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Modelling catalytic combustion in monolith reactors – challenges faced

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Abstract

When searching for a design concept in which a catalytic combustor is utilised, or looking for areas where improvements can be made to an existing design, then mathematical modelling is an important tool. However, models are only as good as the way in which the physico-chemical processes are modelled and the quality of the physical and chemical parameters (e.g. kinetic expressions, physical properties) acquired for use in the models. When selecting a basis for a model, there are many questions that need to be asked and answered by the developer of the chemical reaction engineering model of the catalytic combustor. Many challenges arise from having to make decisions on compromises that need to be made, and in recognising the consequences of such action. Examples of such challenges are outlined and, for some, clues are offered as to where the answers may lie. The examples include challenges in: the selection of appropriate kinetic expressions, recognition of the role that intraphase diffusion may play, the choice of pressure for catalytic kinetic and pilot scale studies, the selection of heat and mass transfer correlations, and the modelling of transients. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

The general principles of catalytic combustion and developments in this field have been described in a number of review articles (e.g. [1–11,32]), where an introduction to this topic may be obtained.

The catalytic monolith reactor consists of a honeycomb structure with a large number of parallel channels through which the gas flows. The walls of the channels may be:

1. coated with a high surface area washcoat (e.g. γ-alumina) that contains the dispersed catalyst(s), or
2. the wall itself may already contain the catalyst as an integral part of its structure.

The size and the shape of the channels may vary; however, in catalytic combustion applications it is not unusual to encounter square-shaped cells of a size: 1 mm×1 mm. As the channels are of a small size, it is not easy to obtain measurements of concentration or temperature within the channels. This in turn causes experimental difficulties regardless of whether or not they are providing data to support empirical work, or supporting the development of physico-chemical models of the reacting system. If a reliable model of the catalytic combustor was available, then it could provide useful data about conditions within a channel, which in turn would guide development and design.

The modelling of catalytic combustion reactions in a monolith reactor is not an easy task, and some of the challenges faced are described in this paper. First of all the interactions between the various physical and

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chemical processes occurring within the reactor need to be considered. As natural gas is frequently considered to be a suitable fuel in catalytic combustion applications and methane is one of its principle components, in the descriptions that follow, methane has been selected to represent the fuel.

2. Reaction chemistry and mechanisms

In a catalytic combustor, combustion reactions may occur

1. in/on a catalyst layer, and
2. in the gas phase, if conditions are suitable to initiate and sustain homogeneous reactions.

It is important to recognise that both of these can occur simultaneously and that the reaction mechanisms are different although interactions may exist between the two.

The homogeneous gas phase combustion reaction may be represented in simplified form as

$$CH_4 + \tfrac{3}{2}O_2 \rightarrow CO + 2H_2O \tag{1}$$

$$CO + \tfrac{1}{2}O_2 \rightarrow CO_2 \tag{2}$$

with CO, H_2O and CO_2 appearing as products of the combustion reaction. It is well-known that reactions (1) and (2) are a gross simplification of the reaction scheme that may be taking place, e.g. in [12] there are a total of 149 reactions listed. The developer of the model has an important decision to make, should they represent the gas phase reactions in simple or complex terms. Regardless of the scheme selected, it is important to note that when making measurements of species present, CO is one of the key intermediates detected and therefore features in both simplified reaction schemes, e.g. [13], and also in the complex models. On the other hand, the catalytic combustion of methane may be represented by

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \tag{3}$$

with the products CO_2 and H_2O being formed.

Reaction (3) is also clearly a gross over-simplification of what is taking place and fails to account for the mechanistic steps that are taking place. However, as Trimm [14] stated: ‘despite considerable study of the generalities of catalytic combustion, much less is known about the particular aspects of the process’. This situation still applies today. Having worked with a number of catalyst suppliers and/or developers, if they are asked to provide rate expressions to describe the reaction summarised by reaction (3), then one might obtain an expression of the power law form and possibly even a Langmuir–Hinshelwood–Hougen–Watson form, with inhibition terms in the expression. However, neither of these expressions describe the mechanistic steps that may be taking place, although they may be of value in a model, with their use being restricted to a limited range of operating conditions.

