Numerical investigation of wall heat conduction effects on catalytic combustion in split and continuous monolith tubes

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Abstract

The optimum length of a monolith tube is one for which near-hundred percent conversion is attained, and at the same time, the catalyst over the entire length of the tube is utilized. In practice, the length is adjusted by stacking monolith plugs end-to-end. In this study, the repercussions of such a practice are investigated numerically with the goal to determine if a tube of length $2L$ demonstrates the same behavior as two tubes of length $L$ each, stacked end-to-end. Catalytic combustion of methane–air mixture on a platinum catalyst is considered. The studies are conducted using a multi-step reaction mechanism involving 24 surface reactions between 19 species. Two different materials are considered for the walls of the monolith tube, namely silicon carbide and cordierite. Both steady state and transient simulations are performed. Results indicate that the ignition and blowout limits can be significantly different between split and continuous tubes when the wall is made up of a high thermal conductivity material, such as silicon carbide. For steady state combustion, for both wall materials, the point of attachment of the flame to the wall is altered by splitting the tube—the effect being more pronounced for silicon carbide and at relatively high Reynolds numbers. These results imply that axial heat conduction, or lack thereof due to thermal contact resistance, through the walls of the monolith results in thermal non-equilibrium between the solid and fluid phase, and subsequently affects ignition and flame stability in catalytic combustion.

Keywords: CFD modeling; Catalytic combustion; Catalyst support; Heat conduction; Thermal non-equilibrium

1. Introduction

Catalytic converters are employed in many engineering applications, such as automotive exhaust after-treatment, environmental detoxification of exhaust gases from chemical plants, low temperature combustion for reduced emissions in gas turbine applications, among numerous others. With an increasing trend towards miniaturization and growing interest in alternative energy conversion technologies, catalytic conversion is expected to play an even bigger role in the future. A monolithic ceramic plug, coated with a catalyst, typically serves as the surface on which heterogeneous chemical reactions occur, enabling conversion. Numerical simulations have found prolific usage within the industry and research communities for better understanding of the operation of catalytic converters.

One of the critical parameters in the design of catalytic converters is the length of the monolith plug to be used. Ideally, the length should be such that no catalyst is wasted and at the same time, the conversion percentage is close to 100%. In practice, the length is adjusted to near-optimum by stacking together several monolith plugs end-to-end. Typically, plugs of 1 in. length are used as building blocks. One critical question that arises out of this practice is whether a monolith tube of length $2L$ will exhibit the exact same behavior as two tubes of length $L$ stacked end-to-end (Fig. 1). Recent experimental observations for lean NO$_x$ trap (LNT) converters (Choi, Prikhodko, Chakravarthy, & Daw, 2006) appear to indicate that the behavior seen in the two scenarios, just described, may not always be the same. Such experimental data are very limited and require corroboration through further carefully controlled experimental studies. Furthermore, because of the complex nature of operation of LNT converters, it is not clear if any discrepancies are due to the splitting of the monolith tube or some other unforeseen experimental errors. Nevertheless, from a theoretical standpoint, it is credible that a difference may exist. One of the possible reasons for differences in the behavior between continuous and split monoliths...
The objective of the present study is to elucidate behavioral differences in ignition and steady state combustion characteristics in split and continuous monolith tubes through computational fluid dynamic (CFD) simulations. While evidence of CFD modeling of single monolith tubes is abundant in the literature (Deutschmann et al., 1994; Raja et al., 2000), and also because catalytic combustion of hydrocarbons constitutes an important class of applications. To enable study of wall heat conduction effects, two different commonly used ceramic materials are chosen: silicon carbide, which is a high thermal conductivity material, and cordierite, which is a low thermal conductivity material. Both steady state and transient simulations are performed for both wall materials for various flow rates (Reynolds number) and fuel–air mixtures (equivalence ratios). The simulations are performed using the commercial CFD code CFD-ACE+TM, and include fluid flow, heat transfer by all three modes, multi-component species transport and multi-step finite-rate heterogeneous chemistry. Predicted results for species mass fraction distributions, temperature distributions and conversion percentages are carefully analyzed to gain insight into the ignition, blowout and steady state combustion phenomena in such systems, with particular emphasis on the differences between split and continuous tubes.

