CFD Modeling of the Combustion Characteristics and Stability in Catalytic Micro-Channels

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Abstract

Combustion characteristics and stability of premixed methane-air mixtures in catalytic micro-channels is studied numerically, using CFD (computational fluid dynamics). The characteristics of the fluid mechanics are also analyzed. A two-dimensional CFD model with detailed reaction mechanisms and multicomponent transport is developed to evaluate the effect of operating conditions on the combustion stability. The laminar flow is assumed, and steady-steady simulations are performed. The fuel-lean equivalence ratio operation limit for the system is determined by the analysis of Reynolds number. The primary focus is on CFD as a means of understanding thermal management at small scales. It is shown that an appropriate choice of the flow velocity is crucial in achieving the self-sustained operation. Large gradients in temperature and species concentration are observed, despite the small scales of the system under certain conditions. The flow velocity is very important as it determines the flame location. Furthermore, the flow velocity plays a dual, competing role in the stability of the system. Low flow velocities reduce the heat generation, whereas high flow velocities reduce the convective time-scale. There is a narrow regime of flow velocities that allows self-sustained operation. When a low-power system is desired, highly insulating materials should be preferred, whereas a high-power system would favor highly conductive materials. Engineering maps that delineate combustion stability are constructed. In order to gain further insight into the combustion characteristics of the system, the optimum Reynolds number is determined. Finally, design recommendations are made.

Keywords: Micro-combustion; Combustion Stability; Thermal Management; Catalytic Combustion; Laminar Flow; Computational Fluid Dynamics

1. Introduction

The push toward the miniaturization of electromechanical devices and the resulting need for micropower generation with low-weight devices has greatly promoted the development of the field of combustion in small channels (Fernandez-Pello, 2002). The characteristic length of the micro-electro-mechanical systems being developed is sufficiently larger than the molecular mean free...
path, and the physical-chemical behavior of the fluids is fundamentally the same as in their macro-scale counterparts. Consequently, the continuum assumption of fluid mechanics and the no-slip condition will still apply (Harley et al., 1995). However, the small characteristic length scale of these systems causes particular characteristics of the combustion, heat transfer, and fluid mechanics involved in the operation of the device.

The small characteristic length scales of these systems constrain the flows to relatively small Reynolds numbers (Kaisare and Vlachos, 2012). The flow will be mainly laminar for these systems and thus the mixing of different species will be determined by diffusion (Ho and Tai, 1998). In addition, viscous effects become significant, and the aspect ratios of these systems are large. Therefore, frictional losses will be high and increased pumping will be required (Walther and Ahn, 2011). On the other hand, viscous forces might be advantages, such as leakage of gases through moving surfaces or through joints (Guo and Zheng, 2008).

Due to the small length scales of these systems, unexpected behavior might be encountered. The most obvious effect is that the flow in these cases operates in the near-Stokesian regime in which the Reynolds numbers are typically very small (Chen and Tian, 2010). Small changes in temperature not only can strongly affect the volumetric flow rate but also can significantly change the fluid-wall interactions that are usually ignored in macroscopic flows (Gat et al., 2008). In addition, the small characteristic length scales of these systems will cause large gradients in flow velocity. This will lead to high convective heat transfer coefficients and high wall frictional losses (Asako and Toriyama, 2005). These in turn may lead to enhanced diffusive mixing, high transverse heat transfer, and large pressure losses within the system (Hong and Asako, 2007).

These systems must use the ambient air as the source of oxygen in order to maintain their performance edge in energy density as well as to reduce volume, as found in their large-scale counterparts. This indicates that the liquid hydrocarbon fuel should be injected directly into the combustion chamber in a non-premixed system, or evaporated and mixed with the air prior to entering the combustion chamber in a premixed system (Mitroglou et al., 2014). The small length scales of these systems could strongly affect the dynamics and energy requirements associated with phase change and multiphase flows (Kavehpour et al., 1997). Furthermore, it has been found that the liquid-vapor phase transition in these systems occurs abruptly in an unstable manner and abruptly (Serizawa et al., 2002). This behavior is entirely differently from the phase transition found in large-scale devices (Chung and Kawaji, 2004). This is partially because of the high surface-area-to-volume ratio that enhances the nucleation and wetting effects at the wall and to the behavior of bubbles that is significantly affected by the small characteristic length scales and distinctly differs from the macroscopic behavior.

Mixing of the reactants in micro-chemical systems encounters a potential problem due to the planar nature of devices and the low Reynolds number. Although the small characteristic length scales of these systems are conducive to their diffusive mixing, the residence time of the fluids is still relatively short and might be insufficient to ensure complete mixing under certain conditions (Oechsler et al., 2016). Furthermore, the overall length of the system might be too long, causing significant pressure losses (Ferrouillat et al., 2006). In some cases, different dynamic mixing approaches might have to be implemented in order to improve and enhance the mixing of species, even though they will consume valuable energy and will introduce fabrication complexities. Several
approaches have been employed to increase mixing rates within the system, primarily of liquids (Villermaux and David, 1983). The main problems with these dynamic mixing approaches are system size, fabrication complexity, and increased pressure drops (Mark et al., 2010). Although these studies have been geared to mix liquids, similar concepts might be applied to mixing gases.

