

Corrugated Cross Flow Catalyst with Permeable Walls

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ABSTRACT

This paper presents a new metallic catalyst substrate design that allows an integrated mixing behaviour and flow homogenisation within the substrate. In addition it offers a high mass transfer from the flow to the interior substrate surface.

The design consists of corrugated walls with crosswise orientation of adjacent layers. The walls are permeable so that their high specific surface is directly accessible for the flow and consequently the length for diffusion is reduced and the mass transfer is enhanced. The crosswise corrugated design causes the intensive internal mixing of the flow within the substrate. Thereby local differences in velocity as well as in temperature and in concentration are equalized.

Thus the new metallic substrate design concept offers homogeneous reaction conditions throughout the entire substrate. It allows a compact design and in case of a Diesel oxidation catalyst an additional soot reduction due to the design with permeable walls.

INTRODUCTION

For decades ceramic monoliths have been used for catalytic converters in gasoline cars. When Diesel oxidation catalysts were introduced the monoliths were adapted to fit this application as well. Additionally for some years metallic monoliths have found a number of applications. They feature a lower heat capacity and therefore a faster light-off and - depending on their design - a turbulent flow inside the catalyst for higher mass transfer coefficients [1, 9]. Different measures to improve the macroscopic mass transfer and achieve a certain mixing within the whole substrate have been reported in literature [1, 9]. Enhancing mass transfer will allow the catalyst substrate volume to be reduced at the same overall conversion [9, 10]. At the same time it is aimed at an improved usage of the active sites at to allow a lower amount of precious metal.

As space requirement in close coupled arrangement is a key issue in both current and future applications, a

compact catalyst design is required which additionally is insensitive to short inlet cones and non-uniform inflow.

In the following a new approach for metallic substrates is described which complies with these requirements. These structures offer a high mass transfer, an intensive mixing characteristics and an additional soot reduction.

CHARACTERISTICS OF CROSS CORRUGATED STRUCTURES

Structures of corrugated layers with alternating orientation of adjacent layers (called cross corrugated structures here) are well known in chemical industry and process industry applications. They have been used for plate heat exchangers, regenerators, mass transfer packings and static mixers for years [3]. Cross corrugated structures coated with a catalytic active layer have been suggested in literature as well. Results on these catalytic cross corrugated structures have been reported in [2], [3] and [4]. Those investigations show that these structures feature a high mass transfer coefficient. Furthermore they show a good radial heat transport as well [4].

The properties of these structures have to be adapted to the specific application here. The left hand side of figure 1 shows an example of these cross corrugated structures. On the right hand side of the picture a scheme of the cross corrugated structure with some of the major geometrical parameters is shown. These comprise the amplitude a , the wavelength λ and the corrugation inclination angle φ . A smaller value of φ means a stronger deflection of the flow while an angle of 90° would result in channels parallel to the flow direction.

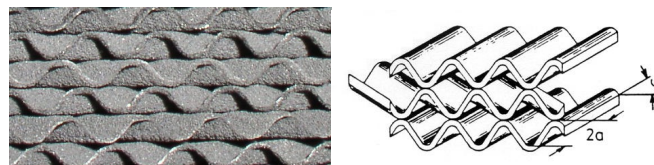


Figure 1: Example of cross corrugated structure

Heat and mass transfer as well as the pressure drop and the mixing characteristics of the flow in cross corrugated structures can be varied in a wide range by proper choice of these parameters.

High local heat and mass transfer

The effects of geometrical parameters on the heat and mass transfer coefficients in these structures have been reported in [2]. By suitable design these structures feature a high heat and mass transfer.

Figure 2 shows a comparison between the local mass transfer in a cross corrugated structure and a square-duct monolith. Both structures have been cut open to investigate the transport phenomena at the surface. A method for visualisation and determination of local mass transfer has been used in these experiments [2, 3]. The local colour intensity on the surface corresponds to the locally transferred mass or in the case of a catalyst application to the local reaction rate.