Having digressed a little, it is now important to return to the interactions that may be taking place in the catalytic coated channel. As the fuel (CH_4) and air flow down the channel, the reactants, CH_4 and O_2 are transported to the catalyst surface where they diffuse into the porous structure and react on catalytically active sites. The products from the reaction then diffuse through the porous structure and back into the gas phase as it continues to flow down the channel. As the combustion reaction proceeds in the catalyst layer, energy is released and this is accompanied by a rise in temperature. As the temperature of the catalyst layer is higher than that of the bulk gas, energy is transferred by convection and the temperature of the gas increases in the axial direction. Energy exchange also occurs in the walls of the structure in the axial direction by conduction, and between the walls by radiation. As the gas temperature increases down the axial direction, a temperature is reached when gas phase homogeneous reactions become initiated; for CH_4 this could start to become significant at temperatures in the region of 800°C. A point has now been reached where both catalytic and homogeneous reactions are occurring and interactions exist between them. The role that the catalyst is playing may or may not be known – is it enhancing or inhibiting gas phase reactions? As an example of a situation where both have occurred, Pfefferle et al. [15] reported the results of studies into the oxidation of ethane over a platinum catalyst surface. At equivalence ratios less than 0.35, platinum was found to promote gas phase oxidation, whereas at values greater than 0.35 it was shown to have an inhibiting effect. In order to attempt even to start modelling some of these interacting effects, we need rate expressions at the mechanistic level and these are not readily available. Some clues as to how these may develop in the future are available in:

1. Warnatz et al. [16] where the oxidation of hydrogen was studied on a platinum surface. A total of 14 steps describe the gas phase reactions and 11 steps the processes occurring on the catalytic sites.
2. Bond et al. [17] who report the results of modelling the combustion of methane in a catalytic monolith reactor using platinum as the catalyst. They combined four computer codes to model gas phase and surface kinetics. The gas phase chemical mechanism code contained 32 species and 175 reactions, and is known as CHEMKIN and was developed by the Gas Research Institute. The surface mechanisms consisted of 25 steps, based on surface reactions published by Hickman and Schmidt [18], Deutschmann et al. [19]. The computer code known as Surface CHEMKIN was used to model the heterogeneous chemical kinetics.

In any model, the way the catalytic and gas phase reaction chemistry are described is clearly an important factor as it has an impact on the complexity of the equations describing the system, the extent of physical and chemical data that needs to be provided in order to model the system, and the nature of experiments that will need to be conducted in order to validate the model. Herein lie a number of challenges.

3. Intraphase diffusion

In applications of catalysts shaped in the form of pellets, it is generally recognised throughout the process industry that diffusion within the porous structure of the catalyst may start to restrict the rate of reaction. To describe this limitation, a term known as the effectiveness factor, η, may be used, defined as

$$\eta = \frac{[\text{measured reaction rate}]}{[\text{rate of reaction in the absence of diffusional resistances}]} \tag{4}$$

For an isothermal pellet, the denominator would be evaluated using the intrinsic rate expression in combination with known values of concentration and temperature on the surface of the catalyst pellet. If η has a value close to one, then intraphase diffusion is not significant, whereas a value of $\eta<1$, e.g. 0.4, would indicate that catalyst utilisation was only 40% efficient.