Combustion at the micro-scale can offer advantages. Aside from the larger radial heat transfers, faster ignition can happen, and low temperatures could be envisioned that could be beneficial for reduction of nitrogen oxides (Ju and Maruta, 2011). However, loss of flame stability due to blowout, extinction, and high wall temperatures are main issues that require careful thermal management. Overall heat management has been found to play a critical role in determining flame stability in micro-chemical systems (Kang and Veeraragavan, 2015). From a practical standpoint, these systems enable easy integration with other elements, such as endothermic microreactors, in multifunctional devices (Ramaswamy et al., 2006). Such devices integrate multiple reactions coupled via facilitated heat exchange through a conductive wall with fewer or no heat transfer boundary layers being involved. Thermal coupling between exothermic and endothermic reactions can be enhanced in a number of ways. One strategy might be to use “nanofluids” (Bouchta et al., 2017). Nanofluids are made up of small nanosized particles suspended in a base fluid (Xuan and Li, 2000). These added nanoparticles have been found to increase thermo-physical properties such as thermal conductivity, thermal diffusivity, viscosity, and convection heat transfer compared to their base fluids (Wang and Mujumdar, 2007). As a consequence, higher thermal efficiencies are achievable, and the device performance can be improved greatly (Turkyilmazoglu, 2016). Furthermore, the anomalous heat transfer enhancement by slip has been reported in the literature (Turkyilmazoglu, 2015). The utilization of nanofluids to enhance the heat transfer performance in micro-chemical systems shows promise but is a difficult task (Molana, 2016). Optimization techniques may be necessary to achieve this goal.

While flame propagation at the micro-scale is feasible, the interaction between transport and kinetics in combustion characteristics and stability of these systems is still poorly understood. The inability of conducting spatially resolved measurements, inherent to the micro-scale, underscores the need for detailed CFD modeling. Additionally, due to the high surface-area-to-volume ratio, the potential for the loss of combustion stability needs to be exploited for robust design. Furthermore, to design micro-chemical systems, the effects of wall thermal conductivity and channel dimension on combustion stability must be explored for optimal thermal management. This study focuses on a simple single-channel system. From a practical standpoint, this system enables easy integration with other elements, such as thermo-electrics and/or endothermic micro-reactors, in integrated devices. Such devices integrate multiple reactions coupled through facilitated heat exchange through a conductive wall with fewer or no heat transfer boundary layers being involved. Two-dimensional CFD simulations are performed, using detailed reaction mechanisms and multicomponent transport. The effect of flow velocity on the combustion characteristics and stability are delineated. Stability diagrams that delineate combustion stability are constructed. Based on these insights, design recommendations are finally made. The objective of this study is to model the combustion characteristics of methane-air mixtures in catalytic micro-channels. Of special interest in this study is on understanding the effect of flow velocity within the system.

2. Computational Fluid Dynamics Model

Considerable progress has been made on the development of micro-power generation systems
Due to the small nature of these systems, it is extremely difficult to be able to determine the heat and mass transfer characteristics (i.e. temperature distribution, flow distribution, etc.) internal to the reactors. Numerical simulations are critical to understand heat and mass transfer phenomena occurring in the systems and help guide the further improvements (Chein et al., 2012). They are also able to improve the designs to achieve the desired performance. Furthermore, the numerical simulation tools have proven to be extremely useful in evaluating new designs, understanding the manifold impact on flow distribution, and projecting thermal distribution and efficiency (Holladay and Wang, 2015).

2.1. Geometric Model

Figure 1 shows the schematic diagram of the catalytic micro-channel. The arrow indicates the direction of flow. To illustrate the primary mechanism, a relatively simple system may provide valuable insights. Premixed, stoichiometric methane-air mixtures are fed to the micro-channel, and hot product gases exit. The catalytic micro-channel consists of two parallel, infinitely wide plates of 0.8 mm apart, and length of 5.0 mm. The walls have finite thickness 0.2 mm. Platinum catalysts are coated on the internal walls of the parallel plates. The micro-channel is modeled as a two-dimensional system due to the large aspect ratio. In order to minimize the computational intensity, only half of the system is modeled, given the inherent symmetry of the geometry. The methane fuel is examined, as it is more challenging to the development of micro-power generation due to the significantly increase in reaction time as compared to hydrogen.

