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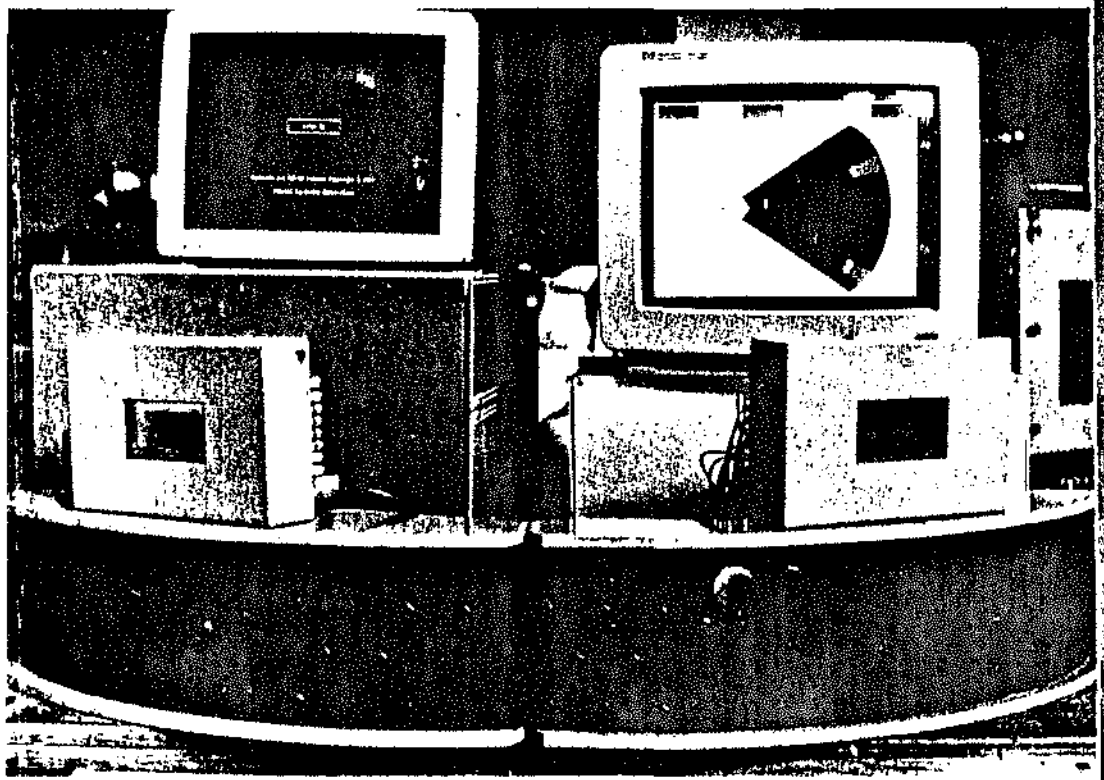
SILVER JUBILEE CONFERENCE ISATA 25 1992

PROCEEDINGS

25th ISATA Silver Jubilee International Symposium on Automotive Technology and Automation

Dedicated Conference on THE MOTOR VEHICLE AND THE ENVIRONMENT - DEMANDS OF THE NINETIES AND BEYOND

Florence, Italy 1st–5th June 1992



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Computational Fluid Dynamics and Its Application to Catalytic Exhaust Systems

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SYNOPSIS Computational Fluid Dynamics has become more accessible to engineers with the advent of more user-friendly codes. In particular, these codes are being used to solve the fluid motion, heat and mass transfer and even the chemical kinetics of catalyst exhaust systems. The Harwell FLOW3D code has proved adaptable and capable of predicting the flow, heat and mass transfer within a single channel, and of predicting the flow through a two-dimensional model of a converter.