2. Governing equations

The governing equations are the equations of conservation of mass (both overall and individual species), momentum and
energy, and are written as (Bird, Stewart, & Lightfoot, 2001; Kuo, 1986):

overall mass: \[ \frac{\partial}{\partial t} (\rho) + \nabla \cdot (\rho \mathbf{U}) = 0 \]  

momentum: \[ \frac{\partial}{\partial t} (\rho \mathbf{U}) + \nabla \cdot (\rho \mathbf{U} \mathbf{U}) = -\nabla p + \nabla \cdot \mathbf{\tau} + \rho \mathbf{B} \]  

energy: \[ \frac{\partial}{\partial t} (\rho h) + \nabla \cdot (\rho h \mathbf{U}) = -\nabla \cdot \mathbf{q} + \dot{S}_h \]  

species mass: \[ \frac{\partial}{\partial t} (\rho Y_k) + \nabla \cdot (\rho U Y_k) = -\nabla \cdot \mathbf{J}_k + \dot{S}_k \quad \forall k = 1, 2, \ldots, N \]  

where \( \rho \) is the mixture density, \( p \) the pressure, \( \mathbf{\tau} \) the shear stress tensor, and \( \mathbf{B} \) the body force vector. Eqs. (1) and (2) are the well known Navier-Stokes equations, and need no further discussion. In Eq. (4), \( Y_k \) is the mass fraction of the k-th species, \( \mathbf{J}_k \) the mass diffusion flux of the k-th species and \( \dot{S}_k \) is the production rate of the k-th species due to homogeneous chemical reactions. The total number of gas-phase species in the system is denoted by \( N \). In Eq. (3), \( \dot{S}_h \) represents the net source due to viscous dissipation and other work and heat interactions, and \( \mathbf{q} \) denotes the net heat flux due to molecular conduction, radiation and inter-species diffusion, and is written as (Bird et al., 2001):

\[ \mathbf{q} = \mathbf{q}_C + \mathbf{q}_R + \mathbf{q}_D = -k_c \nabla T + \mathbf{q}_R + \sum_{k=1}^{N} \mathbf{J}_k h_k \]  

where \( h_k \) is the enthalpy of the k-th species, \( k_c \) the thermal conductivity of the mixture and \( h \) the enthalpy of the mixture (= \( \sum_{k=1}^{N} h_k Y_k \)). The heat flux due to inter-diffusion of species, \( \mathbf{q}_D \), can be comparable or larger than the Fourier conduction flux \( \mathbf{q}_C \), and can result in net heat flux that is opposite in direction to the imposed temperature gradient (Kumar & Mazumder, 2007a). In the above formulation, the enthalpy of the k-th species, \( h_k \), includes the enthalpy of formation and the sensible enthalpy, and is written as

\[ h_k(T) = h_{f,k}^0 + \int_{T_0}^{T} c_{p,k}(T) \, dT \]  

where \( h_{f,k}^0 \) is the enthalpy of formation of species \( k \) at the standard state, and \( c_{p,k} \) is the specific heat capacity of species \( k \). The species enthalpy is generally computed using standard thermodynamic databases, such as the JANNAF database.

The diffusion flux in a multi-component system is often modeled using the so-called dilute approximation (Bird et al., 2001; Desilets, Proulx, & Soucy, 1997; Sutton & Gnofo, 1998; Wangard, Dandy, & Miller, 2001). In a recent study (Kumar & Mazumder, 2007b), it has been shown that for catalytic combustion applications, the dilute approximation is comparable in accuracy to a rigorous multi-component diffusion formulation derived from the Stefan-Maxwell equations, while being computationally about twice as efficient. Thus, the dilute approximation is used here. Using the dilute approximation, the diffusion flux is written as (Bird et al., 2001; Desilets et al., 1997; Sutton & Gnofo, 1998; Wangard et al., 2001)

\[ \mathbf{J}_k = -\rho \mathbf{D}_{km} \nabla Y_k \]  

where \( \mathbf{D}_{km} \) is the effective diffusivity of species \( k \) into the mixture, and is henceforth denoted by \( \mathbf{D}_k \) for simplicity. The effective diffusivity is given by the relation (Bird et al., 2001)

\[ \mathbf{D}_k = \frac{1}{\sum_{i=1}^{N} \frac{X_i}{D_{ki}}} \quad i \neq k \]  