2.2. Mathematical Model

CFD simulations can play an important role in micro-channel technology by providing analysis of specific micro-chemical systems, design of new systems, and evaluation of potential performance advantages relative to conventional chemical systems. The small length scales of the channel imply that transport processes are strongly affected by diffusion. Yet, the characteristic length scales of these systems are still sufficiently large compared to the mean free path of gas molecules, i.e., the Knudsen number is very small. Therefore, classical, continuum descriptions are appropriate. Furthermore, reactive flows in the channel are laminar and more easily quantified than the complex, often turbulent flows characteristic of conventional chemical systems. A two-dimensional numerical model is developed using the commercial CFD software ANSYS Fluent, incorporated with detailed reaction mechanisms and multicomponent transport. The Reynolds number is less than 800, and thus the laminar flow assumption within the system is valid. Steady-steady simulations are performed. A finite-volume method is used to discretize the two-dimensional steady-state energy equation in the wall and the two-dimensional steady-state continuity, momentum, energy and species conservation equations in the fluid (Appel et al., 2002).
Continuity equation:
\[
\frac{\partial (\rho u)}{\partial x} + \frac{\partial (\rho v)}{\partial y} = 0. 
\]  

(1)

Momentum equations:
\[
\frac{\partial (\rho uu)}{\partial x} + \frac{\partial (\rho vv)}{\partial y} + \frac{\partial p}{\partial x} - \frac{\partial \left(2\mu \frac{\partial u}{\partial x} + \frac{2}{3} \mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y}\right)\right)}{\partial x} - \frac{\partial \mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y}\right)}{\partial y} = 0. 
\]

(2)

\[
\frac{\partial (\rho uu)}{\partial x} + \frac{\partial (\rho vv)}{\partial y} + \frac{\partial p}{\partial x} - \frac{\partial \left(2\mu \frac{\partial v}{\partial y} + \frac{2}{3} \mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y}\right)\right)}{\partial y} - \frac{\partial \mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y}\right)}{\partial x} = 0. 
\]

(3)

Energy equation:
\[
\frac{\partial (\rho u h)}{\partial x} + \frac{\partial (\rho v h)}{\partial y} + \frac{\partial}{\partial x} \left(\rho \sum_{k=1}^{K_g} Y_k V_{k,x} - \lambda_g \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y} \left(\rho \sum_{k=1}^{K_g} Y_k V_{k,y} - \lambda_g \frac{\partial T}{\partial y}\right) = 0. 
\]

(4)

Gas phase species equation:
\[
\frac{\partial (\rho u Y_k)}{\partial x} + \frac{\partial (\rho v Y_k)}{\partial y} + \frac{\partial}{\partial x} \left(\rho Y_k V_{k,x}\right) - \frac{\partial}{\partial y} \left(\rho Y_k V_{k,y}\right) - \omega_k W_k = 0, \quad k = 1, \ldots, K_g. 
\]

(5)

Surface species coverage equations:
\[
\sigma_m s_m = 0, \quad m = K_g + 1, \ldots, K_g + K_s. 
\]

(6)

In the above equations, \(\rho\) is the density of the gas mixture, \(x\) and \(y\) are the axial co-ordinate and transverse co-ordinate, \(u\) and \(v\) are the corresponding flow velocity components, \(\mu\) is the viscosity of the gas mixture, \(\lambda_g\) is the thermal conductivity of the gas mixture, \(p\) is the pressure, \(h\) is the total enthalpy of the gas mixture, \(K_g\) is the total number of gas phase species, \(T\) is the temperature, \(Y_k, \omega_k\), \(W_k\), and \(h_k\) are the mass fraction, molar production rate, relative molecular mass, and total enthalpy of the \(k\)th gas phase species respectively, and \(V_{k,x}, V_{k,y}\) are the \(x\)- and \(y\)-components of the \(k\)th species diffusion velocity. Finally, \(\sigma_m\) and \(s_m\) denote the number of occupied surface sites and the molar production rate of the \(m\)th surface species, respectively; \(K_s\) is the total number of surface species.

Species diffusion velocities \(V_k\) are computed using mixture average diffusion, including thermal diffusion for the light species (Kee et al., 1998):
\[
V_k = \frac{-D_{k,m}}{T} \left[ \ln \left( \frac{Y_k W_k}{W_k} \right) \right] + \left[ D_k^T \left( \rho Y_k W_k \right) \right] \nabla (\ln T). 
\]

(7)

Here, \(D_{k,m}\) and \(D_k^T\) are the mixture-average diffusion coefficient and thermal diffusion coefficient of \(k\)th gaseous species, respectively; and \(W_k\) is the mixture average relative molecular mass. Since the heat transfer along the walls significantly affects stability (Federici and Vlachos, 2008), the two-dimensional energy equation is explicitly accounted for within the walls:
\[
\frac{\partial}{\partial x} \left( \lambda_s \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( \lambda_s \frac{\partial T}{\partial y} \right) = 0. 
\]

(8)

Here, \(\lambda_s\) is the thermal conductivity of the solid wall. The interfacial boundary conditions for the gaseous species at the specified gas-wall interface \((y = d/2)\) are written as
\[
(\rho Y_k V_{k_y})_{y=d/2} + W_k (s_k)_{y=d/2} = 0, \quad k = 1, \ldots, K_g.
\]  \(9\)