NOTATION

ε	the void fraction of the monolith
$[X]$	concentration of species X, (mole fraction)
A_{geo}	geometric surface area per unit volume (m^2/m^3)
$k_{m,X}$	mass transfer coefficient (m/s)
h	heat transfer coefficient (J/m^2sK)
λ	thermal conductivity of substrate (J/msK)
ρ_g	density of the gas (kg/m^3)
Cp_g	heat capacity of the gas (J/kgK)
A_{cat}	catalytic surface area per unit volume (m^2/m^3)
R_X	specific reaction rate for species X (mole s^{-1} $m^{-2}Pt$)
$-(\Delta H)_X$	enthaply of reaction for species X (J/mole)
$D_{m,X}$	molecular diffusivity of X (m^2/s)
E_A	activation energy (J/mole)
Q	heat flux (W/m^2)
T_g, T_s	temperature of the gas, temperature of the solid
U_o	initial gas veolcity

1 INTRODUCTION

In the last few years, computational fluid dynamics (CFD) has evolved from a highbrow mathematical science into a useful tool for engineering. Improvements in computational power, from small personal computers to large main-frames, mean that ever larger problems may be solved. Developments in the CFD codes themselves have produced faster, more robust algorithms and the codes have become much more user-friendly, enhancing their appeal to engineers.

Researchers in the automotive emissions field are turning their attention to CFD as a means of predicting the emissions from a range of catalytic systems, without the need for expensive experimental testing. Currently, there are no accurate predictive techniques capable of handling the extremely transient nature of a catalytic exhaust system. Researchers throughout the world, however, have modelled simple catalytic systems (such as one-dimensional, adiabatic reactors), and are using them as a basis for more complex models.

Complete modelling of the whole system requires i) an understanding of the flow and heat transfer of the exhaust system, particularly in the converter; and ii) a thorough knowledge of the chemistry of catalytic converters. At this point in time, little is known about the fluid dynamics within converters, and even less about the kinetics of the catalytic reactions. As experimental programmes reveal more about both these areas, faster computers and more robust codes will be needed in order to solve transient models of ever increasing complexity.

Research engineers, Oh and Cavendish, at General Motors Research Laboratories in the USA developed their own code[1,2,3] based initially on single pellet studies. This one-dimensional model was then extended to a three-dimensional transient model through the work of Chen *et al*[4,5], where it was used on monolithic converters with flow maldistribution.

The GM approach to the transient three-dimensional problem of monolithic converters was to adopt a dual-solver technique, treating the monolith as a porous medium with gas flow. With this approach, the fluid flow and the mass transfer equations were solved, using a finite element solver, for the gases in the void fraction of the monolith. The resulting temperature and concentration fields were then passed to a separate solver which calculated the solid temperature distribution and used the reaction kinetics to calculate a new concentration field. The modified temperature and concentration fields were then passed back to the fluids solver and the process iterated to convergence. These processes were repeated for each time step in a transient calculation.

This model was the first of its kind to consider a three-dimensional non-adiabatic monolithic converter operating, transiently, under conditions of non-uniform flow. The model's predicted results gave good qualitative agreement, but owing to a lack of accurate kinetic data for the reactions considered, the quantitative agreement was only good in some areas.

Many other models have been formulated by various researchers[6,7,8,9,10,11] but these mainly considered one-dimensional models which were adiabatic and/or had uniform flow. Other workers considering three-dimensional models and/or non-uniform flow have been Flytztani-Stephanopoulos *et al*[12], Becker and Zygourakis[13], Zygourakis[14] and Leclerc and Schweich[15]. The models

have generally proved qualitatively correct, but not matching the experimental data too closely.

All of the above approaches have involved the researchers in writing and developing suitable codes of their own, a very costly and time-consuming exercise. With the advent of more user-friendly, versatile, commercially-available CFD codes, researchers are adapting these codes to the particular needs of catalytic converters. Developments are still needed in the area of reaction kinetics, whose highly non-linear mathematical equations do not easily integrate into these codes. The largest single problem in this field is the lack of reliable and accurate experimental data with which to validate any predictions.