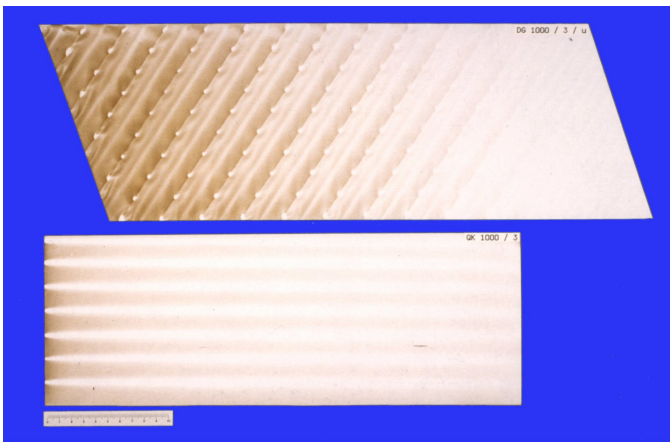


Figure 2: Local mass transfer distribution in a cross corrugated structure compared to a monolith, from [2]

In the case of the square-duct monolith there is a high mass transfer at the front of the structure. Along the flow route the mass transfer declines due to the laminar flow conditions in the square duct. Towards the end of the structure local mass transfer attains a constant but low value.

In the case of the corrugated structure the intensive colour indicates the high mass transfer. The higher mass transfer is due to the turbulent flow regime in these cross corrugated structures. In addition the rather homogeneous colour density distribution shows the homogeneous local mass transfer distribution.

Along the flow route the colour intensity declines due to impoverishment in mass concentration. The light colour intensity in this case corresponds to a decreased concentration of the reactive species. This is proved in figure 3. There a part of the corrugated structure has been placed behind the square duct monolith and then the experiment has been performed in the same way as

before. The dark colour on the corrugated structure proves that at the outlet of the square duct monolith there is still a high mass concentration of the reactive species. So in case of the square duct monolith the light colour is due to the low mass transfer coefficients.

The darker colour of the cross corrugated structure already qualitatively demonstrates the significantly higher mass transfer of cross corrugated structures compared to monoliths. Quantitative measurements on the mass transfer are reported in [3].

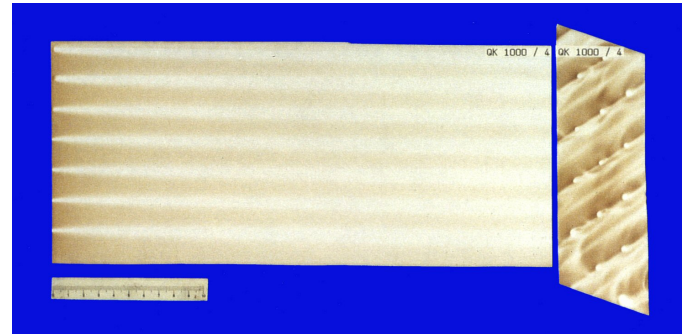


Figure 3: Higher overall mass transfer in a cross corrugated structure, from [2]

Flow with intensive mixing characteristics

The turbulent flow inside the cross corrugated structure is not only responsible for a high mass transfer coefficient to the wall surface but additionally for a high turbulent exchange term inside the fluid phase. The alternating orientation of the corrugated layers opens up flow areas which have a different flow direction in each layer and therefore a zone of high shear stresses in-between. This leads to an excellent mixing and flow homogenisation.

Figure 4 shows an experiment with a local tracer added locally to the flow in a corrugated structure. As can be seen from figure 4 there is an intensive mixing of the flow and after a short length the tracer is distributed uniformly across the entire structure.