The energy boundary conditions at the specified gas-wall interface \((y = d/2)\) are given as

\[
q_{\text{rad}} - \lambda_y \left(\frac{\partial T}{\partial y}\right)_{y=d/2} + \lambda_y \left(\frac{\partial T}{\partial y}\right)_{y=d/2} + \sum_{k=1}^{K_g} (s_k W_k)_{y=d/2} = 0.
\]  \(10\)

Here, the subscripts (-) and (+) denote the properties just below and above the gas-wall interface, respectively. To compute the radiation exchange within the system, the net radiation method for diffuse-gray areas is used. The discrete ordinates radiation model is adopted to model the surface-to-surface radiation. The emissivity of each surface element is assumed to be 0.8. Gas radiative emission and absorption are not considered, given the small optical paths and the large nitrogen dilution. Solid-phase radiative boundary conditions are applied at the inlet and outlet vertical wall faces. For all the external surfaces, the total heat loss to the surroundings is taken into account:

\[
q = h_{\text{o}} (T_{w,o} - T_{\text{amb}}) + \varepsilon \sigma (T_{w,o}^4 - T_{\text{amb}}^4).
\]  \(11\)

Here, the external heat transfer coefficient \(h_{\text{o}}\) is assumed to be 20 W/(m²·K); \(\varepsilon\) is the surface emissivity; \(\sigma\) is the Stephan-Boltzmann constant; \(T_{\text{amb}}\) is the ambient temperature.

2.3. Chemical Kinetics

There have been numerous efforts to model catalytic microreactors. Many of these models are two-dimensional models that include detailed heat and mass transfer in an attempt to accurately model the reactor operation. Unfortunately, because of the numerical complexity involved in solving the resulting partial differential equations, simplified reaction mechanisms are often used to describe the homogeneous reactions. Many of the models assume that gas-phase chemistry is negligible in the system being modeled. Other models have used simplified heterogeneous mechanisms to avoid solving the highly non-linear surface-species balances along the length of the catalyst. Simplified kinetic mechanisms can give reasonable understanding of the rates of reaction and evolution of heat in microreactors, but because the mechanisms themselves contain assumptions regarding the product distributions, they provide little insight into the chemistry taking place in the reactor. The high temperatures observed in short-contact-time catalytic microreactors, in conjunction with operation well into the flammability limits, renders it important to model simultaneously both the homogeneous and heterogeneous chemistry using detailed reaction mechanisms to better understand the role of each mechanism in determining the product distributions and reaction rates in the reactor. A parallel plate reactor geometry can be assumed to reduce the governing partial differential equations to easily integrable ordinary differential equations coupled with algebraic expressions to describe the heterogeneous chemistry. It is therefore of great interest to model the chemistry that takes place under certain conditions to better understand the combustion characteristics and stability in catalytic micro-channels. The reaction system considered in this study is the catalytic combustion of methane-air mixtures over supported platinum catalysts. Depending on the operation conditions and especially at high temperatures, homogeneous reactions may also take place. In this study, the initiation of homogeneous reactions is possible at the reactor operating temperature, providing another route for heat generation. Therefore, homogeneous reactions are not negligible, resulting to the complexity of the CFD model.
Thermal quenching occurs when sufficient heat is removed through the walls, that combustion cannot be self-sustained (Vlachos et al., 1993). Radical quenching occurs via adsorption of radicals on the system walls and subsequent recombination, resulting in the lack of homogeneous chemistry (Aghalayam et al., 1998). The small scales of these systems make them significantly more prone to both quenching mechanisms due to the high surface area to volume ratios, i.e., enhanced heat transfer from the flame to the walls and increased radical mass transfer. Both the heat loss and wall radical quenching are key interfacial phenomena for understanding flame stability in micro-channels (Raimondeau et al., 2002). The focus here is on understanding the overall heat-transfer characteristics in catalytic micro-channels and developing guidelines for appropriate thermal management that can result in more robust flame behavior. Detailed chemical reaction mechanisms are a useful tool for describing flame dynamics and flame responses to external perturbations (Simmie, 2003). Therefore, detailed heterogeneous/homogeneous reaction mechanisms are incorporated in the CFD model. The detailed heterogeneous reaction scheme is based on the mechanism proposed by Deutschmann et al. (2000). The mechanism has been developed to predict ignition and steady-state operation for the catalytic combustion of methane-air mixtures on platinum surfaces. The mechanism consists of 9 gaseous and 11 surface species involved in 24 elementary reactions, including the adsorption and desorption of reactants and products, as well as surface reaction steps. Note that the mechanism assumes hydrogen adsorption is first order in surface sites even though hydrogen adsorbs dissociatively. Furthermore, methane is assumed to be irreversibly adsorbed, and carbon monoxide adsorption is assumed to be second order in platinum sites (Deutschmann et al., 1996). A surface site density is specified to be $2.72 \times 10^{-9}$ mol/cm$^2$ for the supported platinum catalyst. Gas phase species adsorption rates are described in terms of sticking coefficient; the probability that a collision of the particular species $k$ with the surface will result in adsorption. The adsorption rate constant $k_{ads,k}$ of $k$-th gaseous species is modeled using a modified Motz-Wise correction (Dogwiler et al., 1999):

$$k_{ads,k} = \left( \frac{2}{2-\gamma_k \theta_{free}} \right) \frac{\gamma_k}{\Gamma^m} \sqrt{\frac{RT}{2\pi W_k}}.$$  