Substitution of Eq. (7) into Eq. (4) yields the appropriate species transport equation for the dilute approximation formulation:

\[ \frac{\partial}{\partial t} (\rho Y_k) + \nabla \cdot (\rho U Y_k) = \nabla \cdot (\rho D_k \nabla Y_k) + \dot{S}_k \quad \forall k = 1, 2, \ldots, N \]  

Radiation is the dominant mode of heat transfer in such high-temperature applications. In a separate related study (Mazumder & Grimm, 2007), detailed participating medium radiation calculations using Planck mean absorption coefficients of the gas mixture were performed, and it was found that the combustion gas within the monolith tube hardly affects the radiation field, since these tubes are narrow (2 mm diameter). The optical thickness of the gas channels is, therefore, very small, and the gas can be considered non-participating. Thus, for the present study, only radiation exchange between surfaces was considered. Such an approximation has also been used by past researchers (Boehman, 1998) for modeling radiative transport in monolith tubes. In the present study, the discrete ordinates method with the S4 approximation was used to solve the radiative transport equation. The relevant discrete ordinates equations are readily available from the text by Modest (2003), and are omitted here for the sake of brevity.

The governing equations, just described, require boundary conditions. The boundary conditions for the mass and momentum conservation equations are the no-slip conditions at walls, and appropriate mass flux or pressure boundary conditions at inflow and outflow boundaries. These boundary conditions and their numerical implementation are well known and need no further discussion. The focus of this discussion is the boundary conditions for species and energy associated with heterogeneous chemical reactions at fluid–solid interfaces.

At a reacting surface, the diffusion flux of species is balanced by the reaction flux since the surface cannot store any mass. At the heart of surface reaction processes is adsorption and desorption of species at the surface, the treatment of which requires inclusion of so-called surface-adsorbed species (Coltrin, Kee, & Rupley, 1991). At steady state, the net production rate of the surface-adsorbed species is zero. In the absence of etching or deposition of material from the surface (i.e., zero Stefan flux), the reaction-diffusion balance equation at the surface may be written as (Coltrin et al., 1991; Mazumder & Lowry, 2001)

\[ \mathbf{J}_k \circ \mathbf{n} = M_k R_k \quad \forall k \in \text{gas-phase species} \]  

(10a)
\[ R_k = \begin{cases} \frac{dA_k}{dt} & \text{for unsteady} \\ 0 & \text{for steady} \end{cases} \quad \forall k \in \text{surface-adsorbed species} \] (10b)

where \( R_k \) is the molar production rate of species \( k \) due to heterogeneous chemical reactions, \( A_k \) the molar concentration of species \( k \) at the fluid–solid interface and \( \hat{n} \) is the outward unit surface normal. Since \( R_k \) is an extremely non-linear function of the molar concentrations (or mass fractions) (Coltrin et al., 1991; Mazumder & Lowry, 2001), Eqs. (10) represents a non-linear set of differential algebraic equations (DAE). The solution of this stiff set of non-linear DAE is generally obtained using the Newton method, but requires special pre-conditioning to address stiffness and ill-posed-ness in the case of steady state solutions. Details pertaining to these numerical issues may be found elsewhere (Mazumder & Lowry, 2001). The solution of Eqs. (10) provides the near-wall mass fractions and mass fluxes (represented by the left-hand side of Eq. (10b)) of all gas-phase species, which appear as sources/sinks for control volumes adjacent to the surface in a finite-volume formulation (Mazumder & Lowry, 2001).