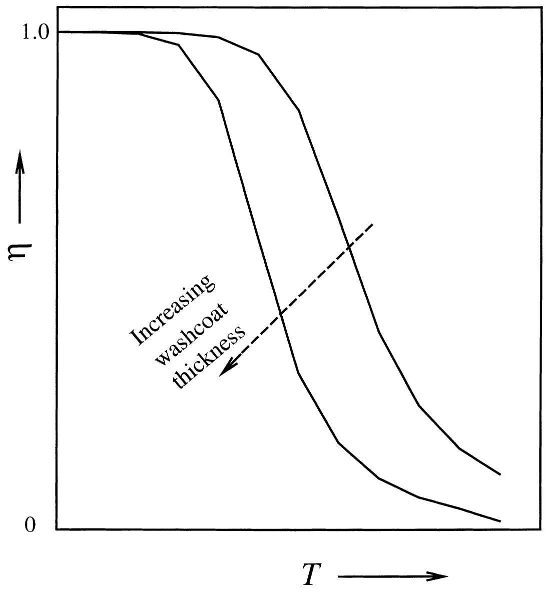

Fig. 1. Sketch illustrating variations in the effectiveness factor with temperature for varying thicknesses of washcoat, for a constant surface mole fraction of methane and constant pressure (methane is assumed to represent the fuel).

In catalytic monoliths, as the thickness of the washcoat layer is very thin, e.g. 50 μm [20] and 10–15 μm [21], it is not unusual to find in modelling studies that this aspect has been neglected. However, as shown in a number of recent studies (e.g. [21–23]), diffusion limitations may become significant (e.g. η=0.8–0.4) at temperatures as low as 700–800 K, depending on the type of fuel and catalyst/washcoat properties. For the catalyst system being studied, it is important to identify the shape of the effectiveness factor plot. An interesting method of plotting these data is illustrated in Fig. 1, where variations of η with the surface temperature of the catalyst may be seen clearly and also the effect of the washcoat thickness. Failure to recognise the significance of diffusion limitations has in turn led to much confusion about the magnitude of *Sh* and *Nu* numbers to be assigned in fully developed

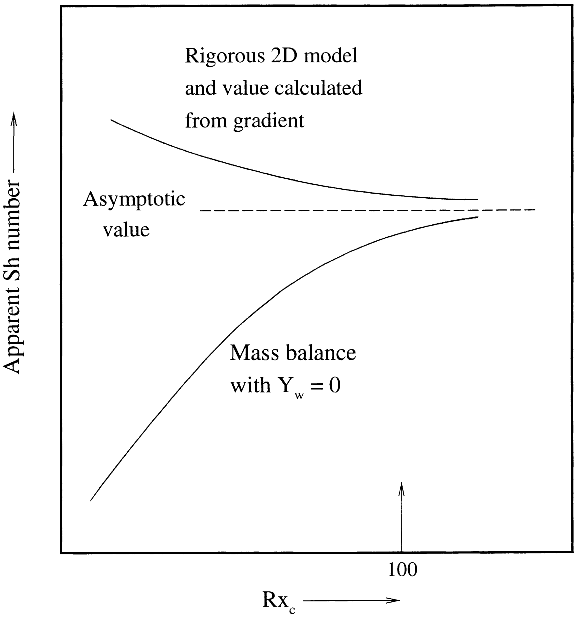

Fig. 2. Sketch illustrating how the computed values of fully developed Sh number vary depending on the method of calculation, based on Hayes and Kolaczkowski [21].

laminar flow. This has arisen from the method used to back-calculate mass transfer coefficients from reaction experiments assuming that intraphase diffusion was not limiting the reaction – calculated values of reaction rate were therefore assumed to be so high at the surface that the reaction was assumed to be completely mass transfer limited and a zero concentration was incorrectly assigned at the surface. This effect was illustrated by Hayes and Kolaczkowski [21] in a circular shaped channel, by defining for a first order reaction, a catalytic reaction number Rx_c and then showing how the calculated values of the Sherwood number varied depending on the method of calculation; this is shown in Fig. 2. The number evolved from one of the boundary conditions at the wall, where

$$-DC_T\frac{\partial Y}{\partial r} = R_w \text{ at } r = R \text{ for all } z \tag{5}$$

and for this first order reaction, the following form of expression was assumed to apply:

