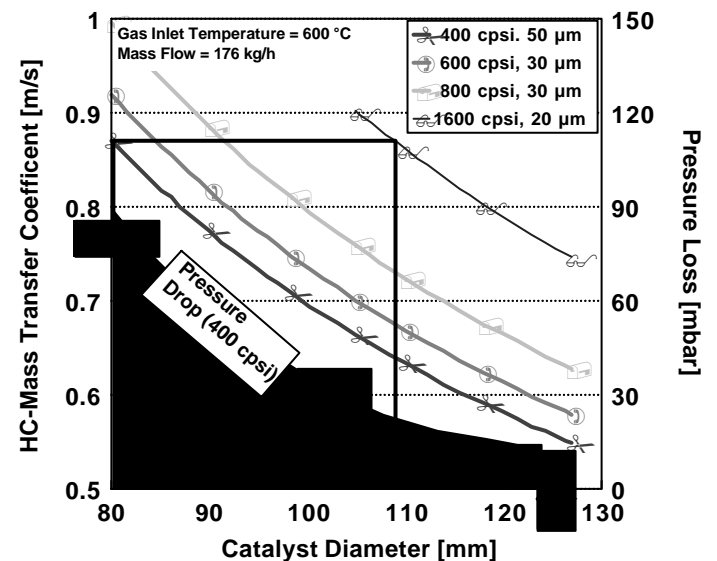


Figure 8: HC mass transfer coefficients as a function of cell density and of the catalyst diameter (gas velocity in catalyst channel), pressure loss of a 400 cpsi structure

Since a catalyst with smaller diameter produces better exhaust gas test results during the cold start phase (see above), a properly designed cascade system with at least two stages will be advantageous. Ideally a catalyst system is to be designed in such a manner that it is possible to meet all emission standards to be fulfilled in the course of the production period of the engine without changing the converter housing. Consequently efficiency has now be regulated merely by varying cell density, foil thickness of the catalyst substrate, as well as by varying the catalytic coating.

3. TEST PROGRAMMS

In order to illustrate the relationships presented in chapter 1, emission tests were conducted on a dynamic engine test bench. The test engine used was the 2.4 litre, 5 cylinder Volvo engine with a modified ULEV engine management system. The engine was equipped with a 5-in-1 manifold which allowed the catalyst to be located approximately 500 mm downstream from the engine exhaust channels. Exhaust gas emissions were recorded and analysed both modally - pre and post catalyst at a sampling rate of 2 Hertz - and as bag results. Background HC-emissions in the range of 0.8 up to 1.2 ppm C₃ were measured and were taken into account evaluating the emission bag results.

In figure 9 the HC engine out emissions, the pre-catalyst gas temperature and the catalyst's structure temperature (10 mm behind the frontal area) for the first 60 seconds of the FTP Test are plotted.

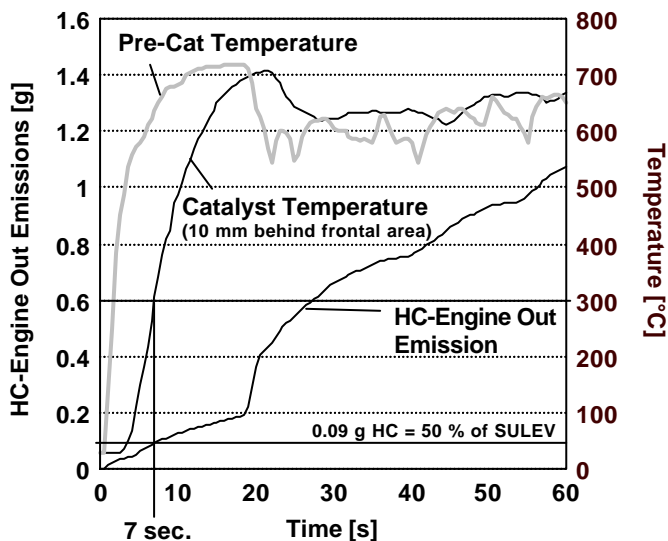


Figure 9: HC engine out emissions, temperature in front of catalyst and catalyst's structure temperature for the first 60 seconds of the FTP-cycle

Due to the modified engine management, the catalyst's structure temperature achieves the level of 300 °C representing the light-off temperature after 7 seconds.