The balance of energy at the surface yields the following equation

\[
\left[ \begin{array}{c} -k \nabla T + q_R + \sum_{k=1}^N j_{khk} \\
\end{array} \right]_F \circ \hat{n} = \left[ -k \nabla T + q_R \right]_S \circ \hat{n} \] (11)

where the subscript “F” denotes quantities on the fluid side of the fluid–solid interface, while the subscript “S” denotes quantities on the solid side of the same interface. The solution of Eq. (11), which is also a non-linear equation, yields the temperature at the fluid–solid interface, and subsequently provides the flux of energy at the interface, which can then be used as a source/sink for the cells adjacent to the interface after appropriate linearization. In this enthalpy formulation, the heat of surface reaction actually manifests itself through the \( \sum j_{khk} \) term.

Eqs. (1)–(3) along with Eq. (9), when solved along with the appropriate boundary conditions just described, will produce flow, temperature and mass fraction distributions of all species within the monolith tube.

3. Results and discussion

The governing equations, described in the preceding section, were solved using the commercial CFD code CFDE-ACE+. It is customary to treat individual channels within a monolith catalytic converter as channels of circular cross-section (Deutschmann & Schmidt, 1998; Raja et al., 2000; Stutz & Poulikakos, 2005), as schematically shown in Fig. 2. Thus, simulations were performed in a single channel with circular cross-section of inner diameter \( D = 1.8 \) mm and length \( 2L = 5 \) cm. All simulations were performed on a uniform grid with 150 cells in the axial direction and 48 cells in the radial direction. A grid independence study for nominal parameter values showed that calculations on a \( 300 \times 96 \) grid yield results that are within 1% of the values from the \( 150 \times 48 \) grid, and thus the coarser of the two meshes was used for further parametric studies.

Methane–air mixture of specified composition was introduced into the monolith tube at a prescribed velocity. A plug velocity profile was imposed at the inlet. Variation of the inlet velocity translates to a variation in the inlet Reynolds number, \( Re_D = \rho U_{in} D / \mu \), which is one of the parameters in the simulations. The outer walls of the tubes were treated as adiabatic.
boundaries. Such a choice is consistent with the fact that if a tube in the center of the monolith is considered, it is surrounded by other tubes that have almost the same temperature. Thus, little or no heat transfer can occur from tube to tube, at least in the central core of the monolithic plug. Newton cooling boundary conditions, including both convection and radiation, were employed at the ends (both front and back) of the ceramic tubes. As stated earlier, surface-to-surface gray radiation transport was included. The emissivity of the inner walls of the tube was assumed to be 0.5, due to lack of better information. The same emissivity value was also used for the tube ends to account for external heat loss by radiation. The inlet and the outlet were assumed to have an emissivity of unity, implying that no radiation gets reflected back from these boundaries. Conversion of the fuel is assumed to take place because of heterogeneous (surface) reactions at the walls only. This is justified since the range of temperature observed is such that homogeneous combustion may be neglected (Raja et al., 2000). The tube wall in the region near the entrance was assumed to be non-reacting up to \( x/L = 0.2 \) in order to allow the incoming plug flow to develop to some extent prior to reactions occurring at the walls (see Fig. 2). Since study of ignition and blowout phenomena is one of the objectives of this investigation, a detailed 24-step surface reaction mechanism (Deutschmann et al., 1994; Raja et al., 2000) was chosen for this study, rather than a single-step empirical rate expression. Initially, a single-step reaction mechanism (Song, Williams, Schmidt, & Aris, 1991) was used in this work. It was found that the single-step reaction mechanism is unable to predict ignition and blowout. The fact that detailed multi-step reaction mechanisms are necessary to predict ignition and blowout is well known, and thus, these findings are not surprising. Thus, the detailed reaction mechanism used here is a necessity and not an arbitrary choice. Furthermore, this particular multi-step reaction mechanism has been used and validated extensively (Deutschmann et al., 1994; Raja et al., 2000), including studies conducted by the authors (Mazumder & Sengupta, 2002). Table 1 summarizes the parameters used for the simulations.