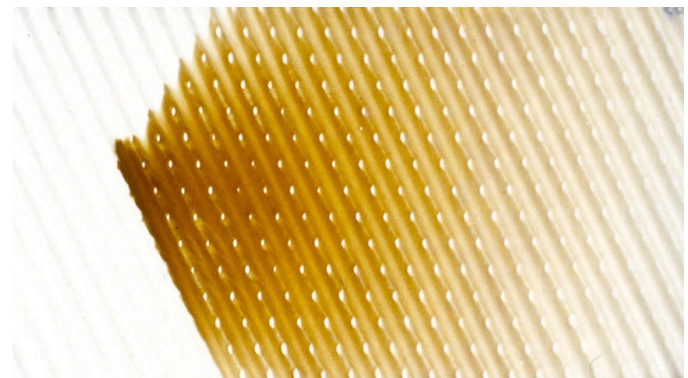


Figure 4: Intensive fluid mixing in a cross corrugated structures, from [3]

The mixing characteristics of the cross corrugated structure does not only equalise the flow field inside the

structure but also homogenise local differences in temperature and concentration of the oncoming flow. Disadvantageous inlet conditions of the flow can be equalised thereby and thus do not have such a big impact on the overall reaction conditions as they do have in the case of square duct monoliths.

CROSS CORRUGATED STRUCTURES WITH PERMEABLE WALLS

The examples up to now have shown cross corrugated structures with non permeable walls. Most commonly they are manufactured of metallic sheets.

The investigations in this work add a new feature to these structures: the walls are made of permeable material. Thus the flow is offered a third way through the walls. This flow is driven by the local pressure difference between luff side and lee side of the corrugated walls. Another driving factor is the local flow inhomogeneity of the oncoming flow. The additional path ways are shown schematically in figure 5.

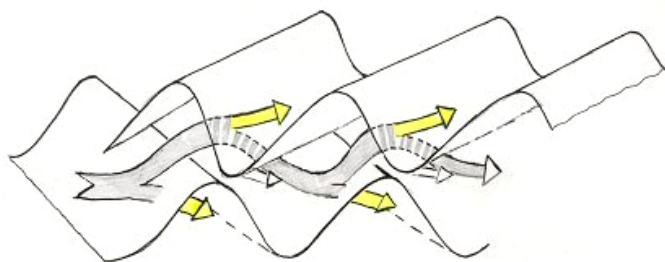


Figure 5: Permeable cross corrugated structure: convective flow through the wall

The flow through the wall opens new possibilities for the transport processes to the reaction sites [7, 8]. This offers new potential for these structures as described in the following.

In general the conversion rate in a catalytic monolith divides in kinetic regions as shown in figure 6. At low temperatures the reaction rate on the catalytic surface is too low because the energy of the molecules is too low to overcome the activation energy of the specific reaction. Although the reactants are accessible at the surface the reaction rate is slow. With increasing temperature the reaction onsets until the consumption of one of the reactants by the reaction equals its transport to the surface by pore diffusion. Latter is enhanced by the further increasing temperature but is still limiting the overall reaction rate. With further increased temperatures the pore diffusion processes quickens and the mass transfer in the channels from the main flow to the pores becomes smaller than the possible pore diffusion transport. Then the mass transfer coefficient in the channels limits the overall reaction rate.

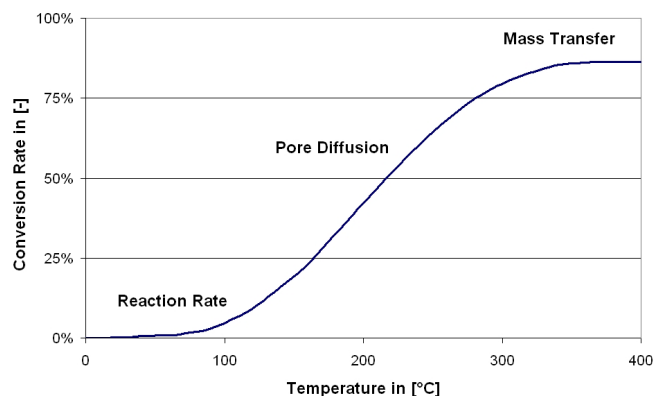


Figure 6: Kinetic regions of a catalyst, according to [11]

Overcoming mass transfer limitation

The use of cross corrugated structures increases the mass transfer in the channels. This has already been shown in the sections before. The use of permeable walls further increases the mass transfer due to the convective transport of the wall flow. So mass transfer limitation can be overcome by suitable choice of the structures parameters. Experiments on the conversion will be shown in a section later on.

