Modelling the catalyst future

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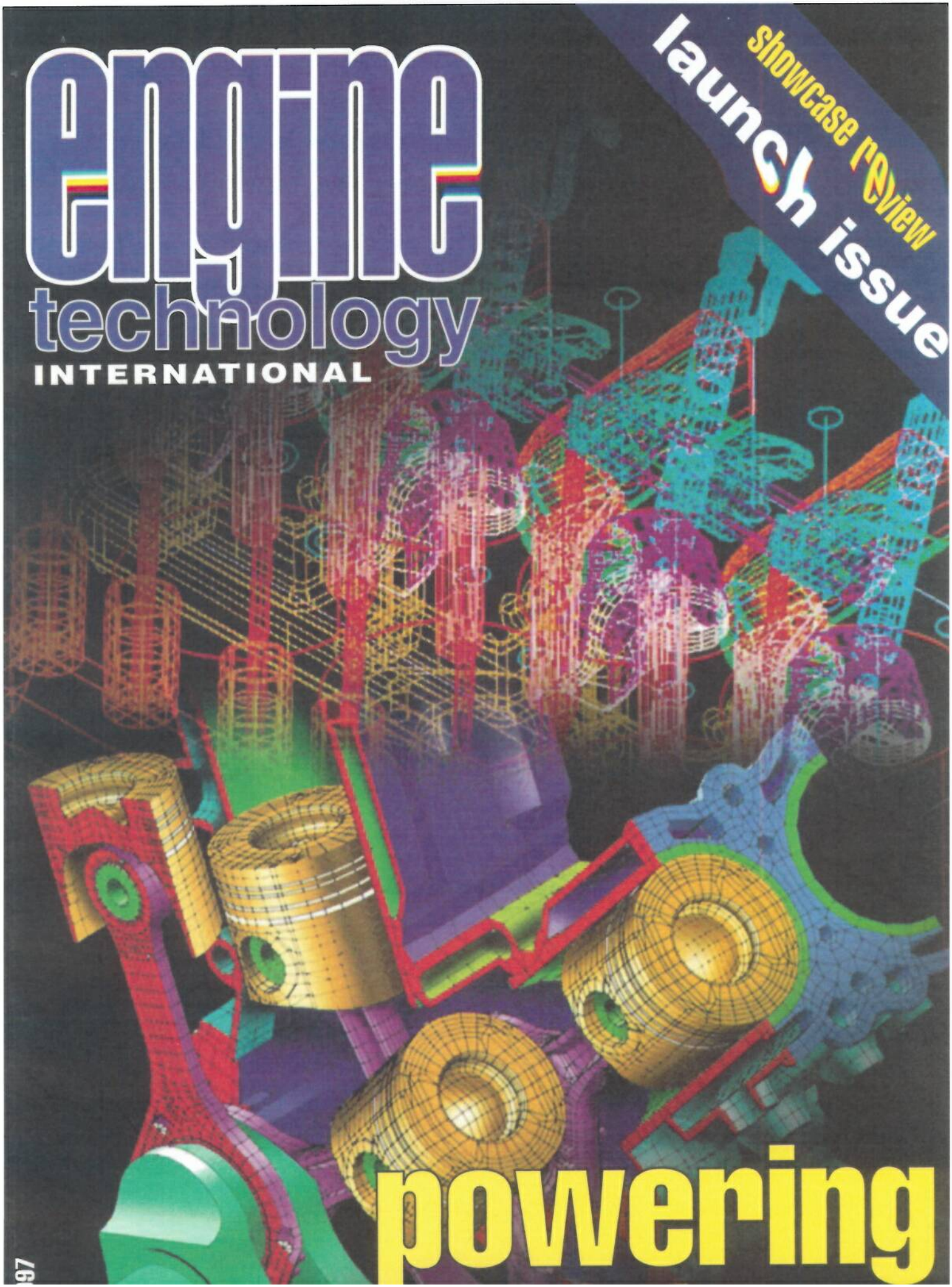

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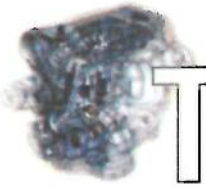
modelling the catalyst future



Three-way catalysts are now used extensively to reduce emissions from spark ignition engines. They operate by oxidising HCs and CO to CO_2 and H_2O while reducing NOx to nitrogen through the catalytic action of noble metals such as platinum, palladium and rhodium. Like many systems, catalysts have a number of performance criteria. The areas of particular interest are their cold start characteristics, steady state emissions, durability and system pressure loss.

