Three-dimensional direct numerical simulations of turbulent fuel-lean H₂/air hetero-/homogeneous combustion over Pt with detailed chemistry

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Abstract

Three-dimensional direct numerical simulations (DNS) with detailed heterogeneous and homogeneous chemistry and transport were carried out to investigate the turbulent combustion of fuel-lean hydrogen/air mixtures (equivalence ratios $\varphi = 0.16$ and 0.18) in a platinum-coated channel with prescribed wall temperatures of 1250 K and 1270 K and an incoming bulk Reynolds number of 5700. Over the channel domain where only catalytic reactions were present, temporally and spatially segregated islands with increased concentration of the limiting hydrogen reactant formed on the walls, a manifestation of finite-rate surface chemistry effects. The formation of such islands correlated with high streamwise vorticity, which was in turn responsible for an increased turbulent mass transport towards the catalyst. Fluctuations of either gaseous or surface species were significant (up to 34%) at the upstream channel locations and dropped farther downstream due to the flow laminarization induced by the heat transfer from the hot walls and the growth of the turbulent boundary layer. Such strong fluctuations exemplify the necessity of appropriate catalytic chemistry turbulence closures similar to those used for gas-phase reactions. Homogeneous ignition exhibited streamwise fluctuations with a standard deviation of three channel half-heights. Downstream of homogeneous ignition, gas-phase combustion was concentrated in elongated streamwise stripes confined close to the walls. Hydrogen was incompletely converted within the gaseous combustion zones, and the leaking fuel reacted on the catalytic walls leading to combined hetero-/homogeneous combustion over the entire post-ignition domain. Local extinction of gaseous combustion was finally observed in spatially isolated islands characterized by high streamwise vorticity. The increased turbulent mass transport towards the walls was hence responsible for the local flame extinction, which in turn led to an increased catalytic conversion at the extinguished flame locations.

Keywords

Turbulent combustion of hydrogen over platinum; 3-D DNS with detailed hetero-/homogeneous chemistry; homogeneous ignition; flame anchoring in turbulent catalytic channels; local flame extinction

1. Introduction

Catalytic combustion is particularly attractive for the latest power generation technologies aiming at mitigating greenhouse CO₂ emissions. For example, post-combustion CO₂ capture in gas turbines can be accomplished by employing large flue gas recycle (FGR) in the reactant stream, so as to increase the CO₂ content in the exhaust gas and thus facilitate its subsequent capture [1]. The reduced reactivity of the heavily FGR-diluted reactants favors the use of catalytic combustion methodologies due to their lower activation energy barrier and the resulting increased combustion stability [2]. Catalytic reactors are made of honeycomb monolithic structures comprising a multitude of channels (each with a hydraulic diameter of 1 to 2 mm), which are typically coated with Pt or Pd. Under the high pressures of modern turbines (up to 30 bar) the incoming Reynolds numbers, based on the individual catalytic channel hydraulic diameter, can reach at full load 20,000 [3]. Moreover, gas-phase chemical reactions cannot be neglected at the elevated pressures of turbines –even at the tight geometrical confinements of typical catalytic reactors [4, 5]. Reliable numerical models for hetero-/homogeneous combustion in transitional or fully turbulent channel-flows are therefore essential for reactor design and development.

Earlier modeling approaches in turbulent catalytic combustion were based on simple turbulence closures stemming from isothermal flows. The first detailed hetero-/homogeneous turbulent combustion models were subsequently developed using Low-Reynolds number (LR) submodels within the Reynolds Averaging Navier Stokes (RANS) description along with appropriate surface/gas-phase chemistry closures [6, 7]. Comparisons with in situ laser-based measurements (1-D Raman of major species concentrations across the channel boundary layer and planar Laser Induced Fluorescence (LIF) of the OH radical) have shown [7] that the heterogeneous and homogeneous combustion processes crucially depended on the turbulent flow laminarization induced by the heat transfer from the hot catalytic walls. Three-dimensional Direct Numerical Simulation (DNS) of turbulent channel-flow combustion of hydrogen/air mixtures over platinum was reported

only recently [8] using a one-step catalytic reaction (gas-phase chemistry was not included). This study quantified the effect of flow laminarization on the underlying combustion processes, demonstrated that the fluctuations of the surface reaction rates had temporal frequencies comparable to the thermal response frequencies of typical solid materials used in catalytic reactors, and assessed limitations of RANS models.

The present work undertakes the first 3-D DNS of turbulent catalytic combustion with detailed heterogeneous and homogeneous chemistry and transport. Simulations are performed in a 5-mm-height planar channel coated with platinum, at an incoming Reynolds number of 5,700 (based on the channel height and the mean inlet velocity), wall temperatures of 1250 K and 1270 K (necessary to achieve gas-phase ignition and which fall in the upper thermal stability limit of practical catalytic reactors used in gas turbines) and fuel-lean H_2 /air premixtures (equivalence ratios $\phi = 0.16$ and 0.18).