According to the engine-out emissions this would exhibit a cold start emission result of 50 % of the SULEV standard for HC emissions, assuming 100 % conversion rate immediately after light-off.

3.1 INFLUENCE OF CELL DENSITY

As an initial step the influence of cell density at a constant catalyst volume of 1.64 litres was observed. For these catalysts, the efficiency factors (GSA / d_h) were calculated and are demonstrated in table 1, together with the physical properties of the substrates.

Size [mm]	Cell Density Foil- Thickness	Volume [l]	GSA [m ²]	Thermal Mass [J/K]	GSA / d _h [m ² /mm]
Ø 118 x 150	93 (600) / 0.03	1.64	6.19	689	7,22
Ø 118 x 150	124 (800) / 0.025	1.64	7.09	681	9,47
Ø 118 x 150	155 (1000) / 0.02	1.64	8.01	641	12,14
Ø 118 x 150	186 (1200) / 0.02	1.64	8.79	680	14,55
Ø 118 x 150	248 (1600) / 0.02	1.64	9.97	750	19,02

Table 1: Physical properties of substrates used in the comparison to test the influence of the substrate's cell density in the FTP-tests

As already discussed, higher cell-densities exhibit the disadvantage of higher back pressure, but better mass-transfer. The accumulated HC emissions (unweighted) during the cold-start phase (Bag 1), at normal operating temperature (Bag 2) and over the entire test are shown in figure 10.

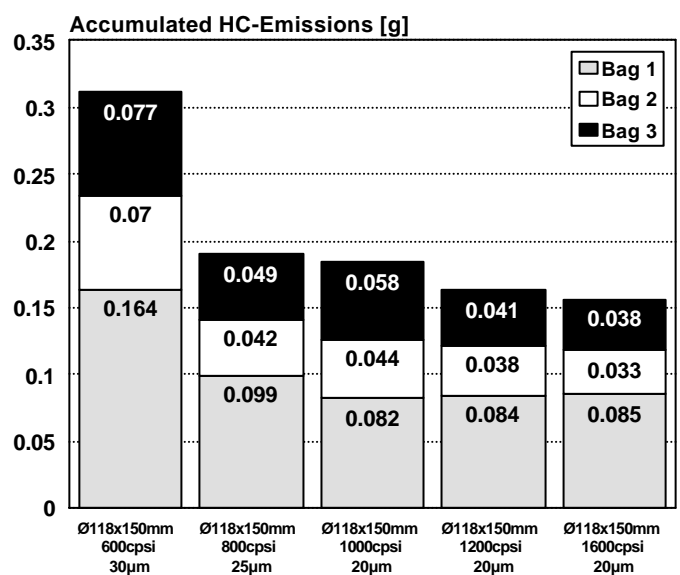


Figure 10: HC emissions during the cold start phase (bag 1), at normal operating temperature (bag 2) and during the warm start phase (bag 3) of the FTP-cycle

Additionally, the influence of cell density on NO_x-emissions in the individual phases of the FTP-cycle are demonstrated in figure 11.

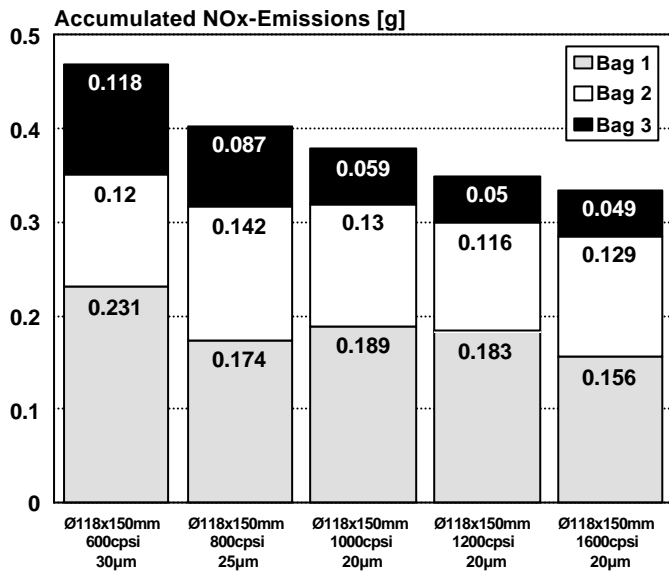


Figure 11: NO_x emissions during the cold start phase (bag 1), at normal operating temperature (bag 2) and during the warm start phase (bag 3) of the FTP-cycle