Catalysts can only convert significant quantities of emissions if their temperature is raised several hundred degrees above ambient. The time taken for significant conversion, usually following a cold start, is known as the light-off time and it can occur over a very narrow temperature range. It has been estimated that as much as 80 per cent of emissions on an EU driving cycle occur during the light-off period and therefore particular attention is being directed towards shortening light-off times.

Catalyst ageing is also important. Thermal degradation and poisoning (by contaminants in the exhaust gases) reduce their conversion efficiencies. Component longevity is important because catalysts are now required to last in excess of 100,000 miles and performance levels need to be maintained throughout their full life-cycle.

Knowing how a catalyst will perform before anything is actually produced has obvious advantages, so engineers will be pleased to learn that an engineering design tool for simulating light-off is a realistic possibility and should be available soon

Due to the chemical kinetics involved, engines are required to operate within a narrow band either side of the stoichiometric air-fuel ratio. This, coupled with system pressure loss, means that designs are required that can meet emission targets without compromising fuel economy.

In an effort to address these issues considerable research is being undertaken to find ways of improving their design and effectiveness. One way of achieving this would be to develop a simulation tool that will predict emissions and system pressure loss. Many design variants could be simulated prior to prototype manufacture and test. Not only would this hold the prospect of producing better designs, but it would also provide the opportunity of reducing development costs by shortening test programmes. This article describes an approach to develop such a model. First it is useful to describe the main factors affecting the performance of catalytic converters and the physio-chemical processes that need to be simulated if a useful model is to be developed.

factors affecting catalyst performance

The majority of auto catalysts consist of monolith structures coated with an alumina washcoat that supports the noble metals. Monoliths can be either ceramic or metallic and normally comprise numerous parallel channels thereby giving the high surface area upon which reactions take place.

Light-off time is a function of many system parameters. Initially the substrate is cold and is subsequently warmed by the hot gases from the engine. The temperature-time history of these inlet gases is a function of the thermal characteristics of the upstream pipework. On entering the catalyst housing, the exhaust gas expands through a diffuser into the channels of the substrate. The

flow and temperature distribution at the front face of the monolith, which can be highly non-uniform, is a function of the geometry of the diffuser, the resistance of the monolith and the condition of the temperature and flow field (turbulence characteristics, degree of swirl, etc) at the entrance to the diffuser. The flow field is largely determined by the complexity of the upstream pipework. On entering the monolith, the flow laminarises as the monolith channel Reynolds number is quite low. The substrate is warmed through convective heat transfer from the gas to the monolith surface and then through heat conduction through the monolith structure itself. This of course depends on the thermal properties of the substrate (i.e. cell density, monolith heat capacity and conductivity). The gas constituents – HC, CO and NOx – are diffused to the washcoat surface and react in the presence of the precious metal catalysts. Initially the reactions are kinetically controlled as the temperature is low. When the monolith has been sufficiently warmed, reactions take place very quickly and the conversion process is then mass transfer limited. Exothermic reactions ensure the substrate subsequently heats up quickly and heat transfer then occurs from the substrate back to the exhaust gas.

development of a mathematical model

Predicting catalyst performance requires a mathematical description of the flow phenomena, heat and mass transfer processes and chemical reactions as a function of both space and time. It is unlikely that a complete mathematical description will ever be developed to represent all these phenomena with sufficient accuracy that experimentation will ever be avoided. Our understanding of the complex physio-chemical processes, while continuously improving, is still far from complete. However the problem can be made tractable by adopting certain simplifications, i.e. by modelling these complex processes. In this way, through a programme of model development coupled with experimental validation, a useful complementary engineering design tool can be developed for design engineers faced with producing systems to meet ever more stringent emission regulations. This is essentially the task that the Centre for Automotive Engineering at Coventry University, UK, has been engaged in for several years.

