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Next generation catalysts are turbulent: development of support and coating

M. Bollig, J. Liebl, R. Zimmer
BMW Group

M. Kraum, O. Seel, S. Siemund
Engelhard Technologies GmbH

R. Brück, J. Diringer, W. Maus
Emitec GmbH

ABSTRACT

Future catalyst systems need to be highly efficient in a limited packaging space which normally leads to a design where the flow distribution in front of the catalyst is not perfectly uniform.

Measurements on the flow test bench show that the implementation of perforated foils for the corrugated and flat foils has the capability to distribute the flow within the channels in radial direction so that the maximum of the given catalyst surface is of use, even under very poor uniformity indices. Therefore a remarkable reduction in back pressure is measured. Emission results demonstrate cold start improvement due to reduced heat capacity.

The use of LS – structured (Longitudinal structured) corrugated foils creates high turbulence level within the single channels. The substrate lights-up earlier and the maximum conversion efficiency is reached much quicker. It can be shown that a 300 cpsi Metalit with 50 µm foil thickness creates the same efficiency during the European Driving cycle like a 600 cpsi with ultra thin wall, even at reduced catalyst volume. The combination of LS type corrugated foil with perforated flat foils further improve catalyst efficiency.

INTRODUCTION

The development of substrates with cell densities up to 1600 cpsi have improved the cold start behavior and also the mass transfer during warmed-up condition [1, 2] Today one can find various cars in serial production that use cell densities up to 1200 cpsi in conjunction with a cascade design where the first small catalyst enhances the heating-up due to increased flow velocities [3].

The developments of high cell densities and at the same time reduced wall thicknesses have been proven to be used in two directions: the increased efficiency leads on one hand to a reduction of precious metals [4]. On the other hand the increased conversion efficiency is reflected in a volume reduction of the catalyst system. In [5] various cell densities from 600 up to 1600 cpsi have been investigated in close-coupled and as well in toeboard position.

The tailpipe emissions of the close-coupled system during the FTP test cycles are shown in graph 1 together with the emission limits BIN 8C. The results of NMHC and CO are almost constant for the all cell densities despite a 32% reduction in catalyst volume. The nitrogen oxide emission has been proven to be sensitive to small changes of the engine management system.

Up to now it could not fully be clarified whether a further increase of the cell density leads to the necessary chemical and thermodynamical improvements to further optimize the catalytic systems. On one hand there is clearly a limit in the necessary reduction of the foil thickness. Also the effect of inhomogeneous flow distributions or tolerances in the air-fuel-ratio can be contrary to an increase in efficiency. An increase in the counter pressure of the monolith can not be accepted.

Under normal driving conditions the gas flow through smooth channels shows laminar characteristics resulting in thick boundary layers which tends to a limited mass transfer from the gas to the wall. In [6] a revised design of metal support " SQ-design" where the walls are broken off and laterally offset after a certain length to form a new passage and therefore create thin boundary layers and turbulence on the surface. This structure on the other hand never went into production because the coating resulted in accumulations of washcoat at each offset of the channel.

Various solutions to create a non – laminar flow with the use of structured metal foil is discussed in this paper. The so called "Perforated Foil" for example leads to a reduction of the heat capacity and also to higher conversion rates in the heated-up condition due to increased mass transfer within the substrate.

IMPLEMENTATION OF METALITS WITH STRUCTURED FOILS

Perforated foils (PE – catalyst)

To disclosure additional physical / chemical potential to enhance the efficiency of a catalyst, the possibility of internal flow across the single channels needs to be investigated. It is the obvious disadvantage of various catalytic systems that an uneven flow distribution in font of the substrates is fixed within the monolith and therefore expensive 2–brick–systems are used to allow additional mixing before the inlet of the second substrate. A transversal motion between the channels and therefore a distortion of the laminar flow would result in an increased efficiency.