Overcoming pore diffusion limitation

The other reaction limitation by pore diffusion as shown in figure 6 can be reduced as well. To better understand the processes in a square duct monolith figure 7 shows the processes with a fully developed laminar flow inside the channels.

The overall reaction processes in such a monolith can be divided into five main steps. The first part is the mass transfer of the reactants from the main flow through the boundary layer to the wall surface. Because of the laminar flow in the channels this mass transfer is purely based on diffusion and therefore is relatively slow. Then the reactants have to be transported from the surface of the porous washcoat to the reactive centres at the inner surface. This transport is determined by the pore diffusion coefficients. These are dependent on the washcoat temperature and the washcoat structure. It is a slow diffusion process, too, depending on the reactant concentrations on the wall surface and inside the washcoat. After the reaction took place the products have to be transported back to the wall surface by pore diffusion and finally the products have to dissolve in the main flow by diffusion. Therefore four of the five steps are characterised by diffusion processes.

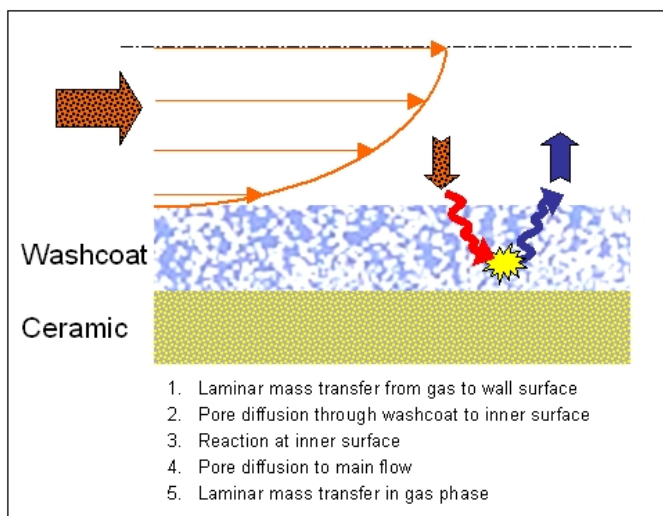


Figure 7: Square duct monolith: pore diffusion to inner surface

The porous cross corrugated structures overcome these slow diffusion processes by installing the convective wall flow as driving mass transfer process. This is illustrated by figure 8.

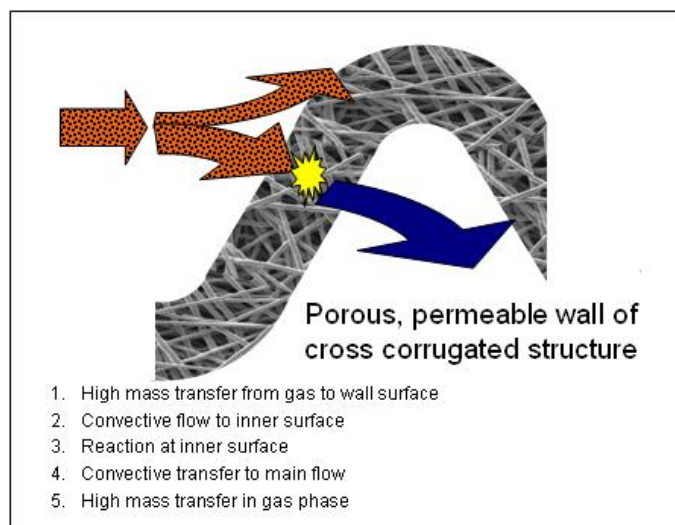


Figure 8: Permeable cross corrugated structure: convective flow to inner surface

Due to the wall flow the mass transport to the wall surface and to the catalytic centres at the inner surface is no more characterised by diffusion processes but by convection. This process is much faster. The diffusion only has to transport the reactants in radial direction from the middle of the pores to the catalytic centres on the inner surface. But these distances are extremely small compared to the macroscopic distances in the channels and in the washcoat pores themselves. So the overall reaction conditions shall be significantly increased by these porous cross corrugated structures.