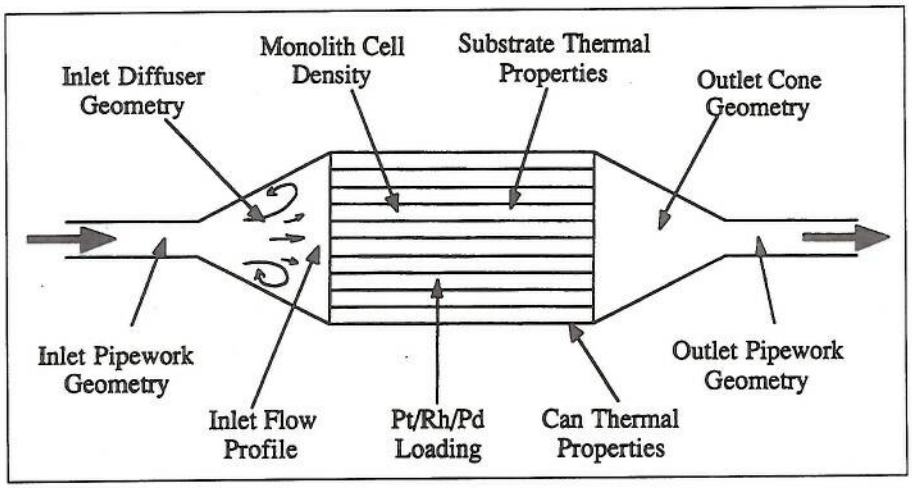


Figure 1: Schematic of converter

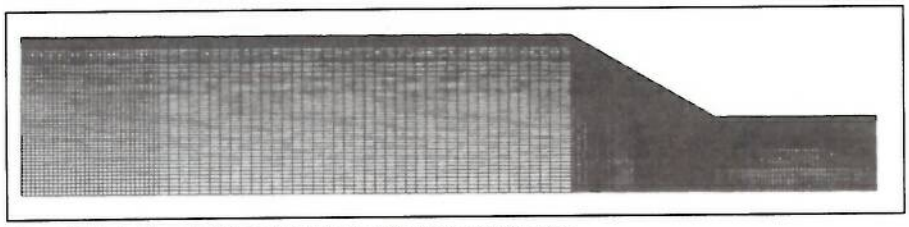


Figure 2: Typical computational grid for an axisymmetric converter

Over the last few years, computational fluid dynamics (CFD) has been widely used in the automotive industry to tackle many engineering problems. The method essentially involves deriving the transport and diffusion equations for the problem variables, i.e. momentum, temperature, species concentration, etc – and solving these numerically at a finite number of points in the flow domain. This is achieved by discretising the equations over a grid of cells superimposed over the flow domain. Solutions are obtained at each cell centre. The greater the number of cells, the more accurate the solution at the expense of increased computing time. Although in principle CFD can be applied to the whole exhaust/catalyst system, there are several issues that need to be addressed.

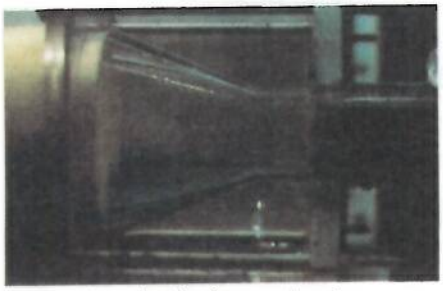


Figure 3: Flow visualisation experiment

In general the problem is 3D, transient and multivariable, and given the current status of computer hardware, it is not feasible to simulate the whole system. Not only must the exhaust pipework be simulated, but also each of the individual channels of the monolith. This would require a prohibitively large number of computational cells.