(12)

In the above equations, $\gamma_k$ is the sticking coefficient of the $k$-th gaseous species, $\theta_{free}$ is the free surface coverage, $\Gamma$ is the total surface site density, and $m$ is the sum of the reactant's stoichiometric coefficients. The detailed homogeneous reaction scheme is based on the Leeds methane oxidation mechanism (Hughes et al., 2001), complemented and corrected by Turányi et al. (2002). The mechanism has been tested against experimental data. Thermodynamic data are taken from the provided schemes, and transport properties are computed using the CHEMKIN transport database. Homogeneous and heterogeneous reaction rates are evaluated with CHEMKIN (Kee et al., 1996) and Surface-CHEMKIN (Coltrin et al., 1996), respectively.

### 2.4. Boundary Conditions

The governing equations must to be solved with appropriate boundary conditions. At the inlet, a fixed, flat velocity profile is used. While this boundary condition fixes the convective component of the flux of energy and species, the diffusive component is highly dependent on the gradient of the computed species or temperature fields within the system. At the fluid centerline, symmetry boundary conditions are employed. At the exit, a fixed pressure is specified and far-field conditions are imposed for the rest of the variables. No-slip boundary condition is employed for both velocity components at the gas-wall interface. At the outlet and the symmetry plane, the transverse flow...
velocity is set to zero and zero-Neumann conditions are employed for all other scalars. The heat flux at the gas-wall interface is computed and continuity in temperature and heat flux links the fluid and solid phases. For the left and right edges of the wall, heat losses are assumed. Furthermore, all internal heat transfer between the fluid and the wall is computed by accounting explicitly for the convective and conductive heat transfer in the two-dimensional CFD model within the system.

2.5. Computation Scheme

In order to perform these CFD simulations, an orthogonal staggered grid is employed. The solid wall is discretized such that the elements at the gas-wall interface have the same axial size as the corresponding elements on the fluid sides. The grid is finer where the gradients in temperature and species concentration are steeper. In order to determine the optimal density as well as node spacing that would minimize computation time and give the desired accuracy, CFD simulations are performed using grids with varying nodal densities. Figure 2 shows the centerline temperature profiles for grids with different nodal densities. There exists a convergence of the solution with the increase of grid density. The coarsest grid, consisting of 8000 nodes in total, fails to accurately capture the maximum temperature as well as the inflection point at the ignition point. Solutions obtained with grids consisting of tens of thousands of nodes are reasonably accurate. All solutions presented herein are obtained using a grid consisting of 20000 nodes, i.e., 20 transverse nodes within the wall as well as 200 axial nodes by 80 transverse nodes within the fluid. Larger grid densities, up to 60000 nodes, yield no obvious advantage.

The fluid density is computed using the ideal gas law. The species specific heat is computed using a piecewise polynomial fit of temperature, and the fluid thermal conductivity, specific heat, and viscosity are computed from a mass fraction weighted average of species properties. In order to discretize the CFD model, the second-order upwind scheme is employed. The “SIMPLE (Semi-Implicit Method for Pressure-Linked Equations)” algorithm is employed to solve for the pressure and velocity fields. In order to solve the governing equations, a segregated solution solver is
employed, using an under-relaxation method. The segregated solver first solves the momentum equations, then solves the continuity equation, and updates the flow rate and pressure. Subsequently, the energy and species equations are solved, and convergence is examined. The latter is monitored through both the values of the residuals of the conservation equations and the difference between subsequent iterations of the solution. Generally, convergence is very difficult due to the disparity between the fluid and the wall thermal conductivities as well as the inherent stiffness of the hetero-/homogeneous chemistry. The simulations are performed on a Beowulf cluster. When parallel processing is employed, the message passing interface is employed to transmit information between nodes. Furthermore, natural parameter continuation is implemented to achieve convergence. The computation time of each simulation varies between several hours and several days, depending on the difficulty of the problem and the initial guess.