The goal is to investigate the impact of turbulence on the catalytic combustion processes, to expose the interactions between turbulence and chemistry leading to the onset of homogeneous ignition, and finally to identify the conditions for local flame extinction. To this direction, equivalence ratios are selected in such a way that they allow for appreciable homogeneous ignition distances, and also for the occurrence of local flame extinction at the selected bulk Reynolds number.

2. Numerical setup

A highly-efficient parallel code based on the spectral element solver Nek5000 [9] is employed to integrate the conservation equations in their low Mach number formulation. The computational domain is split into hexahedral elements, and N th-order Legendre-Lagrangian polynomials are used to express the solution in each spatial direction [10]. The discretized equations are integrated in time with a high-order splitting scheme [11], whereby the continuity and

momentum equations are integrated with a semi-implicit scheme and the thermochemistry is solved by the implicit stiff ODE integrator CVODE [12]. The massive computational cost of integrating stiff ODEs (the large stiffness primarily originates from the surface reaction mechanism, as discussed in [13]) necessitates the use of an efficient adaptive preconditioner, which is constructed using the diagonal of the Jacobian of the surface and gas-phase reactions source terms.

Detailed heterogeneous (catalytic) and homogeneous (gas-phase) chemical reaction mechanisms are employed. For gas-phase chemistry, the elementary H_2/O_2 mechanism of Burke et al. [14] (9 gaseous species and 21 reversible reactions) is used, while the catalytic oxidation of hydrogen on Pt is described by the detailed (mean-field) scheme of Deutschmann et al. [15] (6 gas-phase and 5 surface species, 14 reactions, surface site density $\Gamma = 2.72 \times 10^{-9} \text{ mol/cm}^2$). Validation of the aforementioned hetero-/homogeneous reaction mechanisms has been reported in laminar channel-flow experiments by Ghermay et al. [16]. Gaseous and surface reaction rates are evaluated using Chemkin [17, 18] while a mixture-average transport model is adopted along with the Chemkin transport database [19].

The computational domain comprises a 3-D channel with full-height $H = 2h = 5 \text{ mm}$, width $W = 3h = 7.5 \text{ mm}$ and length $L = 48h = 120 \text{ mm}$ (Fig. 1). The domain is split into a recycling (L_R) and a combustion section, with lengths $12h$ and $36h$, respectively. As in our earlier work [8], in the recycling section the velocity field at $z = 12h$ is copied to the inlet plane $z = 0$ in order to generate a fully-developed turbulent channel flow at the beginning of the reacting section. The friction Reynolds number is $Re_\tau = 182$ based on the channel half-height h and the friction velocity $u_\tau = U_{\text{IN}}\sqrt{C_f/2}$, where the mean inlet velocity is $U_{\text{IN}} = 22.4 \text{ m/s}$ and $C_f = 0.073 Re^{-0.25}$ [20]. Uniform inlet temperature $T_{\text{IN}} = 320 \text{ K}$ and flux boundary conditions for the species are imposed at the inflow. Hydrogen/air premixtures at atmospheric pressure are admitted in the channel, with fuel-to-air equivalence ratios $\phi = 0.16$ and 0.18 (denoted henceforth as Cases I and II, respectively). The upper and lower channel walls are coated with Pt for $z \geq 14h$, leading to a

catalytically-active length $L_{CATAL} = 34h$ (Fig. 1). The wall temperatures are ramped with a hyperbolic tangent function $T_w = T_C + 0.5(T_H - T_C)(1.0 + \tanh(z - 14h)/0.4h))$ from an initial temperature $T_C = T_{IN}$ to a final $T_H = 1250$ K for Case I and $T_H = 1270$ K for Case II. This defines a ramping length $13h \leq z \leq 15h$ (encompassing 0.7% to 99.3% of the temperature rise, see Fig. 1). At the outlet and in the spanwise x -direction, zero-Neumann and periodic boundary conditions are imposed, respectively. The gas-phase species boundary conditions at the gas-wall interface are:

$$\vec{n} \cdot [\rho Y_i (\vec{V}_i + \vec{u}_s)]_w = \dot{s}_i, \quad i = 1, 2, \dots, N_g, \quad (1)$$

where ρ is the gas density, while Y_i , $\vec{V}_i$, $\dot{s}_i$, and N_g are the mass fraction, diffusion velocity vector, mass heterogeneous production rate of the i -th gaseous species, and the number of gaseous species, respectively. The unit outward-pointing vector normal to the catalytic surface is $\vec{n}$ and $\vec{u}_s$ is the Stephan velocity ($\vec{n} \cdot \vec{u}_s = (1/\rho) \sum_{i=1}^{N_g} \dot{s}_i W_i$ [18]). The time-dependent governing equations for the gas-phase have been reported in [21].