To confirm these assumptions cross corrugated catalysts have been set up and compared to a conventional square duct monolith. The results of these experiments will be presented in the following sections.

SET-UP OF CROSS CORRUGATED CATALYST STRUCTURES WITH PERMEABLE WALLS

DESIGN OF MATERIAL PERMEABILITY

The basis of the porous cross corrugation structure is the permeable material of the substrate walls. Two types of materials have been examined: a metallic fibre fleece and a metallic foam. The structure of the fleece can be seen schematically from figure 8. A microscopic photo of the foam is shown in figure 9.

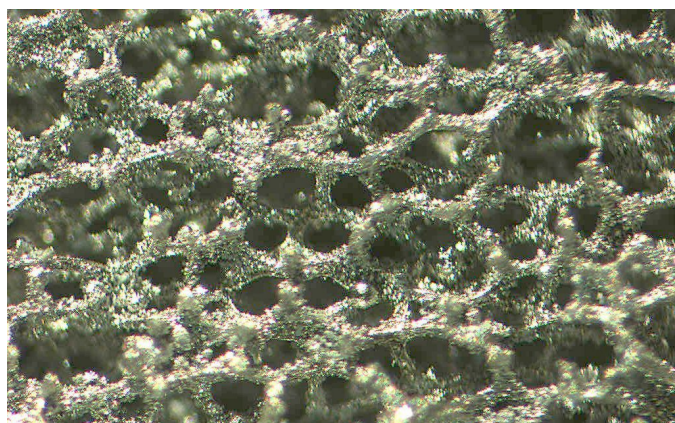


Figure 9: Porous material of the substrate walls

Both materials chosen fulfil the basic requirements. For a better performance of the structures a systematic optimisation has to be undertaken. This mainly concerns the internal pore structure. One option is the pore diameter. It affects the inner surface, the space for the catalytic coating as well as the permeability and therefore the wall flow. And beside the inner structure the material itself has to be optimised. It must fulfil the requirements of the catalytic coatability and a good washcoat adhesion as well as a good resistance concerning corrosiveness and temperature in the exhaust system. Furthermore both the inner structure and the material have to meet the manufacturing requirements. On the one hand it shall be able to be corrugated without damaging the inner structure and on the other hand it shall withstand the mechanical stresses of the canning and the operation without being damaged even at high temperatures.

The currently used felts and foams have been selected from the manufacturers programs. They fulfil the basic requirements but have not yet been tested for all emerging exhaust conditions. The necessary material optimisation has to be undertaken in close cooperation with the porous media manufacturer.

DESIGN OF GEOMETRIC PARAMETERS

Besides the material optimisation the structural parameters have been optimised as well. After some pre-orientating experiments to show the general suitability of the porous cross corrugated structures, a number of catalysts with different geometric parameters has been set up. For the first orientation experiments a rather dense fleece has been used, building rather big channels and featuring rather small orientation angles φ . For the following samples of structures a finer fleece as well as a fine foam has been used. The catalytic activity of these will be presented in the following section. Nevertheless the channels of the porous catalysts examined up to now are wider than those of a monolithic catalyst. For a direct comparison a Cordierite catalyst with a cell density of 400 cpsi has been examined as well.

The cross corrugated catalyst supports have been set up in a round shape with a diameter of 4.66 inch and a length of 3 inch. An example is shown figure 10. A cordierite catalyst of the same size has been used as reference sample. In addition a cross corrugated support with non-permeable walls made of metal sheets has been set up as well to examine the effects of the porous wall.