The second problem relates to describing the complex physio-chemical processes. The flow is extremely complex featuring fully turbulent regimes in the upstream pipes and diffuser and laminarisation within the monolith. The chemical reaction kinetics are also problematic due to the fact that much reaction data is specific to the catalyst itself, which is a complex function of the monolith structure, the nature of the washcoat and the precious metal content – information that is not normally available within the public domain. Given these difficulties, what can be done?

The approach adopted at Coventry is to develop a practical model that can be implemented and used in an engineering design environment, yet at the same time contains sufficient detail to include the important parametric features that are known to affect catalyst performance. This approach requires model development to be closely coupled with an experimental validation programme.

Recognising that the design of the diffuser and the catalyst itself will have a major influence on the overall performance of the system, the approach adopted is to model the system downstream of the diffuser inlet; it being assumed that inlet boundary conditions (inlet flow and temperature profiles) can be measured or calculated elsewhere. One possible method, used successfully in the past, is to simulate the upstream pipework as a one-dimensional system to provide the time-dependent heat flux into the converter.

The flow in the diffuser and converter, however, is highly three-dimensional. Figure 3 shows a flow visualisation experiment that shows the flow structure upstream of the monolith. It can be clearly seen that the flow separates at the throat of the diffuser and jets across the flow domain. The monolith resistance causes the flow to spread out just before entering the monolith channels. The net effect is that the flow and heat flux is maldistributed within the substrate. Maldistributed flow can have several affects on catalyst performance. It can cause premature degradation of the catalysts in areas of high flow, increased pressure loss, and poor utilisation of the catalyst volume. It is, however, possible that the concentrated heat flux at the centre of the monolith can reduce light-off times. Whatever the consequences, it is clear that prediction of the flow distribution within the converter is very important.

As noted previously, the flow problem can be solved in principle by using the solvers of conventional CFD computer codes whereby the flow in the diffuser and monolith are modelled. Resolving the flow domain for each channel of the monolith is, however, impractical. A typical ceramic monolith comprises about 5,000 channels of 1mm^2. Resolving the flow domain for each channel would require, for example, upwards of 1.0E9 computational cells. The approach adopted at Coventry University is to treat the monolith as a porous medium with special properties. In other words, the flow is resolved only at a macroscopic level within the substrate by specifying a distributed resistance downstream of the diffuser. The resistance of the substrate is found from isothermal rig studies conducted under well-controlled conditions. Such an approach circumvents the difficulties of modelling each individual channel. Figure 5 shows an example of measured maldistribution ratios, M, (the ratio of the maximum to mean velocity across the face of the monolith) and the pressure loss as a function of monolith resistance (length), diffuser angle and Reynolds number [1].

Good agreement has been obtained between the predictions and measurements for both parameters in terms of trends and magnitudes. The question now is how can the model be extended to include the effects of heat/mass transfer, chemical kinetics and hence conversion efficiencies?

One approach is to take the isothermal flow field results and use these as input to the monolith, i.e. modelling the monolith separately. This approach, while attractive, neglects the coupling effect between the monolith and the upstream flow. As the monolith temperature increases, so does its resistance and this has the effect of flattening the flow profiles at the front face of the substrate, thereby changing the inlet conditions. An iterative approach would therefore be needed. Clearly it would be better to model the diffuser flow and the substrate as a single integrated system. The problem remains, however, as to how to model all the monolith channels.

One approach is only to model selected channels. This, however, requires specification of the conditions at the chosen channel inlet and difficulties arise where the changing conditions in the substrate influence the upstream flow field for the chosen channel as described earlier. Single channel studies can, however, be useful provided the coupling between adjacent channels is not considered important (i.e. radial heat conduction is secondary) and provided the upstream flow effects are properly accounted for.