3. Results and Discussion

3.1. Numerical Validation

In order to ensure that CFD models accurately predict the phenomena of interest, the validation of models against well-defined experimental data is essential. The numerical model is validated by comparing the results with the experimental data of Dogwiler et al. (1998). Direct comparisons between simulation and experiment allow for an assessment of the CFD model used herein. The methane-air mixtures with the inlet equivalence ratios of $\phi = 0.37$, 0.37, and 0.31 are chosen, corresponding to three catalytic cases further denoted as (A), (B), and (C) of the experimental data, respectively. These catalytic cases with inlet equivalence ratios ($\phi$), inlet velocities ($u_{in}$), and inlet temperatures ($T_{in}$) are described as follows: case (A), $T_{in} = 750$ K, $u_{in} = 1.0$ m/s, $\phi = 0.37$; case (B), $T_{in} = 729$ K, $u_{in} = 2.0$ m/s, $\phi = 0.37$; and case (C), $T_{in} = 754$ K, $u_{in} = 1.0$ m/s, $\phi = 0.31$. The Reynolds number is approximately 186 for cases (A) and (C) and 390 for case (B). The gap size and combustor length are 7 mm and 250 mm, respectively. The measured catalyst temperature distributions serve as the energy boundary conditions at the gas-wall interface. Figure 3 shows the OH concentration profiles along the fluid centerline after homogeneous ignition.

![Fig. 3. OH concentration profiles along the fluid centerline after homogeneous ignition.](image-url)
curves represent the measured OH concentrations, and the dash-color lines represent the predicted OH concentrations. The predicted profiles have been shifted axially to match the measured peak OH positions. The sharp rise in OH concentration along the channel denotes the onset of homogeneous ignition. Good agreement between predicted and measured profiles is observed, since the difference between them is generally less than 20%. On the other hand, the measured positions of homogeneous ignition for cases (A), (B), and (C) are 78, 148, and 83 mm, respectively. The predicted positions of homogeneous ignition are 66, 136, and 70 mm, respectively. Good agreement between predicted and measured ignition positions is also obtained, since the difference between them is within 16% in all the cases studied. Overall, the numerical results, such as OH concentration profiles and ignition positions, are in good agreement with the experimental data. This confirms the reasonable accuracy of the numerical model implemented in this study.

3.2. Contour Plots

Fig. 4. Contour plots of the temperature and mass fraction for a typical set of operating parameters.

Figure 4 shows the contour plots of the temperature and mass fraction for a typical set of operating parameters. The entire micro-channel is presented. Self-sustained combustion can be found within
the system. The exothermic reaction starts at the surfaces of the catalyst and travels towards the fluid centerline. Catalytic reaction takes place very rapidly, combusting most of the fuel in a relatively small region. A significant temperature rise is observed due to the exothermicity of the hetero-/homogeneous combustion, and complete fuel conversion is obtained. Despite the small scales, there exist significant gradients in temperature and species concentration within the fluid near the reaction region. These gradients necessitate the use of a two-dimensional CFD model, since axial diffusion of energy and species cannot be ignored. There are axial gradients in temperature within the walls. Despite the fluid transverse gradients in temperature and species concentration, there are no large transverse gradients within the walls themselves in all cases studied, which can be attributed to the short time-scale for heat conduction across the wall and their large aspect ratio.

3.3. Temperature Profiles

Figure 5 shows the temperature profiles along the fluid centerline and the wall. Three regions can be observed, namely preheating, combustion, and post-combustion or cooling. The width of these regions is highly dependent on operating conditions. In the preheating region, the wall temperature is much higher than that of the fluid, and thus the thermal energy is transferred from the wall to the fluid. The fluid thermal conductivity is significantly lower than that of the wall. Within the walls, therefore, most of the upstream conductive heat transfer occurs. This thermal energy is brought upstream from the post-combustion region in which the wall temperature is considerably high to the cold incoming reactants. Ignition occurs near the catalytic walls and self-sustained combustion can be found at the fluid centerline, as the fluid mixture warms up from the wall towards the fluid centerline. This is consistent with the combustion stability studies carried out in one-dimensional stagnation flows (Karadeniz et al., 2013). However, this catalytic ignition mode is quite different from the case in which outside preheating is employed to ignite the gas mixture. When preheating is employed, ignition occurs at the fluid centerline (Jiménez and Kurdyumov, 2017). Once the fluid reaches the ignition temperature, there is an inflection point in the fluid temperature profile. The reactants are consumed rapidly, releasing heat and then causing a sharp rise in the fluid temperature within the system. This “take-off” point is consistent in temperature with the
experimental findings of Nakamura and Hasegawa (2017). Due to the upstream heat transfer through the walls to the cold incoming reactants, the wall temperature in the preheating region reaches the ignition temperature of approximately 880 K to allow ignition of the fluid in the combustion region. Self-sustained combustion occurs in a relatively narrow region, which is a characteristic of highly activated reactions (Stazio et al., 2016). Despite the small scales of these systems, the rate of heat release is much faster than the transverse heat transfer within the fluid so that the fluid centerline temperature in this region is extremely high, which approaches approximately the adiabatic flame temperature in some cases.