$$R_w = C_T k_w Y_w. \tag{6}$$

Substituting reaction (6) into (5), and rewriting and rearranging in dimensionless form

$$-\frac{\partial Y}{\partial r^*} = \frac{k_w D_T Y_w}{D} = Rx_c Y_w, \tag{7}$$

where $Rx_c = k_w D_T / D$. From Fig. 2, it is clear that if a zero concentration is assumed at the wall, then for low values of Rx_c, very low values of Sherwood number are calculated. However, if the mole balance is solved in a rigorous manner (then as values of Y_w are actually calculated), the Sh number may be determined from

$$Sh = \frac{2R}{(Y_b - Y_w)}\left(\frac{\partial Y}{\partial r}\right)_w. \tag{8}$$

These values now lie close to those expected for fully developed flow in circular channels, see Fig. 2.

At high values of Rx_c, i.e. greater than about 100, then the two methods give similar results. This is not surprising as the mole fraction at the wall Y_w, was less than 3% of the value in the bulk – so assuming it to be zero would not result in a significant difference in calculated values of Sh number.

Not recognising that diffusion limitations exist in the catalyst layer may in turn lead to false conclusions being drawn about the activity of the catalyst. It is easy to think that a more active catalyst is required when in fact a less active one may suffice and higher rates of reaction may be achieved by decreasing the thickness of the catalyst layer in combination with an increase in the geometric surface area per unit volume of the monolith.

The recognition of whether or not diffusion limitations in the catalyst layer exist, and then how to proceed with their incorporation into a reactor model is another challenging task. Especially as one may be dealing with a square shaped cell, where the washcoat is deposited in a non-uniform manner around the perimeter of a cell, e.g. see [21]. To complicate matters further, the extent of catalyst impregnation (or its dispersion) within the washcoat layer may also vary.

4. Performance of low pressure experiments for high pressure applications

Commercial applications of catalytic combustion in stationary gas turbines may necessitate pressures as high as 10–20 bar and turbine inlet temperatures as high as 1500 K. At these experimental conditions, the provision of experimental facilities to perform catalyst screening trials, catalyst kinetic studies and pilot scale trials to evaluate the performances of catalysts and validate models would clearly be an expensive and also a time-consuming task. There is a big difference between the size and weight of a reactor assembly that only needs to operate at 1 bar versus 20 bar pressure, and hence the time and support facilities required even to do a simple change of catalyst. It is therefore not surprising that researchers and their sponsors have tended to work at lower pressures. In the literature there are some studies at elevated pressures. However, these have usually been done by commercial organisations and not all of the information is available on these trials as they have obvious commercial constraints.

Working at lower pressures has the advantage of relative simplicity in experimental rig design. However, there are a number of disadvantages which are not always fully appreciated by the investigators. As the role of scale-up and data extrapolation should be performed by modellers skilled in the reaction engineering aspects of catalytic combustion, some of the traps to be avoided are identified as follows and once again challenges are posed.

4.1. Low pressure catalyst screening trials

Whilst these may be extremely useful to evaluate the performance of a range of catalysts, great care has to be taken to ensure that the adiabatic temperature rise in the catalyst bed could not exceed a critical value at which the catalyst may become damaged and hence lose its activity. Catalytic combustion reactions are very fast and highly exothermic – because of the difficulty of measuring the actual temperature of the catalyst it is easy to become complacent about this aspect. Catalyst screening trials are also frequently performed on powders. The relative performance of a coated or formed monolith may be quite different.