Three different parameters were varied for this particular study, namely the Reynolds number (or flow rate), inlet equivalence ratio, and the wall thermal conductivity. Although the equivalence ratio was varied from 0.8 to 1, it was found that only mixtures very close to stoichiometric (\( \Phi = 1 \)) ignited. Thus, only results for \( \Phi = 1 \) are presented in this article. Initially, a decision was made to treat the gap size (between two building blocks) as a parameter, as well. However, it was found that a gap size of 1 \( \mu \)m did not produce any noticeable differences in results between the split and continuous tube. A gap size of 100 \( \mu \)m (0.1 mm) is unrealistic since 0.1 mm is the thickness of the wall of the tube itself. Thus, simulations were finally performed with a gap size of 10 \( \mu \)m. Again, the choice was prompted by lack of better information, and the fact that the objective of this study is to elucidate behavioral differences, and not necessarily quantitative validation. The mean free path of air at room temperature and pressure is approximately 54 nm, based on kinetic theory calculations (Bird et al., 2001)—about two orders smaller than the gap size used here. Thus, no special thermal contact model was necessary to treat thermal transport within the gap. A simple conductive resistance model was used. It is informative to estimate the radiative and conductive heat fluxes across the gap to determine if radiation through the gap should be taken into account. For example, if the two surfaces across the gap have temperatures equal to \( T_1 = 1100 \) and 1000 K, the maximum radiative heat flux is \( \sigma(T_1^4 - T_2^4) = 26,314 \text{ W/m}^2 \). The conductive flux, on the other hand, is \( k_{\text{gap}}(T_1 - T_2)/d = 800,000 \text{ W/m}^2 \), using a gap size, \( d \), of 10 \( \mu \)m, and a thermal conductivity, \( k_{\text{gap}} \), of 0.08 W/m/K, corresponding to the thermal conductivity of air at 1 bar and

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value/equation/description</th>
<th>Comment/source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length of building block, ( L )</td>
<td>2.5 cm</td>
<td>Typical value</td>
</tr>
<tr>
<td>Inner diameter of tube, ( D )</td>
<td>1.8 mm</td>
<td>Typical value</td>
</tr>
<tr>
<td>Thickness of tube wall</td>
<td>0.1 mm</td>
<td>Typical value</td>
</tr>
<tr>
<td>Emissivity of wall, ( \varepsilon_w )</td>
<td>0.5</td>
<td>Assumed</td>
</tr>
<tr>
<td>Emissivity of inlet, ( \varepsilon_{in} )</td>
<td>1.0</td>
<td>Assumed</td>
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<tr>
<td>Convective heat transfer coefficient, ( h )</td>
<td>200 W m(^{-2}) K(^{-1})</td>
<td>Assumed</td>
</tr>
<tr>
<td>Far field temperature, ( T_{\infty} )</td>
<td>600 K</td>
<td>Assumed</td>
</tr>
<tr>
<td>Inlet temperature, ( T_{in} )</td>
<td>600 K</td>
<td>Typical value</td>
</tr>
<tr>
<td>Inlet axial velocity, ( U_{in} )</td>
<td>Obtained from prescribed ( Re ), using ( U_{in} = Re\mu_{in}/(\rho_{\infty}D) )</td>
<td>Typically used</td>
</tr>
<tr>
<td>Initial temperature for transient simulations</td>
<td>1000 K everywhere</td>
<td>Typically used</td>
</tr>
<tr>
<td>Initial composition for transient simulations</td>
<td>Air everywhere</td>
<td>Typically used</td>
</tr>
<tr>
<td>Cordierite properties</td>
<td>Thermal conductivity</td>
<td>3 W m(^{-1}) K(^{-1})</td>
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<tr>
<td>Density</td>
<td>2511 kg m(^{-3})</td>
<td>Lynch (1975)</td>
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<tr>
<td>Specific heat capacity</td>
<td>1046 J kg(^{-1}) K(^{-1})</td>
<td>Lynch (1975)</td>
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<tr>
<td>Silicon carbide properties</td>
<td>Thermodynamic properties</td>
<td>170.39–0.60967 W m(^{-1}) K(^{-1})</td>
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<tr>
<td>Density</td>
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<td>Riedel (2000)</td>
</tr>
<tr>
<td>Specific heat capacity</td>
<td>490.52 + 0.61227 J kg(^{-1}) K(^{-1})</td>
<td>Riedel (2000)</td>
</tr>
</tbody>
</table>

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1100 K. Thus, it is clear that the highest possible radiative heat flux is at least one order smaller than the actual conductive heat flux across the gap. Actual computed temperature distributions with and without radiation in the gap will be shown shortly to illustrate this point further.