The approach that has been developed at Coventry University [2,3] has been to model the flow and heat transfer in the diffuser and catalyst as a fully integrated system while representing the substrate as a continuum. This integrated approach allows for direct coupling between the substrate and the upstream flow and therefore accounts for the increasing resistance of the monolith at elevated temperatures.

Furthermore, modelling the catalyst as a continuum circumvents the difficulties of modelling each individual channel while representing the physics in a realistic manner. Hence the flow and temperature field in the diffuser is simulated by application of conventional

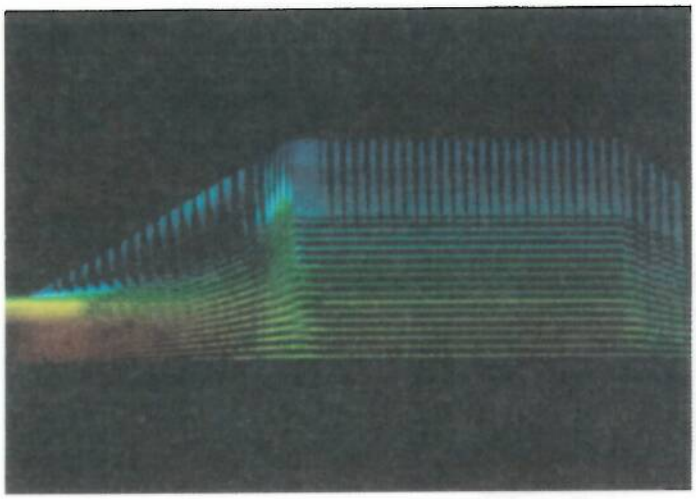


Figure 4: Typical prediction of the flow field in an axisymmetric converter

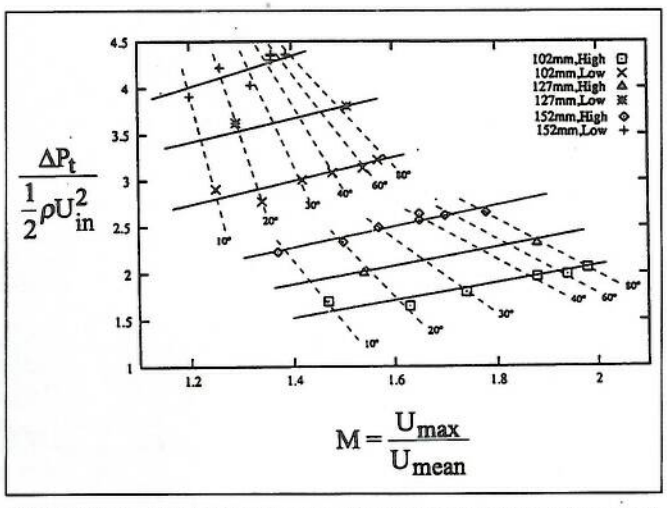


Figure 5: Flow maldistribution and pressure loss in axisymmetric converters [1]

CFD solvers. Downstream of the diffuser the substrate is modelled at a macroscopic level by suitable modification to the CFD code. Essentially the substrate is specified as a porous medium with special properties to account for the particular geometry of the monolith. The link between the gas and solid phases in the substrate is through specification of heat and mass transfer coefficients – these being obtained through a complementary validation programme.

Heat conduction through the solid part of the substrate is modelled by specifying effective conductivities – these being a function of the composite nature of the monolith (e.g. washcoated metallic) and the channel cross-sectional geometry (e.g. square or sinusoidal) – again experimentally determined.

Through model development, coupled with a parallel validation programme, a practical engineering design tool can be developed that represents at the physical level the main influences that control catalyst light-off, i.e. flow maldistribution, substrate properties, etc, while at the same time using experimentally derived data to represent the complex heat and mass transfer processes in the substrate. The following section describes some studies that have been performed using this approach.

simulation studies

Figure 6 shows a simulation of catalyst light-off using the approaches outlined above. The simulation shows the initially cold catalyst being heated by the exhaust gases, which are cooled as they travel through the monolith. After approximately 65-70 seconds the substrate has warmed sufficiently for reactions to take place and the exothermic heat release causes a rapid temperature rise in the substrate and the exhaust gases. CO conversion takes place quickly at this time.