As indicated especially in figure 10, there is a strong reduction of the cold start emission when increasing the cell density while simultaneously reducing the foil thickness. No further improvement can be achieved by further increasing the foil thickness without using thinner foils (1200 cps and higher). Nevertheless, during warm operation (bag 2) the decisive number for optimum conversion is represented by the hydraulic diameter, decreasing between cell densities of 600 and 1600 cps, and resulting in a continuous decrease of bag 2 emissions. Not exactly in the same extend, but at least in tendency, these results are being confirmed by the NO_x-emissions, displayed in figure 11.

3.2 INFLUENCE OF CATALYST DIAMETER

As a second step, catalysts offering similar pressure loss, but with varying catalyst diameters and cell densities were investigated. The reduction of diameter results in a higher flow velocity and consequently in a compensation of the less favourable mass-transfer, which can be found in the case of lower cell densities.

Table 2 shows the physical properties of two of the tested catalysts having been elected for this comparison.

Size [mm]	Cell Density Foil- Thickness	Volume [l]	GSA [m ²]	Thermal Mass [J/K]	Pressure Drop *) [mbar]
Ø 118 x 150	93 (600) / 0.03	1.64	6.19	689	103
Ø 105 x 150	78 (500) / 0.03	1.30	4.59	498	108

*) p = 1.2 bar, T = 750°C, Massflow = 500 kg/h

Table 2: Physical properties of substrates used in the comparison to test the influence of the catalyst diameter in the FTP-cycle

Figure 12 shows the test result as accumulated HC-emissions in bag 1, bag 2 and bag 3 of the FTP-cycle.

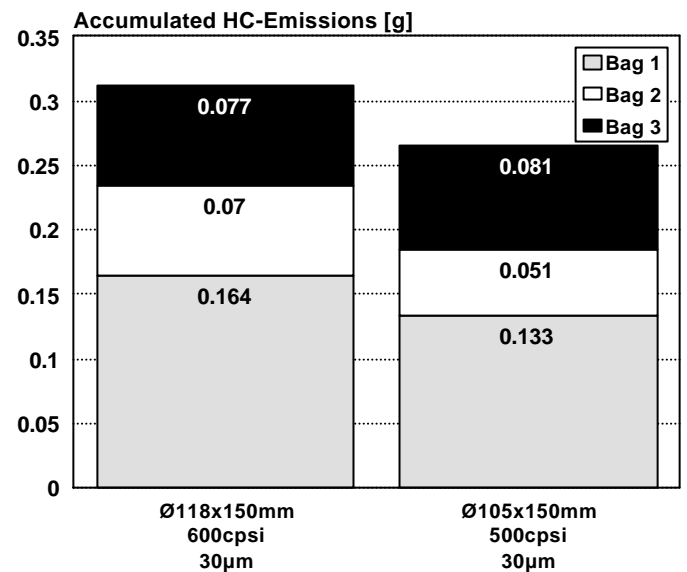


Figure 12: Influence of the catalyst diameter on the HC emissions in bag 1, bag 2 and bag 3 of the FTP-cycle

As a result, the smaller catalyst with diameter 105 mm with a catalyst frontal area being 20% smaller compared to the converter with diameter 118 mm, shows a FTP-bag 1 result and an overall test result which are significantly lower.

Both the influence of cell density and catalyst diameter revealed that a high-cell density, cigar-shaped catalyst system is advantageous in terms of catalyst efficiency in warmed-up condition, but being in disadvantage in terms of pressure-loss. However, the analysis of the exhaust gas mass flow, displayed in figure 6, shows that a catalyst with variable diameter – corresponding to the respective exhaust gas mass flow – would be ideal.

Transferred to reality such a system can only be produced as a so called “cascade”- system, which comprises single catalyst bricks with tiered diameters or conically shaped catalyst bricks.

