

A NOVEL HYBRID CONFIGURATION FOR CH₄ COMBUSTION IN A PEROVSKITE-BASED MONOLITH

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Abstract

Catalytic combustion (CC) is a promising technique for high-efficiency clean combustion. Nevertheless, its practical application has been limited owing to low thermal resistance of the catalytic materials. Thus, hybrid configurations have been proposed in order to overcome these limitations. In this work, we numerically explored the behaviour of a novel hybrid configuration for CH₄ combustion, in which the monolithic honeycomb is partially coated with a perovskite catalyst as the active phase. In particular, the central channels are uncoated, while the catalyst is confined into the external channels of a non-adiabatic 10x10 honeycomb. CFD simulations showed that, at steady state, complete fuel conversion is obtained even if the 60% of the monolith is uncoated (further reduction in the catalyst presence does not allow fuel ignition), thus highlighting the crucial role of homogeneous reactions in both coated and uncoated channels. In particular, homogeneous ignition in the uncoated channels is due to the axial heat transfer from the catalytic (external) to the non-catalytic (internal) channels. Moreover, hybrid configurations show more pronounced thermal profiles, allowing to reduce the peak temperature of the catalytic channel with respect to a fully coated configuration.

Introduction

In view of its ability to burn different fuels beyond their flammability limits with high efficiency even at relatively low temperatures, thus producing extremely low levels of pollutants (NO_x, CO and UHC), catalytic combustion (CC) is a promising technique for high-efficiency clean combustion¹. Even though in practical applications catalytic combustors operate at high pressure (up to 20 bar), most of the literature studies have been performed at atmospheric pressure. However, pressure affects several phenomena occurring during catalytic combustion (mass and heat transfer, surface and homogeneous reactions).

Under high pressure conditions, homogeneous gas-phase chemistry is coupled with heterogeneous surface-phase chemistry for different fuels, such as methane, hydrogen and CO/H₂ mixtures²⁻⁴.

It is worth underlying that the most part of the above studies have been focused on noble metals as active phases. From a general point of view, noble metal catalysts

show very high activity, but exhibit high costs and tendency to volatilize and/or sinter at high temperature⁵. A possible alternative to noble metals for methane combustion is represented by perovskites⁶. Dispersion of the active phases on high surface area supports like La-stabilized alumina or La-stabilized MgO or spinels increases activity and thermal stability⁶. Very recently, we reported experimental and numerical results for methane catalytic combustion over a 20 wt LaMnO₃/La- γ -Al₂O₃ monolith under conditions relevant to small size GT applications (pressure up to 12 bar) showing that, as pressure increases, the catalyst allows ignition of the homogeneous reaction which guarantees the complete fuel conversion². We reported the numerical results of a single channel CFD model showing that the complete methane conversion is attained by the synergic coupling of catalytic and homogeneous reactions². The catalytic reaction has been proven to promote ignition and high wall temperatures. In this work, a multichannel (10 x 10 channel) configuration CFD model is developed in order to identify an optimal reactor configuration in terms of number of catalyst-coated channels.

CFD Model

A two-dimensional CFD model was developed to simulate the coupling of the fluid flow and the chemical processes at the gas-solid interface and in the gas phase for lean CH₄/O₂/N₂ combustion over a 20 wt% LaMnO₃/La- γ -Al₂O₃ catalyst supported on 900 cpsi honeycomb cordierite monolith (NGK) in a shape of cylinder (length, 50 mm; diameter, 11 mm). A section of the whole catalytic monolith was simulated exploiting its symmetries. Figure 2 shows the scheme of the computational domain. Each channel consists of two parallel (infinitely wide) plates.

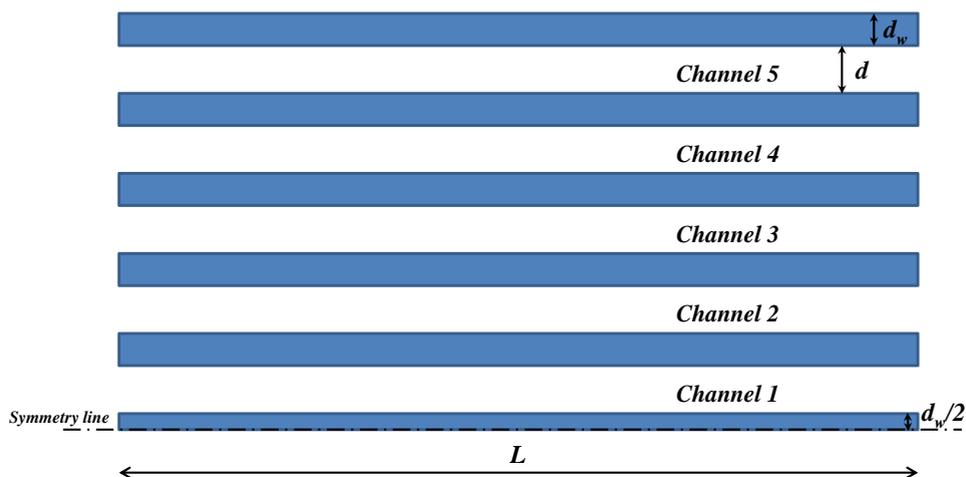


Figure 2. Scheme of the computational domain (not to scale).