2 PROBLEM DISCUSSION

2.1 The fluids problem

The fluid flow in typical monolith channels is known to be laminar after the initial entrance region, so modelling may begin by looking at the development of laminar flow in such channels. The boundary conditions in this case are adiabatic (Q=0), isothermal (T_g=T_s), and uniform inlet velocity ('plug flow').

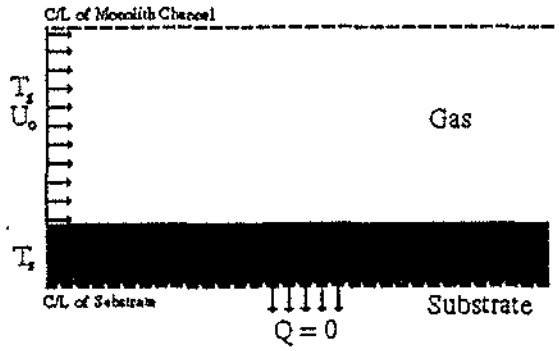

Fig 1. Adiabatic single channel model

Next, the system may be extended to be non-adiabatic and therefore non-isothermal. In this case the substrate acts as either a heat sink (for a cold (or washcoat-only) monolith) or as a heat source (hot, catalytically active monolith). For simplicity, the heat sources or sinks are assumed to be constant down the length of the monolith and constant with time (see Figure 2).

Analytical solutions exist for a variety of channel geometries (square, rectangular, triangular, round, etc), so limiting Nusselt numbers from the codes may be compared with the analytical results. The local heat transfer coefficient 'h' varies along the length of the channel, from a very high value in the developing entrance region, to the limiting value for the duct. This limiting value of 'h' may be validated with the analytical results. The heat transfer coefficient determines how the monolith temperature responds to the inlet gas temperature. This is of critical importance for light-off behaviour, particularly with twin brick systems which have developing laminar flow at the front face of each brick.

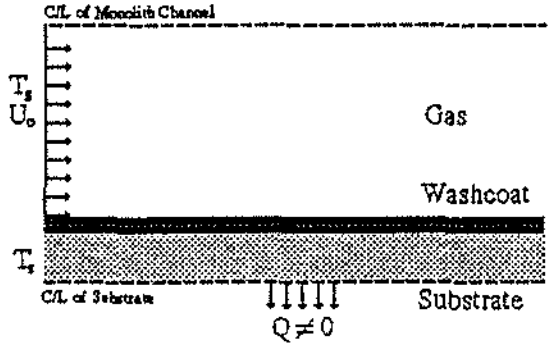

Fig 2. Non-adiabatic single channel model with washcoat

Catalytic converters on a vehicle experience heat losses from the converter (and therefore from the individual monolith channels) which change with time. The temperature distribution within a converter is a function of the mass flowrate, inlet temperature, rate of chemical heat release and rate of cooling from air motion over the external surface, all of which are extremely complex functions of each other and of time.

Owing to the physical limits of computational speed and memory, it is currently impossible to model all these transients with any degree of realism. Pseudo-steady state situations, such as vehicle cruises and idle modes, have almost constant rates of heat loss/input and so simplifying the problem to constant wall heat fluxes is, therefore, not totally unrealistic. Using the Harwell FLOW3D code, simple single channel modelling with heat sources/sinks at the walls has been achieved in a short space of time.

2.2 The chemical problem

The fluids model can be made more complex with the inclusion of mass transfer by means of the diffusion of various catalytically active species. Further, the addition of a porous washcoat adds diffusion and convection of the gas through the porous solid. Treating the system as adiabatic and isothermal, the inlet conditions are uniform inlet gas velocity and concentration of a catalytically active species X (see Figure 3).

The diffusivity of each species through the gas and through the washcoat need to be determined before the codes will produce realistic solutions. As yet, the experimental data is not capable of providing exact diffusivities of all the species under consideration, in particular their diffusion through the washcoat. An approximation is to use the diffusivity of the bulk gas in the washcoat as an average for all species present in the gas and such an assumption has to be

made while awaiting further experimental investigation. As the catalytically active precious metals are dispersed throughout the thickness of the washcoat, the active species are being converted at all depths of washcoat. Thus good models should allow conversion of the active species through the washcoat.