4.2. Low pressure kinetic studies

In order to utilise catalytic rate experiments in reactor models, it is necessary to evaluate experimentally the rate expressions. These expressions are very much both catalyst and system specific and it is not yet possible to determine them a priori from information in the literature. If kinetic studies are performed at low pressures then the effect of pressure is clearly unknown and the term could therefore not feature in the rate expression. Another problem that is very likely to arise results from the difficulty of measuring catalyst surface temperatures at which the reaction may be occurring, and also ensuring that the rate is not limited by intraphase or interphase diffusion effects. A decision may therefore be taken to perform the kinetic experiments at lower temperatures than those likely to be encountered in the commercial application – how valid are these expressions when in turn they are extrapolated to the high temperature region? If kinetic studies are performed on powdered catalysts then how do you adapt the expression to represent conditions in a coated or catalyst formed monolith? In order to overcome some of the difficulties associated with measurements of kinetics at high temperatures, McCarty [24] describes the use of an annular reactor where a thin coating of catalyst (10–40 μm) is deposited on the surface of a small tube, and the reactants flow in an annular gap (0.1–0.3 mm) at a linear velocity of 2–10 m/s. Using this configuration and flow conditions, it was claimed that thermal and concentration gradients across the gas film boundary layer had been minimised. The combustion of methane was studied over a palladium catalyst supported on $La_2O_3{\cdot}11Al_2O_3$, at temperatures in the range of 300–800°C.

4.3. Low pressure pilot scale trials

In this section, the term ‘pilot scale trials’ is used to represent experiments that are performed in a tubular reactor with the aim of obtaining information on performance to aid system scale-up. In these types of experiments the monoliths are usually of a length to achieve conversion levels between 30% and 100% in the reactor. Whereas in the kinetic experiments described in Section 4.2, conversion would normally be restricted to a level of 5–10%, although depending on the method of analysis, higher values may also be encountered.

If low pressure pilot scale trials are performed on a coated or formed monolith, then at low pressures the flow in the channels is likely to be in the laminar flow regime and interphase mass and heat transfer may play a significant role in these experiments. In a theoretical study [22], it was shown how easy it is to over-temperature the catalyst and hence draw false conclusions about the activity of the catalyst. Had the experiments been performed at elevated pressures, then because of the higher values of the heat transfer coefficient, the wall temperatures would have been substantially lower and hence catalyst deactivation would not have actually occurred in practice.

5. Modelling heat and mass transfer in monolith channels

Depending on the Reynolds number, flow in the channels may be in the

- laminar region: $Re<2\,100$,
- turbulent region; $Re>10\,000$, or
- transition region: $2100<Re<10\,000$.

An early assessment of the flow region is important as it provides an indication of the challenges to be faced. If conditions pertain to the transition region, then one may choose to follow the words of caution offered in [25]: “The transition region extending roughly from $Re=2100$ to 10 000 is not well understood and is usually avoided in design if possible”. In the following sections, some of the challenges of working in the laminar and turbulent flow region are discussed.

In considering flow in a monolith channel, it should be appreciated right from the start that in a reacting catalytic monolith channel, where the gas temperature and velocity increase in the axial direction, and the shape of the cell is rarely circular, it is very unlikely that fully developed flow would ever be achieved in a practical application. If it did, then reactions would have stopped and that section of the reactor would not be contributing. Instead, the flow is more likely to be in the developing region and in some applications this feature may be intentionally exploited in the design.

5.1. Laminar region

If a one-dimensional flow model is employed, then heat and mass transfer correlations need to be employed that reflect flow conditions in the channel. In Section 3, the danger of being drawn into correlations with substantially lower values than expected for fully developed flow has already been discussed. However, an area where care still needs to be taken is in the way developing flow is modelled in a channel. If a one-dimensional correlation is used, then axial distance in the channel features in the expression. For example, in Hawthorn [26], for a circular channel, then

$$Sh = 3.66\left[1 + 0.095\frac{D_\mathrm{h}}{L}Pe_\mathrm{m}\right]^{0.45}, \tag{9}$$

$$Nu = 3.66\left[1 + 0.095\frac{D_\mathrm{h}}{L}Pe_\mathrm{h}\right]^{0.45}. \tag{10}$$