Figure 10: Setup of the permeable cross corrugated substrate (example: foam catalyst)

In total five cross corrugated catalysts with different parameters have been set up for the experimental series presented here. All of them have the same amplitude of $a = 1.5$ mm. One has been set up of metal sheets, one consists of a metal foam and three are built from a metal fleece. The parameters of the samples set up are shown in table 1.

No.	walls	material	angle φ	amplitude
1	non permeable	metal sheet	50°	1.5 mm
2	permeable	foam	51°	1.5 mm
3	permeable	fleece	39°	1.5 mm
4	permeable	fleece	49°	1.5 mm
5	permeable	fleece	62°	1.5 mm
6	Cordierite monolith, 400 cpsi			

Table 1: Parameters of samples

The small differences in the inclination angles are due to the manual setup of the supports in this early stage. The Cordierite monolith has a amplitude of its channels of 0.6 mm compared to 1.5 mm for the corrugated structures. The metallic substrates have been canned in the usual way using a fibre mat.

CATALYTIC COATING

The set-up of the catalyst shown in figure 10 already is a catalytic coated one. The six catalysts considered in this paper are all coated as similar as possible. Due to restrictions in the geometrical surface of the monolith and the porosity of the permeable cross corrugated catalysts respectively the amount of washcoat on the catalysts had to be adapted. So in case of the monolith a higher amount of washcoat had to be used than on the porous structures which pores would have been closed in case of too much washcoat. But the composition of the washcoat and especially the composition of the active precious metal and its amount have been kept identical for all catalysts. Therefore comparable reaction behaviour of the coatings can be assumed.

The set-up of the cross corrugated structures in layers opens new ways of coating. While a monolith has to be coated as a whole, the new supports can be coated in two different ways. The traditional way would be the same coating technique as for the monolith in coating the mounted support as a whole. The other possibility is to coat the layers individually and mount the support out of coated layers. The benefit of the second method is that the coating will be more uniform without washcoat accumulation in any corners. The coating method to prefer may be decided by the coating manufacturer.

PARTICULATE MATTER REDUCTION

Another feature of the porous cross corrugated supports is that they offer an additional reduction of particulate matter.

As outlined before the main characteristic of the porous cross corrugated supports is the convective flow through the porous walls. On the one hand side this enhances the mass transfer to the active surface. On the other hand side this also will result in a particulate matter reduction by these porous structures. As a defined part of the flow passes the wall, a part of the particulate

matter will deposit within the porous wall. There it will be oxidised either thermally by oxygen or by NO₂, formed at the catalytic coating.

Concerning the PM oxidation by NO₂ the porous cross corrugated supports feature an improved NO_x-cascade as outlined in figure 11. The exhaust flow passes through the porous walls and NO can be oxidised to NO₂ by the catalytic coating in the porous wall. Downstream the NO₂ can oxidize soot while it gets reduced to NO. Passing a catalytic active site it will be oxidised to NO₂ again. Further downstream the NO₂ again will oxidise soot. Then it can once more pass through the porous wall and thereby can be re-oxidised to NO₂ which means another chance to oxidise the soot particulates. So the overall balance can be more than one oxidised soot molecule by one NO₂-molecule. So the amount of the NO_x-cascade is related to the number of wall passages. Thereby the amount of the wall flow component is a major parameter for this kind of soot oxidation and the parameters of the permeable cross corrugated structure have to be designed according to that.