In the post-combustion region, the reaction is completed as the reactants have been consumed. The fluid cools down to the wall temperature, and the walls are cooled by external heat losses. There exist no significant axial or transverse gradients in temperature and species concentration within this region. Both the fluid and the wall would eventually be cooled to the ambient temperature in non-adiabatic cases, if the channel is sufficiently long. In certain cases, the maximum fluid temperature is higher than the adiabatic flame temperature of methane-air mixtures, which is consistent with the flame behavior reported by Gauthier et al. (2014). Traditional analysis of plug flow reactor or continuous stirred tank reactor suggests that this is not possible to achieve with detailed chemistry and transport (Lignola and Maio, 1990). In this CFD model, however, the walls act as a conduit for upstream heat transfer through the walls from the hot exiting products to the cold incoming reactants. This causes a heat recuperation within the system, resulting in an increase in temperature near the entrance, which allows a maximum temperature that is higher than the adiabatic flame temperature. In these cases, however, the exiting fluid temperature is lower than the adiabatic flame temperature for adiabatic walls, which can be attributed to the overall energy conservation within the system.

### 3.4. Effect of Wall Thermal Conductivity

![Fig. 6. Effect of wall thermal conductivity on the flame location.](image-url)
Figure 6 shows the effect of wall thermal conductivity on the flame location. The wall thermal conductivity is taken as an independent parameter to understand how important thermal management is. The wall plays an important role in overall heat transfer. On one hand, the wall allows the upstream heat transfer, which provides a route for preheating that is necessary for ignition and combustion stability. On the other hand, the wall provides another route for exterior heat losses, which can delay ignition and eventually cause extinction. Since the walls are responsible for the external heat losses as well as the majority of the upstream preheating, the wall thermal conductivity plays an important role in determining combustion stability (Norton and Vlachos, 2003). Low wall thermal conductivity inhibits the upstream heat transfer through the wall, and thus the preheating of the feed is limited, inhibiting the initialization of hetero-/homogeneous combustion, and causing blowout. Additionally, hot spots of high temperatures within the walls may occur, which could damage the catalyst. In contrast, high thermal conductivity walls result in lower operating temperatures, and the isothermality of the system can be expected. However, high thermal conductivity walls offer a larger hot area for external heat losses and become susceptible to spatially global-like extinction (Norton and Vlachos, 2004). To quantitatively differentiate between the different regions and monitor the possible blowout behavior, the flame location is defined as the axial position with the greatest reaction rate (Leach et al., 2006). The flame location in all cases occurs on the fluid centerline. The flow velocity plays an important role in determining the location of the flame in micro-channels. This is consistent with the experimental data (Federici et al., 2009). For high flow velocities, the flame location moves downstream with the increase of flow velocity. This is because of the shorter residence times, i.e., the decrease in the convective time-scales. For low flow velocities, a sharp shift of the reaction region downstream occurs with the decrease of flow velocity. This is because of the decrease in the heat generation rate. The heat generation rate in this case is still much faster than the external heat loss rate, causing a reduced upstream heat-transfer rate. There is a minimum in the location of the flame, which can be attributed to the competition between decreased residence time and increased volumetric heat released with the increase of flow velocity. This minimum approaches the unconfined flame speed for the same feed composition, experimentally determined by Vagelopoulos and Egolfopoulos (1994). The minimum moves slightly toward higher flow velocities for higher wall thermal conductivities, as the latter allow greater upstream heat transfer to compete with the faster convective flow.

3.5. Effect of Channel Dimension

Figure 7 shows the effect of channel dimension on the flame location. When the primary variable is the wall thermal conductivity, the gap distance and wall thickness play an important role in combustion stability also when the flow velocity varies. The blowout velocity decreases with the increase of gap distance. The decrease in combustion stability with respect to flow velocity is mainly because of the reduced time-scales for energy diffusion between the walls and the fluid, which limits the preheating of the feed, and thus shifts the flame location downstream. Conversely, the blowout velocity increases with the increase of wall thickness, while leaving unaffected the combustion stability for slow flows. The increase in combustion stability is mainly because of the increased area for heat flux and upstream preheating rate to the cold incoming reactants. When flow velocities greater than the unconfined flame speed are required, the gap distance must be small, and the wall thickness and thermal conductivity should be sufficiently high to provide effective heat recuperation from the post-combustion region to preheat the cold incoming reactants.
3.6. Stability Diagram