These correlations have frequently been used in the literature. However, because of the way they were derived, for a fixed length L they represent the average value over that length and not the local value at $x=L$. This can have a major effect on the way the performance of the reactor is modelled and calculated values of catalyst/wall temperatures. In order to determine local values of heat and mass transfer coefficients, the paper by Groppi et al. [27] should be studied. In that paper, correlations were refined for a square channel, presenting the following for the constant wall temperature and constant heat flux case:

$$Nu_\mathrm{T} = 2.97 + 6.854\left[\frac{1000}{Gz}\right]^{-0.5174}\exp\left[-\frac{42.49}{Gz}\right] \tag{11}$$

and

$$Nu_\mathrm{H} = 3.095 + 8.933\left[\frac{1000}{Gz}\right]^{-0.5386}\exp\left[-\frac{6.7275}{Gz}\right]. \tag{12}$$

Having mentioned these two conditions, it is worth reflecting on which one may apply, these issues are discussed in [27,28]. To illustrate the extent of variation in simulated experimental data, the results of using two different methods to model a monolith reactor are illustrated in Fig. 3.

If a two-dimensional (2D) model is used, then the rigorous equations may be written which include appropriate boundary conditions to model the developing profile. The complexity of the numerical techniques to be employed is in orders of magnitude more complex than the 1D code. But if one is looking for a

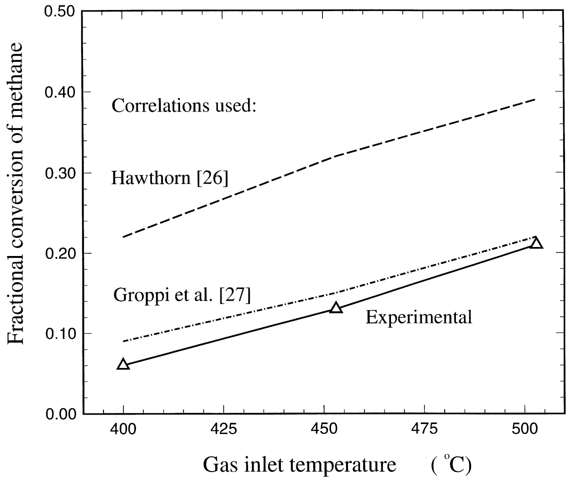

Fig. 3. Catalytic combustion of methane in a monolith reactor: fractional conversion as a function of gas inlet temperature. Inlet mole fraction of methane=0.011; atmospheric pressure; monolith length=0.015 m; channel size=1.2×1.2 mm, square shaped cells with 62 cells/cm^2 (supplied courtesy University of Bath, data acquired in EC Brite Euram Project, see acknowledgements).

rigorous approach then is the 2D approach valid? Monolith channel are generally square, square with rounded corners, triangular or sinusoidal – it is very rare to encounter a circular channel. A three-dimensional (3D) code should therefore be used – the complexity of modelling the reactor would again increase by another order of magnitude and it is not a challenge that has tempted many to follow. A careful balance therefore needs to be struck between the development of a model that can be used readily as a tool to aid reactor design, and the desire to model in a rigorous manner all of the phenomena that may be taking place. For the examples of recent developments and studies of 2D codes, papers by Leung et al., Groppi et al., Hayes and Kolaczkowski, and Hayes et al. [23,27–30] should be consulted.

5.2. *Turbulent region*

For 1D flow models, correlations for fully developed flow are readily available. However, relatively little is known about the effect that the developing profile may have on rates of heat and mass transfer under catalytic combustion conditions in a monolith reactor. For example, if a gas at an elevated pressure (e.g. 20 bar) enters a square shaped cell (1 mm× 1 mm) with a linear velocity of 52 m/s and $Re=1.3\times10^4$, then if the difficulties of attaining a stable flow pattern in a square shaped cell are combined with small variations in surface roughness and of catalyst coating around the perimeter of the cell, and of reactions taking place at the wall leading to thermal expansion, then it is clear that these factors will have an important effect.