The model solves the mass, momentum, chemical species and energy conservation equations in the fluid (coupled to the ideal-gas equation), along with the energy

equation in the solid wall. At the inlet of the channel, fixed flat profiles were assumed for velocity, species and temperature. At the exit, the static pressure was imposed and far-field conditions were specified for the remaining variables. At the fluid-wall interface, a no-slip boundary condition was assigned; heat losses from the ends of the channels were not considered (insulated ends), while Newton's law of convection was used at the outer surface of the external channels.

The reaction rate for homogenous methane combustion was calculated according to the single-step reaction rate by Westbrook and Dryer⁷. The catalytic reaction rate used in this work is a single rate equation and was obtained from an independent experimental campaign on the $\text{LaMnO}_3/\text{La-}\gamma\text{Al}_2\text{O}_3$ catalyst performed in the pressure range 1-12 bar⁸. The spatial discretization of the model equations used second order schemes for all terms. Computations were performed by means of the segregated solver of the ANSYS FLUENT code (release 13.0) that adopts the SIMPLE method for treating the pressure-velocity coupling. All residuals were always smaller than $1.0 \text{ E-}7$.

Main results

In figure 1 (top), the methane conversion obtained at $P = 9$ bar and $Q = 88$ slph is shown as a function of the number of catalyst-coated channels. The complete conversion is obtained in all the monolith configurations, except the one in which only the two central channels are coated by the catalyst, suggesting that, in this case, the catalyst is not sufficient to sustain the homogeneous reaction in the outer channels.

The temperature maps of the fully coated configuration (not reported) show that two main profiles maybe distinguished in the whole reactor: a profile at the channel level, close to the channel entrance, in which reaction is localized, and a profile at the reactor level (macro-profile) which is characterized by significant radial and axial temperature gradients involving the whole monolith. Furthermore, the central channel feels the non adiabaticity of the reactor only in proximity of the outlet section, while the external channels are susceptible to heat losses also in the reaction zone, close to the channel entrance. The heat losses through the external channels weak the homogeneous reaction front with respect to the central channel. However, at these conditions, the complete conversion is attained in all reactor channels. The profiles of the homogeneous reaction and the methane molar fraction (not reported) confirm that the complete methane conversion is attained through the homogeneous reaction front. This result is in agreement with our previous findings² showing that the role of homogeneous reaction is crucial in determining complete methane conversion at all the operating pressures investigated. Indeed, the catalytic reaction has been proven to promote ignition and high wall temperatures, thus initiating and sustaining the homogeneous reaction front which completes the methane conversion.

The bulk gas temperature (figure 1, bottom) decreases as the number of catalyst-coated channels decreases.

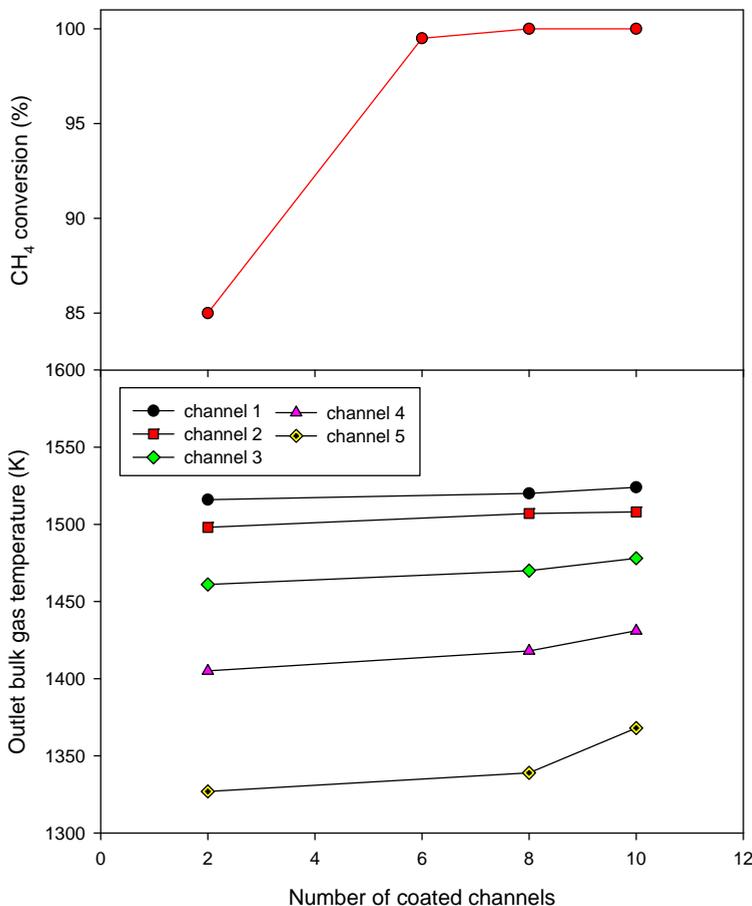


Figure 1. Computed CH₄ conversion (top) and outlet bulk gas temperature in each channel (bottom) versus the number of catalyst-coated channels: P = 9 bar; Q = 88 slph. Channels 1-5 can be identified in Figure 2.

The hybrid configurations show more pronounced thermal profiles (not reported), allowing to reduce the peak temperature of the catalytic channels with respect to a fully coated configuration. The partial coating then allows the control of the temperature level at the catalytic walls.

Conclusions

Numerical results have shown that, at steady state conditions, the catalyst is not required over the entire monolith, being sufficient only over the walls of the outer perimeter channels. As a matter of fact, in the uncoated central region, methane conversion by homogeneous reaction is made possible by the radial heat transfer from the external catalytic channels through the monolith walls.

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