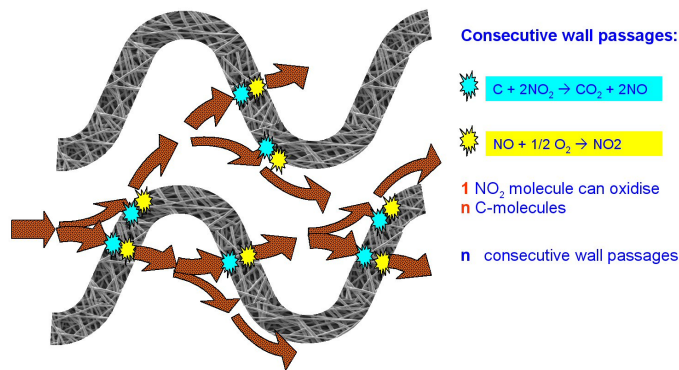


Figure 11: Enhanced NO_x-cascade by consecutive wall passages

The enhanced NO_x-cascade is of special interest at engine calibrations with a low NO_x emission, as it allows a high PM conversion at a low NO_x concentration. Measurements on particulate matter reduction by cross corrugated catalyst supports will be shown later on.

FLOW DISTRIBUTION IN POROUS CROSS CORRUGATED STRUCTURES

The mixing characteristics of the flow in cross corrugated structures with non-permeable walls have already been outlined in the first chapters. In the following section the effects of the wall permeability on the flow distribution in cross corrugated substrates will be looked at.

CFD calculation

A numerical study using computational fluid dynamics has been carried out to determine the flow inside the corrugated structures. The resulting flow field inside the

channels is shown in figure 12 for a specific set of parameters.

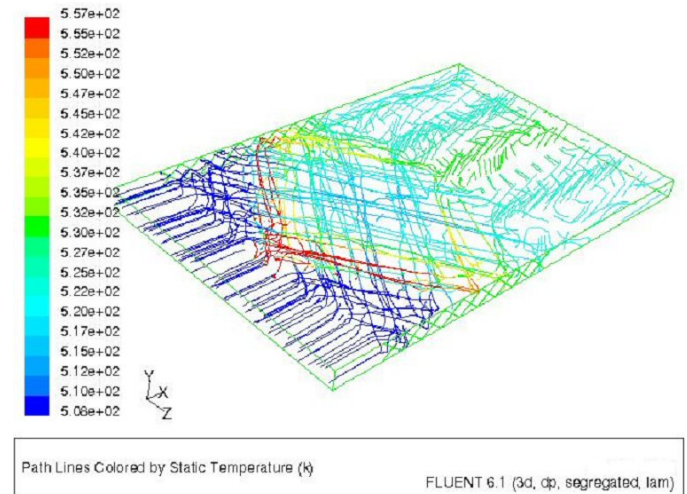


Figure 12: Flow distribution and fluid mixing inside the permeable corrugated structure, from [5]

In this example the flow enters the computational region from the lower left side with a temperature of 235 °C with a superficial velocity of 4.4 m/s. These conditions are typical for engine operation points with partial loads. The walls on the side of the computational region are treated as insulated walls while the borders on the top and at the bottom are linked in a periodic boundary condition so that the structure segment is repeating endlessly in the y-direction.

The structure simulated here consists of a length of 85 mm at a width of 118 mm. The walls have been modelled with the permeability of the material used for the experimental setup [5]. The flow enters the structure and is diverted as shown by the path lines depending on the orientation of the corrugated layers. The flow mainly flows along the corrugation towards the sides. At each crossing of the main flows a certain exchange between these flows takes place. This is the basis for the mixing characteristics of the flow in these structures.

In this example the mixing is shown for the gas temperature which corresponds to the colour of the path lines. To demonstrate the temperature homogenisation in the structure, a high heat source is assumed in the middle of the structure front. This heat source locally leads to wall temperatures of over 700 °C and gas temperatures of more than 550 °C and therefore to a temperature rise of more than 300 °C. Along the flow route the heat distributes across the entire structure. At every crossing of the corrugation the heat distributes bit by bit until the maximum differences in the gas temperature are lower than 20 °C at the end of the structure.

So the intensive mixing in these structures helps for temperature homogenisation and consequently serves for uniform reacting conditions throughout the structure.