If heat transfer rates are enhanced substantially in the entrance to the monolith, then the wall temperature, and reactions will proceed at a slower rate than that predicted with the aid of a model that uses a fully developed flow correlation.

6. Modelling transients

6.1. *Thermocouple lag*

Modelling the transient behaviour of a catalytic monolith is not an easy task. In the literature, there are very few papers that combine model development and simulations with actual experimental results to validate such models. A recent example of such a study is presented in [30]. Transient experiments were performed during periods of heating and cooling. This was achieved by first allowing a steady state to be achieved (either with or without methane in the feed stream), and then the methane was turned either on or off simulating an inlet step change in concentration. Monolith wall temperatures were measured with two thermocouples that were pushed down into two separate channels, and located at a distance of 10 and 120 mm from the inlet (the total length of the monolith was 151 mm). The thermocouple was a tight fit in the channel, ensuring good contact with the wall. One of the interesting aspects described in this paper is the recognition and method of quantification of the lag that arose as a result of the thermal capacity of the thermocouple itself. The location of the thermocouple in the monolith is illustrated in Fig. 4. The thermocouple blocks a channel so reactions do not proceed in that channel. Therefore the walls that surround the thermocouple and the thermocouple itself will respond to a change more slowly than the rest of the monolith. This behaviour was quantified by increasing the value of the effective wall thickness of the channel in the model, and a good match was

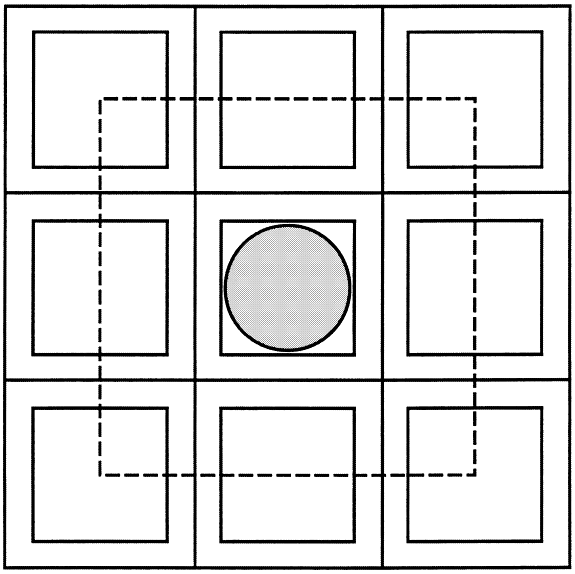

Fig. 4. Sketch illustrating the location of a thermocouple in a monolith, adapted from Hayes et al. [30].

obtained between the experimental and modelled data. When looking for a match between experimental and simulated data, it is important to compare wall temperatures in the monolith as well as gas outlet temperature and composition data; this aspect has been recently emphasised in [31]. The method of monolith wall temperature measurement and allowances for thermal (or instrument) lag are clearly both challenging and important factors.

6.2. *Transient kinetics*

In some applications, it is not necessary to have catalytic rate expressions with transient terms, however, there are situations where this is very important. For example, in the modelling of a catalytic convertor to control emissions from an engine where:

1. carbon monoxide and the hydrocarbons are oxidised,
2. the NO_x is reduced, and
3. conditions in the exhaust may vary from fuel-rich to fuel-lean with the cerium oxide in the catalyst layer acting as an oxygen store,

then it is easy to see that transient terms are necessary. The modelling of a catalytic converter, when every inlet parameter is continuously changing is clearly a challenging task and also an area where steady state models are of little value.