3.3 INFLUENCE OF CASCADE SYSTEMS

A cascade catalyst system comprising a cylindrically shaped substrate as well as a conically shaped converter ("ConiCat") as the first brick, completed with an second brick with a larger diameter, listed in Table 3 together with the 600 cpsi single brick catalyst as a reference, were tested in the FTP-cycle.

Size [mm]	Cell Density Foil- Thickness	Volume [l]	GSA [m ²]	Thermal Mass [J/K]
Ø 118 x 120	93 (600) /0.03	1.64	6.19	689
Ø 90 x 50.8 + Ø 118 x 120	93 (600) / 0.03 + 155 (1000) /0.02	0.32 + <u>1.31</u> = 1.63	1.22 + <u>6.49</u> = 7.71	136 + <u>513</u> = 649
ConiCat Ø 90/114 x 45 + Ø 118 x 120	124 (800/500) / 0.03 + 155 (1000) /0.02	0.37 + <u>1.31</u> = 1.68	1.48 + <u>6.49</u> = 7.97	163 + <u>513</u> = 676

Table 3: Physical properties of cylindrically and conically shaped substrates used in cascade catalyst systems for testing in the FTP-cycle, a 600 cpsi single brick catalyst as a reference

The FTP cold start emissions of both of the cascade catalyst systems during the first 125 seconds of the test are plotted in figure 13, compared to the test results on the single brick catalysts Ø 118 x 150 mm; 600 cpsi and Ø 105 x 150 mm; 500 cpsi.

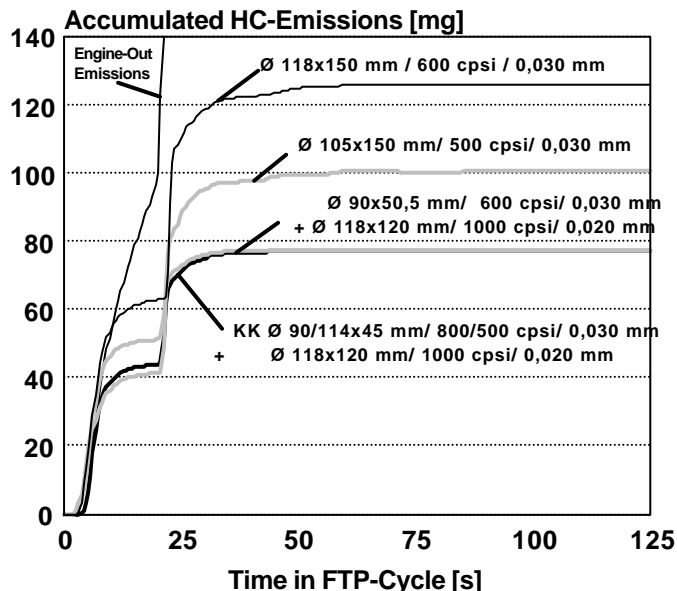


Figure 13: Accumulated cold start HC-emissions of cascade catalyst systems compared to one-brick catalysts during the first 125 seconds of the FTP-cycle

Based on the plotted test results, it can be the first conclusion, that – as shown above – reducing the

catalyst diameter without changing cell density and foil thickness improves cold start behaviour dramatically. Secondly, increasing the cell density while keeping foil thickness constant, as in the case of conically shaped cascade KK Ø 90 mm with 800/500 cpsi, the advantage of the higher GSA is compensated by an increased heat capacity, resulting over all in the same cold start result as showed by the second cascade catalyst Ø 90 mm + Ø 118 mm.

The next step now was to compare the overall test results in the FTP-cycle of several one- and two-brick catalysts, as listed in table 3. Figure 14 shows the weighted emission results.