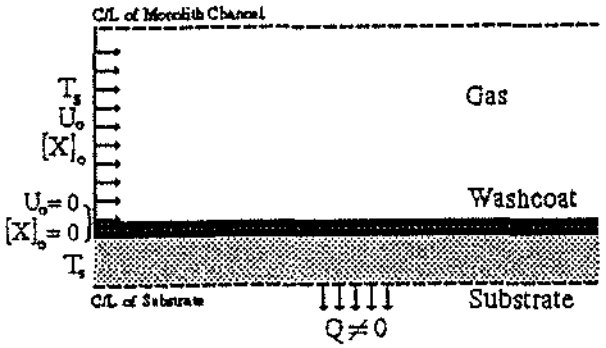

Fig 3. Non-adiabatic single channel model with washcoat and catalytically active species X

Such details have, to date, proved difficult to model. As an alternative, researchers have treated the washcoats simply as active surfaces. The influence of convection within the washcoat on the behaviour of the gases has been ignored. Species conversion at the surface may be defined as some function of both the rate at which the species makes contact with the surface, and the temperature at the surface.

Considering the thin surface to represent a catalytically active washcoat, the rate of conversion of X is proportional to the concentration gradient of species X at the surface, i.e. the flux at the surface equals the rate of conversion of X. The attendant chemical heat release is ignored initially, and included as the modelling becomes more complex. The boundary condition for this case could either be i): that

$$[X]_s = 0 \qquad (1)$$

which makes the conversion of X limited to the rate at which X can diffuse to the surface (diffusion limited),

or ii) that of a constant wall flux of species X:

$$D_{m,X} (\partial[X]/\partial y)\Big|_{surf} = a\ Constant. \qquad (2)$$

Both cases represent X being converted to an inert product of no further interest. Analytical solutions exist for both cases (and are analogous to the heat transfer cases), so the model may be tested for its prediction of $[X]_g$ exiting the channel. The mass transfer coefficient 'k' is important in determining how and where catalytic activity occurs, and the predicted limiting Sherwood number (analogous to the limiting Nusselt number) may be compared to the analytical solutions.

Making the surface catalytically active to simulate the real monolith channel requires that the flux of X at the surface equals the rate of conversion of X, which is proportional to the rate of heat generation. Here the flux of X is not a constant, rather:

$$D_{m,X} (\partial[X]/\partial x)\Big|_{surf} \propto [X]_s e^{-E_a/RT} \qquad (3)$$

Suitable rate constants, activation energies and heats of reaction are required before the models can deal realistically with the chemistry. The simulations may either be adiabatic or non-adiabatic, the latter being the closest a single channel can be to representing a real monolith. The published, but limited, experimental data of temperatures and species concentrations may be used to test model predictions.

3 MULTIPLE CHANNEL (MATRIX) MODELLING

Modelling the whole converter requires that the inlet pipe, inlet cone, catalyst monolith and outlet cone be included in the simulations. The flow in the inlet pipe is highly turbulent and unsteady making accurate predictions of the flow field at the front of the catalyst very difficult. Most researchers have ignored the pulse effects in vehicle exhaust systems, preferring to get the bulk flow correct first. An accurate prediction of the non-uniform flow distribution is necessary, because the chemical behaviour is so heavily dependent on the flow profile. Researchers Lai and Kim *et al*[16] have used a commercially available code to model the flow through various converter designs. Their results have indicated the expected extent of flow maldistribution within various converter designs. Their experimental programme will aim to validate the model predictions beyond just pressure drop correlations.

The fluids problem for gas flow within the catalyst requires a one-dimensional flow through the brick. These channels provide a resistance to the flow which is proportional to the inlet velocity and monolith length. The solid in the monolith has thermal conduction in three dimensions which must be solved simultaneously with the fluid flow.