The development of metal foils with perforation [Fig. 1] has promising advantages:

- homogeneous distribution of flow and concentrations
- compensation of lambda variations
- reduction of heat capacity

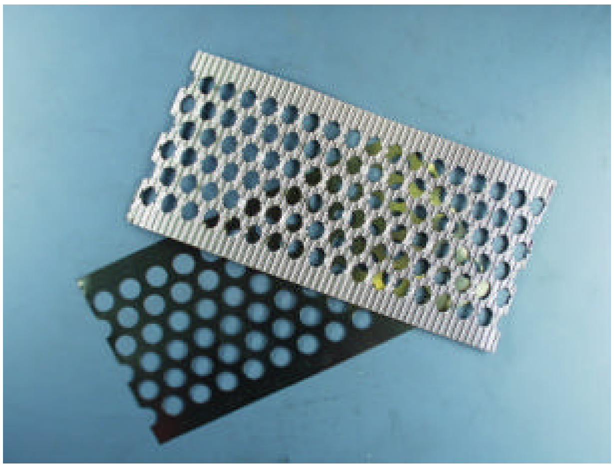


Figure 1: Perforated flat and corrugated foil

It is obvious that the perforation on the other hand leads to a significant loss of geometric surface area. This investigation will show the loss of surface can be compensated by the creation of a " turbulence-like " flow within the channels.

Calculations of the flow within a channel

The influence of the perforation was investigated with a CFD-model (KIVA-II). The calculation shows the velocity vectors and also the pressure gradient of two adjacent channels [Fig. 2]. The boundary conditions were set in a way that the difference of the flow velocities between these channels was 2 m/s before reaching the first perforation. The foil thickness was 50 µm with a coating thickness of 25µm on each side.

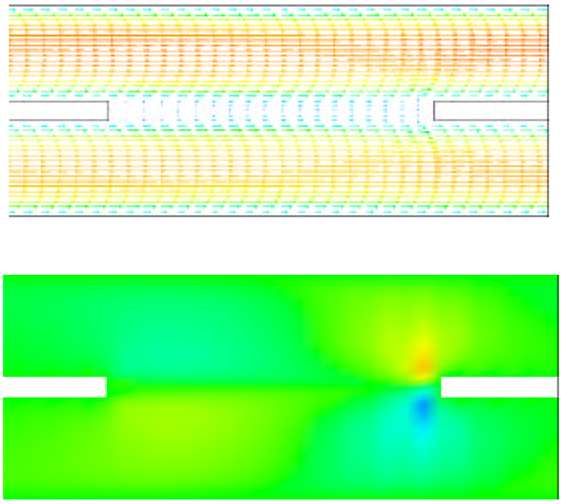


Figure 2: CFD-modelling of a channel with perforation

It is obvious that after 1/3 of the diameter of the hole the flow dives into the opening. The profile of the flow shows an interaction between both channels. The axial flow is disturbed and a radial component is developed so that a pressure gradient is created which leads to an increased mass transfer.

Development of a coating process for structured foils

Perforated foil substrates as well as any combination with otherwise structured foils substrates require a modified process due to the constraints of open channels.

Figure 3 shows the result from a regular prototype production. The caverns formed by the perforation led to plugging as well as to an uneven distributed washcoat.

Modifying the coating process resulted in the expected and desired appearance of the catalytic layers (fig. 4).

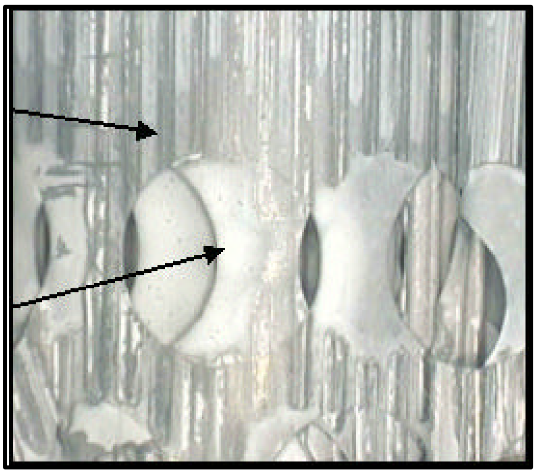


Figure 3: Impression of washcoat appearance for the non-optimized coating process

Modifying the coating process resulted in the expected and desired appearance of the catalytic layers (fig. 4).

Figure 4: Impression of washcoat appearance after process optimization

A similar improvement in the coating results were seen for the combinations of LS with PE designs. LS only type substrates can be produced with todays regular processes.