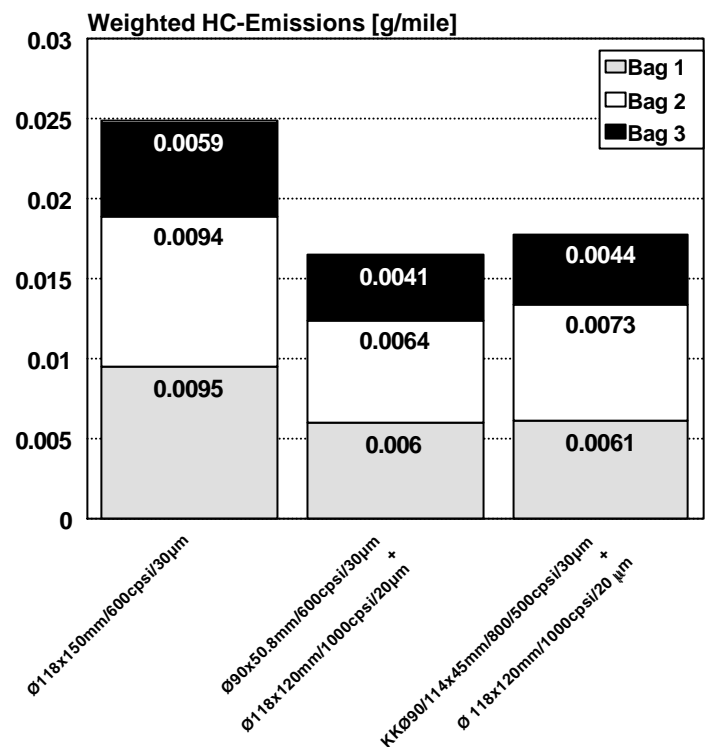


Figure 14: Overall FTP-test results for both of the cascade systems and for a single brick catalyst Ø 118 x 150 mm with a comparable cell density of the first brick

As indicated by the bag results, there seems to be no significant difference between both cascade catalyst systems, as already explained in figure 13. Additionally, both cascades are clearly advantageous compared to the 600 cpsi single brick catalyst with diameter 118 mm, due to the smaller diameter of the first brick at comparable cell densities.

Therefore, with similar cell densities and overall catalyst volumes, it seems to be advantageous to divide the catalyst volume in two pieces in order to receive best cold start performance and mass transfer.

3.4 EMISSION MEASUREMENTS AT A HIGH-LOAD

Since usually emissions being tested only in special exhaust gas test cycles, as for example the FTP-cycle, the question was of interest, what maximum conversion rate of pollutants is achievable using today's catalyst technology. That can be seen also from the background that high speed driving, which is not part of an emission test, should not cause pollution of the environment. To simulate that, the catalysts were tested at a constant load of 200 kg/h exhaust gas mass flow.

The resulting HC, CO, and NOx conversion rates are displayed in Figure 15.

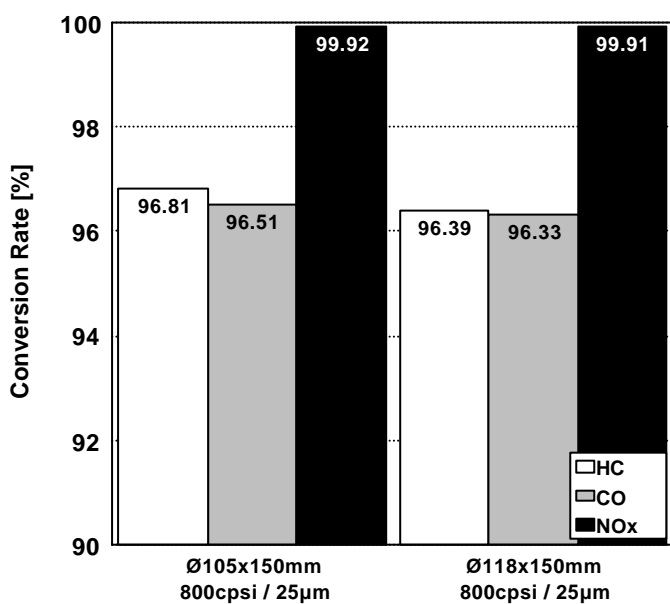


Figure 15: HC-, CO- and NOx-conversion rates of various test catalysts at a high load point with 200 kg/h exhaust gas mass flow

The catalysts compared to each other were converters with 1,3 resp. 1,6 l volume, with a cell density of 800 cps. For HC, CO and NOx, no influence of the catalyst volume could be found. Therefore it can be concluded, that even a small volume converter reveals very high efficiencies of close to 97% resp. 100% in high load points.