All transport properties of the fluid, namely viscosity, thermal conductivity and binary diffusion coefficients were computed using the Chapman-Enskog equations of kinetic theory (Bird et al., 2001; Hirschfelder, Curtiss, & Bird, 1954), available as part of the CFD-ACE+TM code. The Lennard-Jones potentials, which are needed as inputs, were obtained from the CHEMKIN database, which is built into the CFD-ACE+TM code. The solutions were deemed to be converged when the residuals of each of the conservation equations decreased by five orders of magnitude.

Figs. 3 and 4 show the ignition characteristics of heterogeneous combustion of a stoichiometric methane–air mixture on platinum supported on silicon carbide and cordierite tubes, respectively. In these figures, the flow rate (Reynolds number) is treated as a parameter. Several interesting observations may be made. First, the range of Reynolds number for which ignition occurs in the two cases (silicon carbide versus cordierite) are completely different. The ignition occurs over a much wider range in the case of silicon carbide, indicating that it is probably a better candidate for the development of robust monolith plugs. Physically, axial heat conduction through silicon carbide is much more pronounced than in cordierite (the average thermal conductivity of silicon carbide is about 100 W m⁻¹ K⁻¹, while that of cordierite is about 3 W m⁻¹ K⁻¹), enabling heat generated at the point of attachment of the flame to propagate to other locations of the tube, thereby facilitating the stability of the flame. Secondly, in the case of cordierite, when ignition occurs, no noticeable difference is observed between the split and the continuous tube. Both tubes ignite or blow out in a similar fashion. In contrast, for silicon carbide, the split tube ignites over a much wider range of Reynolds number than the continuous case. The final observation to be made is that in the case of cordierite, once ignition occurs, the steady state conversion fraction is approximately 0.95, irrespective of the Reynolds number. In contrast, for silicon carbide, the conversion fraction decreases monotonically with increase in Re until blowout occurs. However, even for silicon carbide, at low Reynolds numbers, the conversion fractions are more or less independent of Reynolds number.

It is clear from the above results that the effect of splitting the tube only manifests itself if the thermal conductivity of the tube is high. Otherwise, wall heat conduction is weak to begin with (as in cordierite), and whether additional conduction resistance is imposed or not plays no role in the overall ignition and flame attachment process. One question that remains for the silicon carbide cases is why there is no difference in conversion fractions between split and continuous tubes at low-intermediate Reynolds number, while there is big difference above a certain Reynolds number. To answer this question, temperature and mass fraction distributions were studied for a case where both tubes ignited (Fig. 5), such as $Re_D = 300$. Comparison of the temperature distributions between the split and continuous tubes show observable differences in wall temperature, particularly near the splitting point. However, these observable differences in temperature do not translate to observable differences in mass fraction of methane. Hence, the conversion fractions are similar. At low flow rates, the flame attachment point is close to the inlet, and almost 85% of the methane already gets consumed in the first tube section. Thus, any difference in wall temperature in the second half of the tube hardly has any effect on the overall conversion fraction. With increase in flow rate, the flame attachment point is expected to shift further downstream, in which case, the effect of splitting the tube is expected to be amplified, as was already corroborated by the results shown in Fig. 3. To elucidate this phenomenon further, snapshots obtained from transient simulations at different instances of time are depicted in Fig. 6. These figures clearly show that the point of attachment of the flame in this case is in the sec-
Fig. 5. Differences between split and continuous tubes for heterogeneous combustion of a stoichiometric methane–air mixture on platinum supported on a silicon carbide monolith at steady state ($Re = 300$).