A catalyst brick of common dimensions e.g. a round 4.66" dia x 6" long has a frontal area of 17.055 in^2. With 400 cells/in^2, the total number of cells is 6822, and, allowing a computational grid of 10 x 10 cells to be fine enough to solve each channel, this equates to 6.8×10^5 mathematical cells. Making the monolith model 100 cells long gives a model size of 6.8×10^7 mathematical cells. The largest common meshes that people currently solve for a steady state problem are of the order 1×10^5 and for transient models of the order 1×10^4.

Obviously, grids with 10^7 mathematical cells cannot be handled using current technology, so the models have to be

simplified. A first stage simplification would be to use only one mathematical cell for each monolith cell. This would require that the boundary of each mathematical cell be a 'thin surface' - the so-called 'baffle' approach. In order to represent the physical situation mathematically, conjugate heat and mass transfer coefficients have to be specified.

These coefficients are a combination of the transfer coefficient from the centre of the cell to the washcoat surface, from the washcoat surface to the substrate surface, and, for heat transfer, from the substrate surface to the centre of the substrate. Specifying these transfer coefficients for use within a multi-channel mathematical model causes problems owing to the experimental difficulty in actually measuring their values.

It may be feasible to use the CFD codes to predict these transfer coefficients for the range of velocities present in a given flow distribution (i.e. a particular catalyst at a nominal flowrate), and use these values in a multi-channel model. Such an approach is necessarily tedious, requiring that all the coefficients to be calculated for each channel of the monolith from the predictions of the single channel model.

A much simpler approach is to use the porous medium approach of the researchers from General Motors[1,2,3,4,5]. This method divorces the physical and mathematical cells completely, with the mathematical cells containing a medium of given porosity. The gas transport equations are solved for the open fraction of the media, and the heat and mass transfer equations for the solid fraction of the media.

Such a porous medium approach has the one-dimensional heat and mass transfer equations:

$$\varepsilon\rho_g C p_g \partial T_g/\partial t = -\rho_g u C p_g \partial T_g/\partial x + hA_{geo}(T_s - T_g) \quad (4)$$

$$\varepsilon\rho_g \partial[X]_g/\partial t = -\rho_g u \partial[X]_g/\partial x - \rho_g k_{m,X} A_{geo}([X]_s - [X]_g) \quad (5)$$

along with the chemical heat release equation and the three-dimensional heat conduction equation:

$$(RT_g/P_g) A_c R_X([X]_s, T_s) = k_{m,X} A_{geo}([X]_g - [X]_s) \quad (6)$$

$$(1-\varepsilon)\rho_s \partial(C_s T_s)/\partial t = (1-\varepsilon)(\lambda_x \partial^2 T_s/\partial x^2 + \lambda_y \partial^2 T_s/\partial y^2 + \lambda z \partial^2 T_s/\partial z^2) + hA_{geo}(T_g - T_s) + A_{cat}(-\Delta H)_X R_X([X]_s, T_s) \quad (7)$$

The present state of catalyst modelling indicates that commercial codes can be adapted to the catalytic converter and produce useful results in a reasonable timeframe. There is an undoubtable learning curve for these codes, but in no way is it comparable to writing a code from scratch. The state of the art modelling is at the level of predicting the drops) and getting some way into non-adiabatic converters with or without some kinetic chemistry.

In order to progress from the current 'build and test' philosophy for designing catalyst exhaust systems to a more theoretical approach, engineers and researchers need predictive tools whose answers can be translated into production. Such tools will not be available for several years to come, but current modelling techniques are now laying the foundations for them.

The adaptation of the CFD codes to this problem has already shown a capability to predict flow maldistribution within the converter[16]. With reliable experimental data to back up such predictions, CFD may be used to improve canister design. Using the predictions of flow distribution, CFD codes will be used to predict converter performance in terms of heat transfer and/or mass transfer. Such models are mostly of a steady state nature, with transient models less well developed, owing to their computational expense.