4. SUMMARY

To achieve future emission standards as for example SULEV or NZEV, beside the cold start, the efficiency in warmed up conditions becomes more and more important. Conversion rates of close to 100% have to be achieved. Examinations and test results of various catalyst systems exhibited following conclusions:

- For cold start both the thermal mass (cp) and the geometric surface area (GSA), together describing the Cold Start Factor GSA / cp , is of major importance
- Cascade catalyst systems with smaller substrate diameters of the first brick improve cold start behavior at a constant cell density
- The catalytic efficiency in warmed-up condition depends mainly on the mass transfer which can be described as diffusion rate
- Increasing the flow velocity in the channels for example by using smaller catalyst diameters improve mass transfer and increase back pressure
- By using smaller catalyst diameters the cell density can be reduced to achieve the same mass transfer rate
- Back pressure relates directly to mass transfer and catalyst efficiency
- Higher cell densities (smaller hydraulic diameters, d_h) result in better mass transfer and therefore in improved catalyst efficiency under warmed-up conditions
- The catalytic efficiency in warmed-up condition can be described with the help of the Efficiency-Factor GSA / d_h
- Even under high load conditions like driving on the highway small catalysts with a volume of 50 – 60% of the engine displacement conversion rates of over 96 % in HC and CO and close to 100% in NOx

An optimized catalyst layout of this kind results in lower system costs; above all due to less need of precious metals.

6. REFERENCES

1. Maus, W., Brück, R.; Emitec GmbH: "Motornahe Katalysatorsysteme; neue technische Herausforderungen"; 18. Internationales Wiener Motorensymposium, 17. April 1997, Vol. I
2. Liebl, J., Erhard, O., Albrecht, F., Zinecker, R.; BMW AG: "Entwicklung von Abgasnachbehandlungskonzepten für die zukünftigen Gesetzgebungen"; MTZ 58 (1997) 12
3. Pfalzgraf, B., Rieger, M., Ottowitz, G.; Audi AG: "Close coupled catalytic converters for compliance with LEV/ULEV and EG III legislation – Influence of support material, cell density and mass on emission results"; SAE 960261
4. Umehara, K., Makino, M., Brayer, M.; NGK Insulators; Becker, E. R., Watson, R.; Environex Inc.: „ Prediction of catalytic performance for ultra thin wall and high cell density substrates"; SAE 2000-01-0494
5. Shinichi Kikuchi, Seiji Hachio, Tatsuya Okoyama, Shyoji Inose; HONDA; Kouichi Ikeshima; NGK Insulators: "High cell density and thin wall substrate for higher conversion rate catalyst", SAE 1999-01-0268
6. Lafyatis, D.S., Will, N.S., Martin, A.P., Rieck, J.S., Cox, J.P., Evans, J.M.; Johnson Matthey: "Use of high cell density substrates and high technology catalysts to significantly reduce vehicle emissions"; SAE 2000-01-0502
7. Umehara, K., Yamada, T., Hijikata, T., Ichikawa, Y., Katsube, F.; NGK Insulators: "Advanced ceramic substrate: Catalytic performance improvement by high geometric surface area and low heat capacity"; SAE 971029
8. Gulati, S. T.; Corning Inc.: "Design Considerations for Advanced Ceramic Catalyst Support"; SAE 2000-01-0493
9. Noriyuki Kishi, Shinichi Kikuchi, Norio Suzuki, Tadayoshi Hayashi; Honda R&D Co.: "Technology for Reducing Exhaust Gas Emissions in Zero Level Emission Vehicles (ZLEV)"; SAE 1999-01-0772
10. Brück, R., Diewald, R., Hirth, P., Kaiser, F.W.; Emitec GmbH: "Design criteria for metallic substrates for catalytic converters"; SAE 950789
11. Leveroni, E., Saroglia, G., Rossi, R.: "Catalyst concepts for small volume SI-engines: The example of the 1.2 l – Punto"; Haus der Technik, Essen, 1996
12. Bremer, H., Wendlandt, K.-P.: "Heterogene Katalyse"; Akademie Verlag, Berlin