In the application of catalytic combustion as a primary combustion process (e.g. as an application in a stationary gas turbines), then inlet conditions to a catalytic monolith are more likely to be of a steady state nature. Obviously if the power from the unit needs to be varied, then inlet conditions will change until a new steady state is reached. However, if as a result of operating conditions, the characteristics of the catalyst change, and these in turn affect the rate of reaction, then transient terms need to be included. One example, is the well recognised transition of the catalyst palladium, from the oxide to the metal:

$$PdO \rightleftharpoons Pd + \tfrac{1}{2}O_2.$$

For example, if the temperature of a PdO catalyst in an air stream at atmospheric pressure is increased, then at about 800°C the catalyst will start to decompose to Pd. The change is gradual, and as a result of it, the activity of the catalyst decreases. If surface temperatures of the catalyst system are maintained below 800°C, then a steady state model may suffice. However, if it is expected to operate the catalytic combustor at temperatures in the transition range, then transient terms to describe the change in catalyst and how the reaction rate expression(s) need to be modified would need to be included.

Another difficult area that faces experimentalists, is one where they notice a change in activity with time. The change may be very noticeable for a fresh catalyst – especially, if it has not been conditioned or aged. For a catalyst in a long service application, then small changes as a result of e.g. sintering, poisoning, may occur. The experimentalist or design engineer often ask if these can be modelled, as they have an important effect on design and the allowances that subsequently need to be made, e.g. how much longer should the monolith be and what effect could this have if the reactor is operated with fresh catalyst?

In reality, if the activities on the surface of the catalyst and interaction with promoters, catalyst support and other contaminants were examined in detail, then it is evident that the situation is very dynamic, and if a rigorous approach was to be followed then these changes would require transient terms. The challenge facing the modeller is to recognise when transient terms are important in kinetic expressions, and then how to formulate and validate the model.

7. Concluding remarks

There are many challenges that face the developer of a mathematical model of a catalytic monolith combustor. In this paper, examples of challenges have been selected, that reflect areas that have not been addressed adequately by researchers over the last five years. For some of these, solutions exist, whilst others will require long term research.

From the description of some of the challenges faced, it should be clear that the developers of mathematical models of catalytic combustors cannot work in isolation either from the pilot scale experimentalists or researchers into new catalyst systems. Despite the difficult challenges faced, modelling has a very important part to play, in the identification of conditions to be tested, scale-up and system design. If modelling proceeds in parallel with experimental studies and concept development/design, and it is applied correctly, then it should result in lower costs in the development phase of a project.

Finally, it is important to emphasise the importance of building up models in a gradual manner, guidance on this aspect has been gathered in the first introductory text book on catalytic combustion by Hayes and Kolaczkowski [32]. Even from a relatively simple model there is a lot that could be learned. Simpler forms of models also offer the advantage of short computer runs and less problems when the cause of a program crash or strange solution needs to be identified. However, the model should represent the physical and chemical processes taking place in the catalytic combustor, and therefore the formulator of the model would need to be skilled in chemical reaction engineering and associated fluid mechanics and heat transfer – as resources and the time available for a research project are limited, compromises in the development of a model will need to be made.

8. Nomenclature

C_T	total molar concentration (mol/m^3)
D	molecular diffusion coefficient (m^2/s)
D_h	hydraulic diameter of channel (m)
D_T	diameter of the channel (m)
Gz	Graetz number
h_d	mass transfer coefficient (m/s)
ΔH	heat of reaction (J/mol of methane)
k_w	catalytic rate constant (m/s)
L	length of monolith (m)
Nu_T, Nu_H	Nusselt number, constant wall temperature and constant heat flux case
Pe_m	Peclet number for mass transfer
Pe_h	Peclet number for heat transfer
$^*r$	dimensionless radial distance
r	radial coordinate (m)
R	radius of the channel (m)
Re	Reynolds number
R_w	rate of catalytic reaction, e.g. combustion of methane (mol/m^2 s)
Rx_c	dimensionless catalytic reaction number for a first order reaction
Sh	Sherwood number
Y	mole fraction of fuel, e.g. methane
Y_b	mole fraction of fuel in the bulk stream, or average value
Y_w	mole fraction of fuel at the wall
z	axial coordinate (m)

Greek letters

η	effectiveness factor of the catalyst

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