4 FLOW3D Theoretical Predictions

The Harwell FLOW3D code was used to predict the development of the laminar velocity profile for compressible flow between parallel plates. The plates were 0.001 m apart (consistent with the size of a monolith channel) and the inlet velocity was set to 5 ms^{-1} (consistent with typical monolith channel gas velocities) and at 600K. The walls of the channel had a constant positive heat flux of 1500 Wm^{-2}, to crudely simulate a chemical exotherm in the channel.

The results in Figure 4 show the development of the laminar flow profile in the channel. The increase in magnitude down the channel is caused by the increasing temperature lowering the gas density. Figure 5 shows the development of the temperature profile at the same points along the channel.

The transition from the plug profile (at the inlet) to the parabola (for fully developed flow) exhibits peaks which gradually merge and produce the parabola. This effect was noted by Wang and Longwell[17] when they numerically evaluated the development of the parabola for an incompressible Newtonian fluid. In their analysis, the pressure gradient $\partial P/\partial y$ (and thereby the momentum equation for u_y) was not necessarily small in the entrance region and so could not be ignored. As CFD may be considered to be a generalised extension of their numerical technique, it should not be too surprising that FLOW3D produced the same results.

The FLOW3D code has also been used to predict the flow profile within a two-dimensional slice of an elliptical type catalyst. A thin slice through the converter has been taken with the x-direction defined as the direction of flow and the y-direction as the major axis of the converter ellipse. The z-direction bounding planes have been declared as planes of symmetry, so the model is not a true representation of the

catalyst. However, if the slice is thin compared to the other major dimensions, the behaviour of the physical system is nearly two-dimensional, and so the model is not without value.

The mean gas velocity through such a slice will be higher than the average for the whole brick because of wall effects. The mean velocity through the two-dimensional slice will be nearly twice the true mean for the brick, implying that the predicted pressure drop will be nearly twice that of the experimental pressure drop.

The catalytic converter model used in these predictions was taken from a two-dimensional slice through an elliptical (6.68" x 3.18") monolith brick 5" long. The included angle for both the inlet and outlet cones on the major catalyst axis was 60°. This configuration was chosen to match experimental data from Johnson Matthey[18]. Using air at ambient conditions, the flow profiles were measured in the inlet cone 4 cm from the front face of the catalyst, and in the outlet cone 1 cm from the rear face. The profiles were measured for a range of space velocities (45000, 67500 and 90000 GHSV (Gas Hourly Space Velocity)) along with the pressure drop across the converter.

The FLOW3D code was then used to predict the flow distribution through the 2-D slice model. The inlet and outlet profiles shown in Figs 6, 7, 8 and 9 are consistent with the positions used to obtain the Johnson Matthey experimental data. The predicted results agree well with their experimental data.

5 CONCLUSIONS

The main conclusions from this work show that it possible to take a commercially available CFD code and adapt it to the problem of catalytic converters. There are, of course problem areas, which may be different for various codes, but these can be overcome. The learning period for a CFD suite is at least six months for someone with no prior experience, and a year or more for the user to become totally proficient i.e. be able to generate an excellent mesh with the refinement in the necessary areas. At least one User Course is recommended after about a month of familiarisation with the code.

The representation of the porous catalyst medium has proved non-trivial within the FLOW3D code. The abrupt pressure change in the flow caused by the catalyst results in the model easily diverging if the porous medium is not handled correctly. Forcing the flow to be one-dimensional through the monolith is also non-trivial within FLOW3D, though the code seems otherwise very robust. Large changes in neighbouring cell sizes has not caused any problems.

Refining the grid near the wall definitely improved convergence, indicating that it is important to solve the turbulent boundary layer well. A long inlet pipe upstream of the inlet cone was used to ensure that a fully developed turbulent profile was achieved (from the plug profile specified at the beginning of the pipe) at the inlet cone. This improved the convergence and resulted in a well defined recirculation pattern.