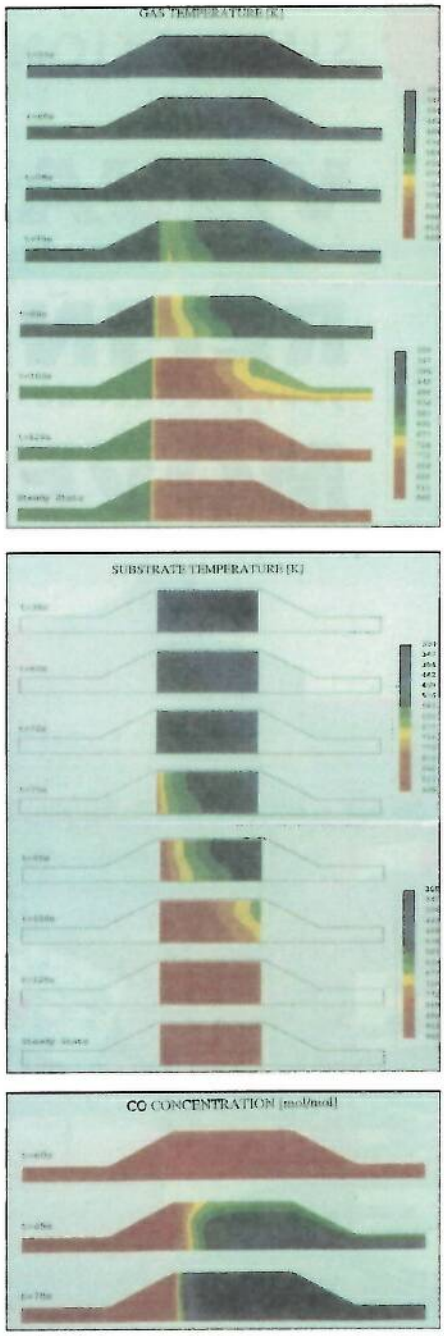


Figure 6: Simulation of catalyst light-off

Figure 7 shows an example of predicted and measured temperature rise in a monolith. What is particularly interesting about this case is that a temperature plateau is observed in the measurements, which was not predicted in the early simulation. This is believed to be associated with the condensation of the exhaust water vapour on the initially cold monolith and its subsequent evaporation. Much better agreement was achieved once the moisture effect was incorporated into the model as seen in Figure 7. The intriguing question is: can this moisture phenomenon affect light-off times?

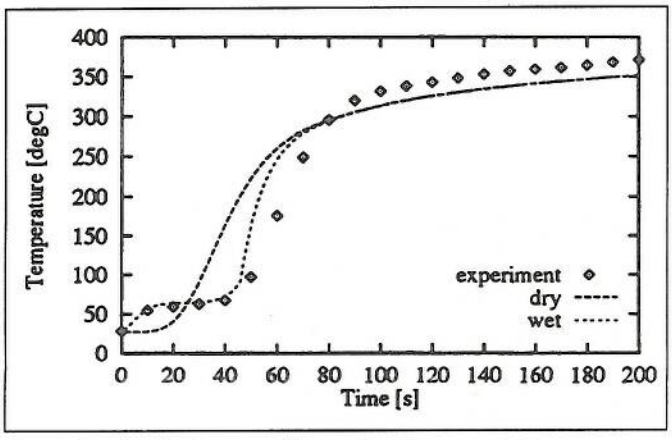


Figure 7: Predicted and measured temperatures in a converter[3]

In conclusion, it would appear that the aim of developing a practical engineering design tool for simulating light-off is realistic. The problem is complex but, coupled with experimental validation programmes, a useful complementary design tool can be generated to help the design engineer faced with the difficult task of optimising catalyst systems.

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