Fig. 6. Comparison of temperature distribution between split and continuous silicon carbide monolith tubes from the onset of ignition to steady state ($Re = 600$).

For the continuous monolith case, the heat is transferred upstream by axial conduction, and is eventually lost by both conduction and radiation to the relatively cold inlet. In the split monolith case, the heat is prevented from traveling upstream by the thermal resistance of the small air gap, and therefore, cannot be dissipated as effectively. This facilitates attachment of the flame and enhances its stability at steady state.

In order to corroborate the earlier claim that radiation transport through the gap has no impact on the results, the simulations just described for $Re = 300$ and 600 were repeated with radiation through the gap. Fig. 7 shows the axial temperature distributions along the tube wall with and without radiation through the gap. The results with and without radiation through the gap are so close that they cannot be identified separately on this figure. Table 2 shows the actual temperature values across

| $T_1$ (K) | $T_2$ (K) | $\Delta T = T_1 - T_2$ (K) | $q_{\text{rad}} = |\varepsilon\sigma(T_1^4 - T_2^4)|$ (W/m$^2$) | $q_{\text{con}} = |k_{\text{gap}}\Delta T/d|$ (W/m$^2$) |
|-----------|-----------|-----------------|-----------------|-----------------|
| No radiation | 1345.21 | 1302.54 | 42.67 | 0 | 341,360 |
| Radiation, gap surface $\varepsilon = 1.0$ | 1345.05 | 1302.69 | 42.36 | 22,297 | 338,880 |

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Fig. 7. Axial temperature distributions along the monolith tube middle at various Reynolds numbers with and without the gap, and with and without radiation transport through the gap.

the gap for one of the cases. While radiation transport through the gap decreases the temperature drop across the gap, it is evident from the computed results that the effect is too small to be of any consequence to the overall conclusions.

From an engineering perspective, the results of this investigation indicate that high thermal conductivity ceramic supports are more robust as design materials since they allow ignition and flame stability over a wider range of conditions. The downside is that such materials may be susceptible to different behavior when the monolith is built with smaller building blocks as compared to continuous monolith tubes.

4. Summary and conclusions

Computational fluid dynamics calculations have been performed to elucidate key behavioral differences in heterogeneous combustion within continuous and split monolith tubes. Methane–air combustion on platinum is chosen as the candidate system, and a 24-step reaction mechanism is used to describe the surface chemistry. In the split tube case, two building blocks of length 1 in. are used, and an air gap of 10 μm is introduced between the two blocks. Two commonly used ceramic materials, namely cordierite and silicon carbide are used for the supporting monolith material. Silicon carbide is representative of a material with high thermal conductivity, while cordierite is representative of a material with low thermal conductivity. Both materials have comparable thermal capacitance.

Simulation results showed that ignition occurs in cordierite tubes over a very narrow range of Reynolds number, typically ranging between 35 and 80. In contrast, ignition occurred in silicon carbide tubes in the range of Reynolds numbers between 20 and 1000, indicating that silicon carbide monoliths are likely to be much more robust and less susceptible in uncertainties in the Reynolds number. Within the narrow range of Reynolds numbers for which ignition occurred in cordierite tubes, it is observed that the steady state conversion fraction is virtually independent of the Reynolds number: in all cases, about 95% conversion was obtained. For silicon carbide, on the other hand, the conversion fraction decreased monotonically with increasing Reynolds number, until blowout occurred.

The difference in behavior between single and split monolith tubes manifested itself only for the high thermal conductivity material, namely silicon carbide. For cordierite, both tubes behaved almost identically. For silicon carbide, it was found that the introduction of a thin gap between the two sections of the tube reduced heat loss by axial conduction, facilitating attachment of the flame and subsequent steady state combustion. In the case of continuous tubes, even though ignition occurred, the flame could not be sustained due to rapid heat loss by axial conduction.

From an engineering perspective, the simulation results presented here can serve as guidelines for future design and troubleshooting of catalytic combustion processes. The work is easily extendable to other practical chemical systems, pending availability of reasonably accurate reaction mechanisms.

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