Full three-dimensional models are of course necessary in order to truly match any experimental data, though the models are an order of magnitude more difficult to set up and refine as well as for the code to solve. Two-dimensional models are not without value, and may be used as quick models to 'tune' parameters, grid, etc., prior to a three-dimensional model.

Many solutions have only concentrated on predicting the flow profiles within the converters - a much larger area lies in getting the codes to predict the chemical kinetics. The lack of good experimental data means that progress in this field will not be easy, but researchers throughout the world are looking at these problems, so full catalytic exhaust system modelling should be possible in the future.

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Figure 4 Development of the hydrodynamic profile.

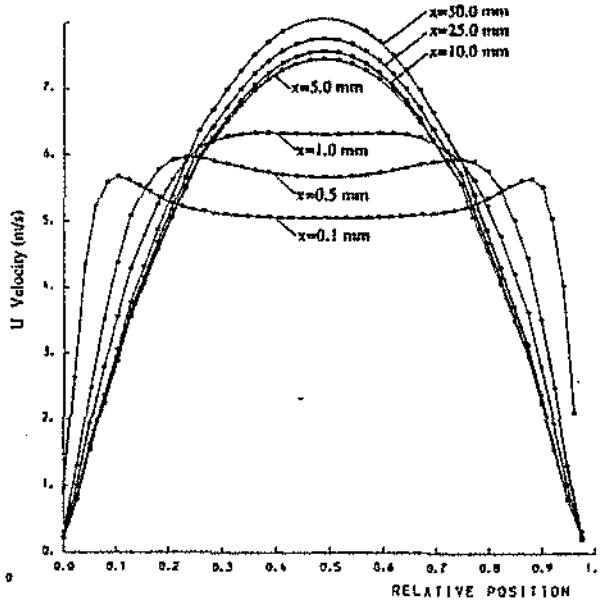
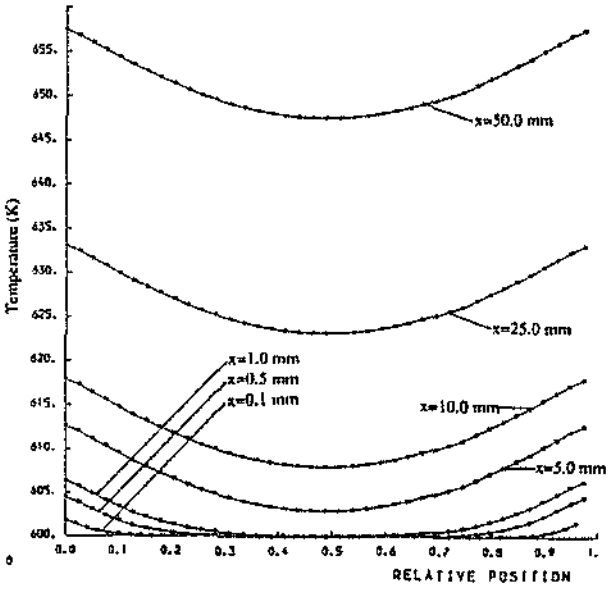


Figure 5 Development of the thermodynamic profile.



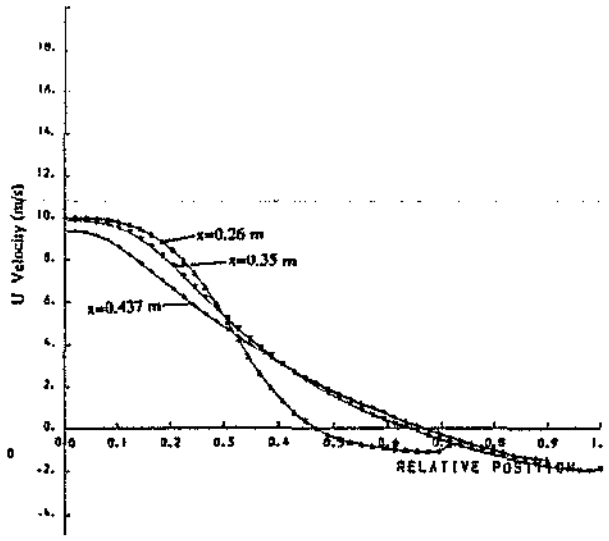


Figure 6 Two-dimensional catalyst can - no catalyst - 45000 GHSV.