Influence of the perforation on the flow distribution

The influence of the perforation on the flow distribution and also on the back pressure was investigated by using different substrates with a various grades of perforation / porosities [Tab.1]:

Ø95x101,5mm 1200 cpsi, 40µm	Metalit 1	Metalit 2	Metalit 3
Hole diameter [mm]	----	8 mm	4 mm
Porosity [%]	0 %	35 %	19 %
Geom. surface area [m²]	3,57	2,32	2,89

Table 1: Tested substrates

The measurement was performed on a test bench using a manifold with 3 cylinders creating an almost concentric flow into the converter with a mass flow rate of 300 kg/h and a temperature of 100°C. All substrates have been coated with a loading 150 g/ft³ and with a constant washcoat thickness and therefore lower absolute washcoat mass. The systems have been aged according the procedure of the German Car Consortium at a maximum temperature of 950°C.

In a first test the uniformity index of the flow was 0,92. Fig. 5 shows the flow distribution measured at the outlet of the substrates.

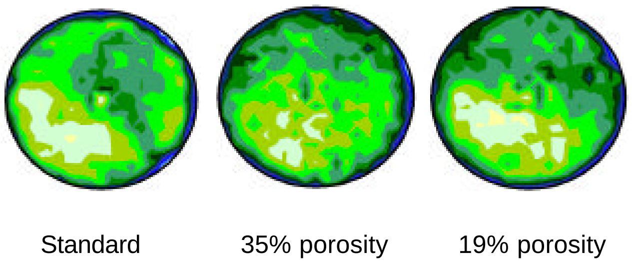


Figure 5: Flow distribution of the tested Metalits

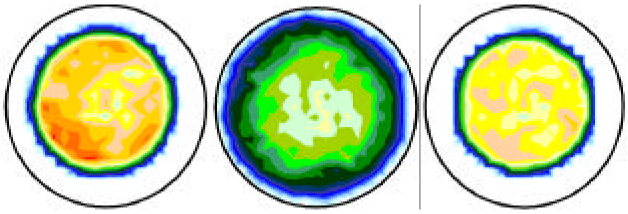
In case of the standard substrate the flow is directed into the lower left part of the substrate inlet and shows a concentration of high flow velocities. It can be clearly seen that using a Metalit with a porosity of 35% the area of high velocities is distributed more equally while a porosity of 19% does not clearly show a major difference. Nevertheless the measurement of the pressure drop of each system show a remarkable reduction which can be referred to the homogenization of the velocities of each channel [Tab. 2].

In order to simulate an uneven flow the inlet side of each Metalit was covered by ring of 15 mm around the outer edge of the substrates. The flow was therefore forced into the middle of the substrate with a free cross section of dia 65 mm. The results of these measurements are shown in Fig. 6.

Ø95x101,5mm 1200cpsi, 40µm Porosity [%]	Metalit 1 0 %	Metalit 2 35 %	Metalit 3 19 %
Uniformity Index [-]	0,92	0,92	0,92
Back Pressure [mbar] at 300kg/h	18,4	14,4	16,5

Table 2: Results of flow measurements

The flow velocities increase essentially in case of the non perforated substrate with a similar concentration as seen in the tests before. With the support of the 35% perforation it is possible during the length of 101,5 mm to distribute the flow again towards the substrate mantle and enhance the measured uniformity index by around and consequentially the back pressure by around 40% [Tab. 3]. The lower porosity again is only able to equalize the area of the high velocities which has no visible effect on the distribution but leads to a almost 20% reduction of the pressure drop.



Standard 35% porosity 19% porosity

Figure 6: Flow distribution of the tested Metalits

95 x 101,5mm, 1200cpsi,40µm	Metalit 1	Metalit 2	Metalit 3
Porosity [%]	0 %	35 %	19 %
Uniformity Index [-]	0,49	0,78	0,49
Back Pressure [mbar]	28,5	15,4	23,1

Table 3: Results of flow measurements at low

Influence of the perforation on heat capacity

The perforation of the flat and the corrugated foils reduces the thermal mass of the Metalits accordingly. In Tab. 4 the influence of the perforation of 35% on the heat capacity is shown for a substrate 600 cpsi with various foil thicknesses. Measurements have proven that a that using a thickness of 50µm with the perforation leads to a 3,5 sec reduction during cold start to reach light-off temperature. It is therefore possible to build a 600 cpsi with perforated 50µm foil at constant heat capacity compared to the 30µm foil.