Figure 8 shows the stability diagram in terms of the critical flow velocity as a function of the wall thermal conductivity. The upper curve represents the high-velocity limits, leading to blowout because of the reduced convective time-scales. The lower curve represents the low-velocity limits, leading to the loss of combustion stability because of the insufficient heat generation. Between these curves, self-sustained operation is allowed, whereas outside the regime, stabilized combustion is impossible. The horizontal dashed line represents the experimentally determined laminar flame speed for a stoichiometric methane-air mixture (Vagelopoulos and Egolfopoulos, 1990).
The laminar flame speed is an important property of a reacting mixture (Bradley et al., 1998). Furthermore, the accurate knowledge of laminar flame speed is essential for validation of reaction mechanisms, CFD modeling of combustion, and reactor design (Clavin, 1985). The phenomena of flame extinction, stabilization, and blowout can be described by using the laminar flame speed (Law and Sung, 2000). Lower wall thermal conductivities allow self-sustained operation for lower flow velocities, and the latter require less upstream heat transfer and more robust against external heat losses. In contrast, higher wall thermal conductivities cause maximum allowable flow velocities. However, the increased external heat losses prohibit low flow velocities. This relationship becomes significant when designing micro-devices. When a high-power system is desired, highly conductive materials should be preferred. In contrast, when a low-power system is desired, highly insulating materials should be favored to minimize external heat losses. On the other hand, the moderate wall thermal conductivity is essential for transferring heat upstream to cause ignition and stabilize the flame within the system. For systems with a high wall thermal conductivity, the wall thermal conductivity plays a minimal role in flame stability. In contrast, for higher external heat losses, high wall thermal conductivities cause the flame location to be shifted downstream and may cause extinction. The focus here is on the combustion stability. However, other wall thermal properties, such as mechanical strength, radical sticking, and allowable operating temperatures, along with the system efficiency, must be considered when designing device dimensions and choosing a material for construction. Note that complete fuel conversion can be found herein for a stoichiometric methane-air mixture using detailed chemistry and transport.

3.7. Effect of Reynolds Number

![Graph: Effect of Reynolds number on the minimum allowable equivalence ratio.](image)

**Fig. 9.** Effect of Reynolds number on the minimum allowable equivalence ratio. The Reynolds number is computed using the gap distance, velocity, density, and kinematic viscosity at the inlet. In fluid mechanics, the Reynolds number is an important
dimensionless quantity, which can be used to help predict flow patterns in different fluid flow situations (Wu et al., 2016). The Reynolds number is a criterion of whether fluid flow is absolutely steady or on the average steady with small unsteady fluctuations (Kim et al., 1987). The ability of the system to operate under fuel-lean conditions can be expected to be beneficial, since it could reduce unwanted combustion products such as coke, nitric oxide, and carbon monoxide (Hernández-Pérez et al., 2015). It could also reduce the operating temperature, which in turn could increase the lifetime of the system (Okuno et al., 2017). There are two regimes of low and high Reynolds number. For high Reynolds number, blowout occurs due to the decreased convective time-scale. On the other hand, for low Reynolds number, combustion stability is lost with the decrease of Reynolds number due to the reduced heat generation rate. At the transition between these low and high Reynolds number regimes, there appears to be a deep minimum in the fuel-lean operation limit and possibly a turning point over a narrow regime of Reynolds number. In order to fully characterize this situation, however, arc-length continuation is necessary.

Experimental studies using a heat-recirculating combustion system have found that there is an optimum Reynolds number at a relatively shallow minimum fuel-air equivalence ratio (Ahn et al., 2005). The optimum Reynolds number and the minimum equivalence ratio are generally consistent with the findings in this study. The differences between these computational findings and the experimental results perhaps due to the enhanced insulation and preheating that are achieved with the heat-recirculating design and the gap distance, which is considerably larger in the experiments (Rana et al., 2017). In order to fully understand these differences, more studies are required. One interesting topic includes how the operation and wall thermal properties affect combustion stability and the self-sustained combustion envelope through the change in reactivity and the Lewis number effect (Yan et al., 2016). Another interesting topic includes how the choice of combustor geometry affects the design and operation of micro-power generation systems (Zhang et al., 2017).

4. Conclusions

The combustion characteristics and stability of methane-air mixtures in catalytic micro-channels are studied using a two-dimensional CFD model. The effect of flow velocity on the combustion stability is evaluated to analyze the characteristics of the fluid mechanics. The minimum allowable equivalence ratio for the system is determined by the analysis of Reynolds number. It is shown that the catalytic combustion of methane-air mixtures can be stabilized in narrow channels but very careful design is necessary. Despite the small characteristic length scales of these systems, large axial gradients in temperature may exist in the walls as well as large transverse gradients in temperature and species concentration exist in the fluid. The wall thickness and thermal conductivity are important parameters in design, since they strongly affect the combustion characteristics. The flow velocity plays an important role in determining the flame location. It also plays a competing role in combustion stability. Low flow velocities reduce the power generation. In contrast, high flow velocities reduce the convective time-scale below that of the upstream heat transfer through the walls. Consequently, there exists only a relatively narrow regime of flow velocities within which self-sustained operation is allowed. A high-power system would favor highly conductive materials. In contrast, when a low-power system is being designed, highly insulating materials should be favored to minimize external heat losses. However, other wall thermal properties and the system efficiency must be considered in design. The optimum Reynolds number is determined to gain insight into the combustion characteristics of the system.
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