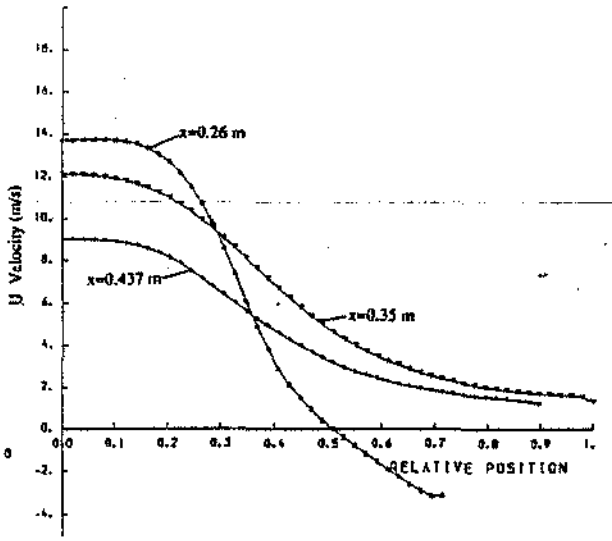


Figure 8 Two-dimensional catalyst can - with catalyst - 67500 GHSV.

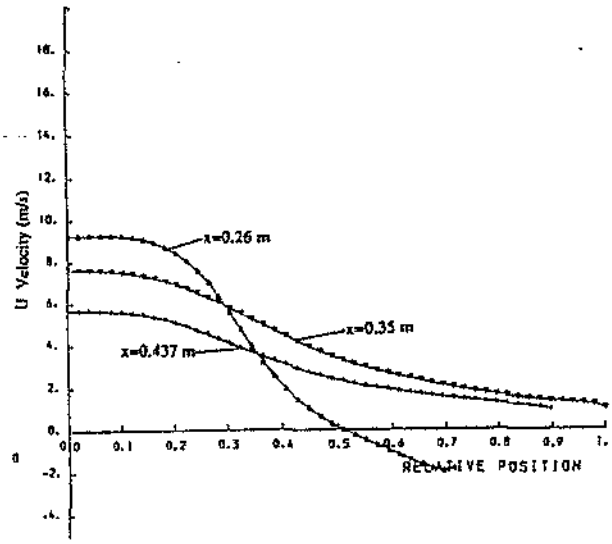


Figure 7 Two-dimensional catalyst can - with catalyst - 45000 GHSV.

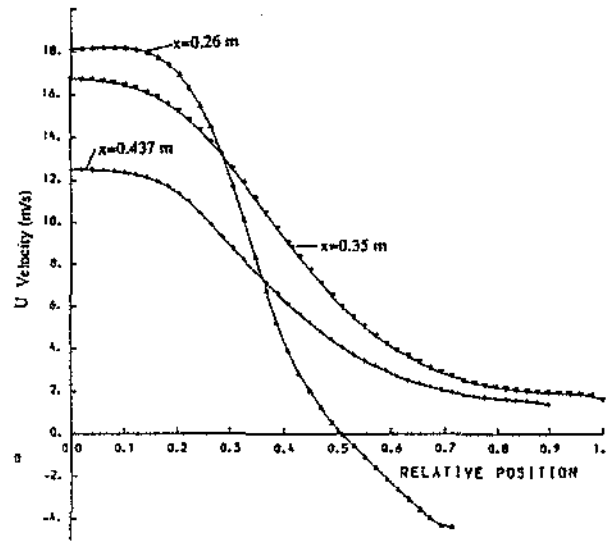


Figure 9 Two-dimensional catalyst can - with catalyst - 90000 GHSV.

Figure 10 Isometric view of a 3.18" x 6.68" x 5.00" 400/6 ceramic substrate (Johnson Matthey).