	600cpsi 40µm	600cpsi 30µm	600cpsi 50µm perforated	600cpsi 40µm perforated
Heat Capacity [J/K]	0,500	0,410	0,405	0,355

Table 4: Influence of perforated flat and corrugated foils on the heat capacity (loading 150 g/ft³)

Emission results

The emission measurements have been performed by Engelhard Technologies on an 6-cylinder, 3,0 l engine with ULEV application during the European driving cycle. The substrates have been tested with the standard non-perforated foil as well as with perforation of 19% and 35%. It was of major interest whether the improvement in the mass transport within the channels was able to compensate the loss of geometric surface area.

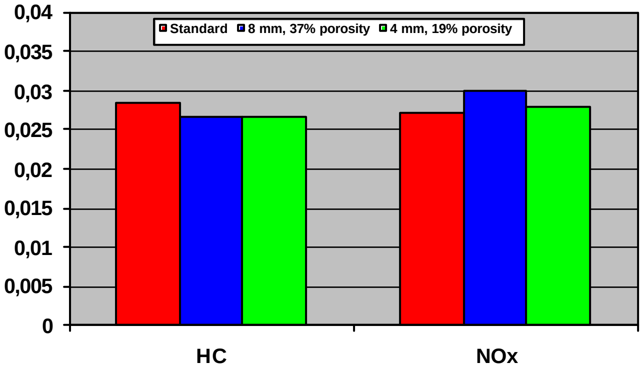


Figure 7: Accumulated emission results during the EU driving cycle

The advantage of the heat capacity reduces the cold start HC emission for both of the perforated systems. The total results on the other hand are not only determined by the heating-up behavior. Due to the limited mass transfer during heated-up conditions plays the velocity of the diffusion the major role in the process especially when the concentrations are low. Since the advantage in HC emission which was achieved during cold start is not lost during the complete cycle it can be shown that the loss of surface area is compensated by the creation of a radial flow and therefore improved mass transfer to the walls.

The conversion of the NOx emission is not driven by the heat capacity, but by the surface area and the velocity of diffusion. The results show a clear function of the porosity which also proves that the distortion of the flow within the channels and the equal distribution of the concentrations can almost balance the loss of 19% resp. 35% of surface area.

Implementation of Metalits with LS structured foils

The first idea to enhance the diffusion process within the channels was created already in 1994 when Emitec developed the Transversal Structure (TS) [7, 8]. The counter corrugation leads to radial flow components which let to increased conversion efficiency [Fig.8]

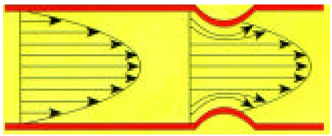


Figure 8: Flow field within a channel using the TS - structure

This technology is widely used in the industry either to increase the conversion rates at a given substrate design or to reduce the cell density and therefore the back pressure of the system at a constant emission performance.

To further improves the formation of a turbulence-like flow in the channels, the flow should be forced to change the channels by an appropriate geometry. Such structures are already known since several years but the use was very limited since the coating technique was not suitable so far. But new production technologies which has been developed in recent years prove the implementation of structured foils in the near future.

Fig. 9 shows an example of such structures, the “Longitudinal Structure LS“ where the geometry of the channel is slotted and broken up by a counter corrugation in axial direction. So the laminar flow is interrupted and especially at low concentrations the diffusion process is not longer limited to the velocity.

The look into the inlet side of an LS – structured Metalit with 300 cpsi leads visually to the impression that, due to the counter corrugation a Metalit with 600 cpsi is used. For an examination to show its potential in a comparison of a 600 cpsi substrate a 300 LS was used [Tab. 7].

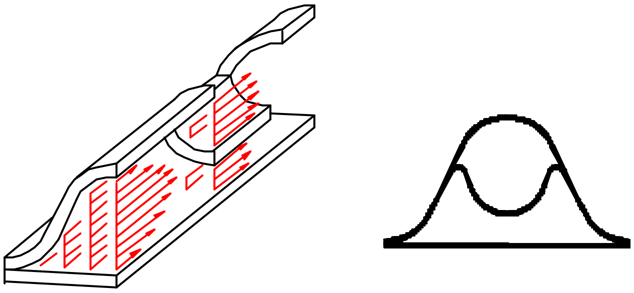
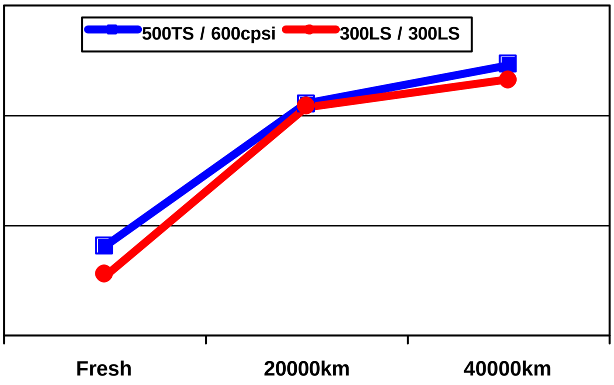


Figure 9: Schematic of LS – structured foils

First emission experience with LS-structure

The serial production system of the BMW 3,0 l engine consists of a cascade design Ø 80 x 50,8 mm, 500TS



followed by Ø 98,4 x 101,5 mm, 600 cpsi. This system was compared to a cascade of the same dimensions where both substrates were built of 300 LS-structure. The systems were measured in fresh conditions and then aged in a car using a high load driving cycle.

The emission results are shown in fig. 10 for Hydrocarbon and for NOx in fig.11.

Figure 10: Hydrocarbon emission during the FTP-test cycle

The performance of the 300LS Metalits behave equal or even partially better than the substrates with the higher cell densities. A replacement of todays systems by reduced but structured cell densities is therefore possible.

Figure 11: NOx emission during the FTP-test cycle

Further Emission testing

New systems were also tested in a car with a 4-cylinder, 1,6 l engine. The manifold uses a 4-into-1 manifold with a close-coupled main converter [Fig. 12]. The design of the exhaust system shows a slight bend to the right in front of the converter, so that the flow can be expected to be guided mainly in that direction.

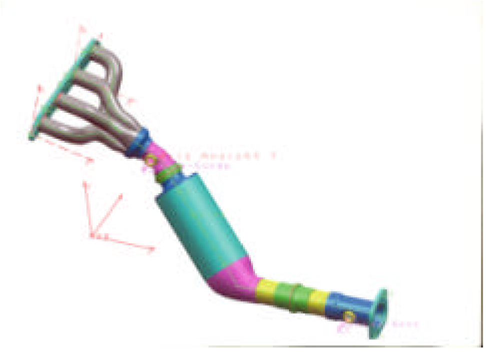
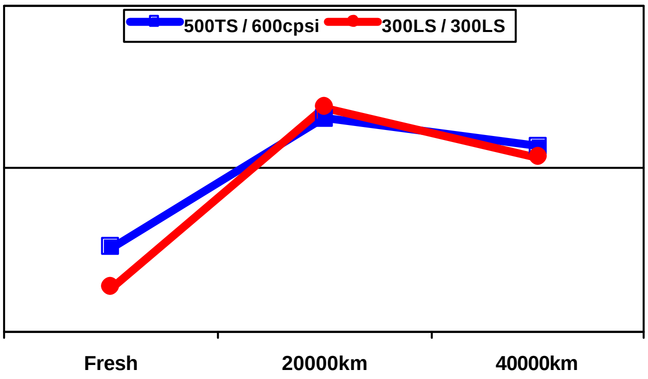


Figure 12: Catalytic exhaust system

The substrates have been zone-coated: 50g/ft³ in the first 50 mm, 20 g/ft³ in the remaining area. The aging procedure of the German Car Consortium was again used with a max. temperature of 950°C



	Volume [l]	Heat capacity [kJ/K]	Geometric surface area [m²]
Ø105x135mm 300LS, 50µm	1,17	0,49	3,21
Ø105x150mm 600 cpsi, 3 mil	1,33	0,52	4,39

Table 7: Physical data of the tested systems

The results shown in Fig. 13 demonstrate the immense potential of the slotted foils: despite the fact that in case of the LS structured 300 cpsi support a 25 % reduction of the surface area (due to the reduced cell density **and** reduced volume) is realized all emission results are almost similar. The production of turbulence and the split-up of the thick boundary layers results in an intensive exchange of the exhaust gas between the core stream and the surface.

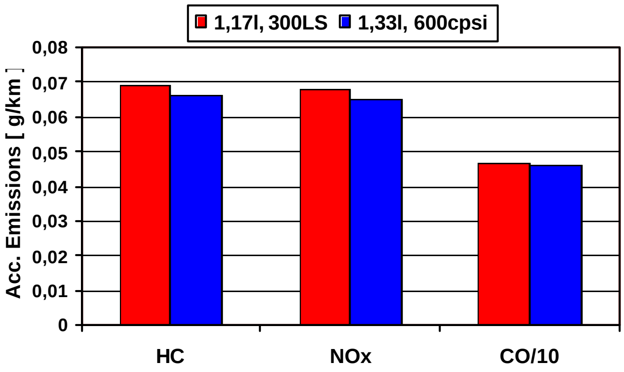


Figure 13: Emission results during the EU driving cycles

This technology now offers a similar potential like the various high cell densities: the structured foils can either result in a lower catalyst volume or a reduced loading of the support. Along with this option goes an almost 30% cost saving of the support material.

Emission Measurements with slotted LS foils in combination with perforated flat foils

The use of the slotted LS structure allows an exchange of the flow only between neighboring channels driven by the turbulent forces. The success of implementing perforated foils to further enhance the radial flow through the catalyst led to the idea to combine both structures. A smooth perforation was used for the flat foils with a hole diameter of 8 mm and a low porosity of around 6 % of the total surface area [Tab. 8].

The measurements of the converter efficiency have been again tested in the EU driving cycle and compared to the previous test results [Fig. 14].

	Volume [l]	Heat capacity [kJ/K]	Geometric surface area [m²]
Ø105x135mm 300LS, 50µm	1,17	0,49	3,21
Ø105x135mm 300LS/PE 6%, 50µm	1,17	0,40	2,62
Ø105x150mm 600 cpsi	1,33	0,52	4,39

Table 8: Physical data of the tested systems

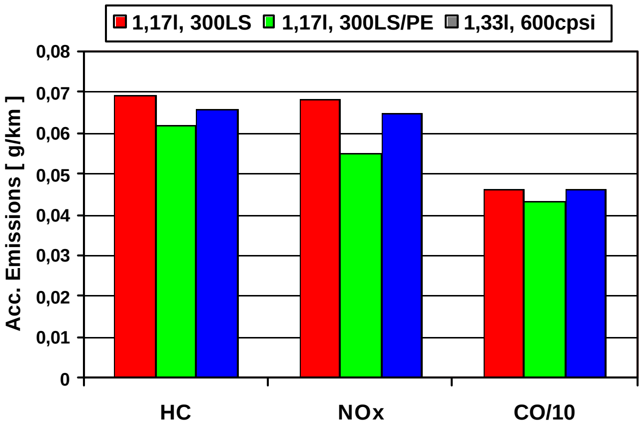


Figure 14: Emission result od a combined LS / PE substrate compared to standard and only LS-structured supports

The additional implementation of the perforation to the flat foils leads again to an improved HC and CO emission of almost 10%. Since the reduction of heat capacity is quite small (just 6% porosity), the major effect of the increased performance appears to be a smoother aging due to a rather optimized flow distribution through the substrate. The improvements in NOx emission are around 20%. The perforation allows the exchange of the flow through the channels so that a better use of the given surface is possible which leads to an improvement in NOx emission of around 20%.

In a next step these investigations will be repeated with an increased porosity up to 15% and also cell densities from 200 to 400 LS.

CONCLUSION

The new technologies Perforated Foils and LS-structure for metal substrates offer a variety of potential improvements for the next generation of catalysts.

The major goal for future supports must be to break up the laminar flow within the channels and create at least a turbulence-like flow to enhance the mass transfer. The perforation of the metal foils constitutes a first step to an internal, radial flow exchange between the channels. The intense interaction between the flow and the wall allows the same efficiency like a standard substrate despite a 35% reduction in surface area. Due to the lower heat capacity the perforation ultra thin foil not necessarily need be used for high cell densities and allows additional temperature resistance e.g. for high temperature turbo engines. Also the back pressure is remarkably reduced.

The slotted LS – structure disturbs the flow right in the middle of the channel to break up the boundary layers and creates a turbulent flow at each slot. The conversion efficiency is therefore much higher so that a reduction in the cell densities is possible eg. 300 LS instead of a 600 cpsi. Additionally even the catalyst volume can be reduced compared to the standard system.

These new types of metallic substrates with structured foils require, based on their new designs, innovative catalyst technologies as well as a modified coating process. The set goals for product and process development were met: the loss of GSA for the PE and the LS/PE type substrates could be compensated. Even the reduced cell dimensions for LS and LS/PE type substrates, in comparison to regular flow through substrates, resulted in an improved performance.

ACKNOWLEDGMENTS

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