The domain is discretized with 20 uniform elements in the spanwise x -direction, and 20 elements clustered close to walls in the y -direction with the first point near the wall located at $y^+ = 0.146$ wall units. In the streamwise z -direction, 20 elements are allocated to the recycling section with uniform distribution, while 180 uniform elements are distributed in the reacting section in order to resolve the steep thermoscalars gradients. By using seventh-order interpolating polynomials in each spatial direction and element, the computational domain is discretized by a total of 27,440,000 grid points. The resolution in the most demanding y -direction ranges from 4 to 10 μm over the transverse distance where the flames are typically located.

Variables are non-dimensionalized using as reference values the mean inlet velocity $U_{ref} = U_{IN} = 22.4$ m/s, the channel half-height $L_{ref} = h = 2.5$ mm, and the inlet temperature $T_{ref} =$

$T_{IN} = 320$ K. In the non-dimensional coordinate system, the walls are located at $y = \pm 1$, the spanwise bounds are $x = \pm 1.5$, and $z = 0$ refers to the beginning of the recycling section.

3. Result and discussion

Time integration is initially carried out to reach a statistically-steady reactive flow. At longer times and within the statistically-steady operation, only catalytic combustion is present in Case I (homogeneous chemistry is ignited during the transient period, but eventually is blown out at long times) whereas combined hetero-/homogeneous combustion occurs in Case II. The heterogeneous combustion processes are discussed first, the conditions leading to homogeneous ignition follow, and finally the turbulence and hetero-/homogeneous chemistry interactions are elaborated downstream of homogeneous ignition. All statistical quantities presented henceforth have been averaged over both time and spanwise x -direction.

3.1. Heterogeneous combustion

The reactor length with only catalytic reactions (without contribution of gas-phase chemistry) is herein considered, encompassing the lengths $14 \leq z \leq 48$ and $14 \leq z \leq 29$ for Cases I and II, respectively. Typical two-dimensional distributions of the instantaneous hydrogen concentration and of the quasi-streamwise vortical structures are illustrated in Fig. 2. In particular, color maps of the ratio of hydrogen mass fraction at the upper wall ($y = 1$) to the inlet mass fraction ($Y_{H_2,w}/Y_{H_2,IN}$) are depicted, with superposed isolines of streamwise vorticity $\omega_z = -0.75$ (black lines) and $\omega_z = 0.75$ (white lines) on the zx -plane and at $y = 0.9$. The ratio $Y_{H_2,w}/Y_{H_2,IN}$ is a measure of the degree of finite-rate chemistry. For infinitely-fast chemistry (transport-limited hydrogen conversion) this ratio is identically zero on both catalytic walls; the higher this ratio, the higher the degree of finite-rate chemistry. Although a true measure of finite-rate chemistry is the wall-to-center ratio $Y_{H_2,w}/Y_{H_2}|_{y=0}$,

the center plane hydrogen mass fraction drops weakly over $14 \leq z \leq 34$ from $Y_{\text{H}_2,\text{IN}}$ to about $0.97Y_{\text{H}_2,\text{IN}}$ such that $Y_{\text{H}_2}|_{y=0} \approx Y_{\text{H}_2,\text{IN}}$.

The decrease in the occurrence frequency of the vortical structures along the streamwise z -direction is evident in Fig. 2 (see black and white isolines). This is an outcome of the flow laminarization due to heat transfer from the hot walls that dampens the vortical structures, such that they eventually almost vanish at the channel outflow. For Case II with higher wall temperature, the effect of flow laminarization is stronger (vortical structures persist farther downstream in Fig. 2a rather than in Fig. 2b). The results in Fig. 2 are qualitatively similar to those reported in our earlier purely heterogeneous DNS investigation with one-step catalytic chemistry [8], however, here the flow laminarization is stronger due to the substantially higher wall temperatures (1250 or 1270 K as opposed to 960 K in [8]).

A correlation between counter-rotating vortices and regions of high $Y_{\text{H}_2,\text{w}}$ is readily seen in Fig. 2. The quasi-streamwise vortices and turbulent structures bring fresh mixture from the channel core towards the wall, and the concentration of hydrogen near the wall becomes appreciably higher than zero. The hydrogen mass fraction at the wall reaches its highest value at the beginning of the platinum-coated section ($z = 14$) and drops rapidly to a few percent of $Y_{\text{H}_2,\text{IN}}$ farther downstream, despite the isolated “islands” of high hydrogen concentration at various spanwise x -locations seen in Fig. 2. This indicates that catalytic reactions are, over most of the catalytic surface area, fast compared to the turbulent transport and that hydrogen conversion is not far from the transport limit. There exist however isolated local zones with significantly higher values of $Y_{\text{H}_2,\text{w}}/Y_{\text{H}_2,\text{IN}}$ (5% to 8%) at $z > 20$ (reddish/greenish islands in Fig. 2) typically associated with the high vorticity indicated by the black/white isolines. The formation of such islands is a clear manifestation of finite-rate surface chemistry, indicating that the catalytic reactions are not fast enough to cope with the locally and instantaneously high turbulent transport towards the wall.

Probability density functions (PDFs) of $Y_{H_2,w}/Y_{H_2,IN}$ are presented in Fig. 3. In the channel section with only catalytic conversion, the growth of the species boundary layer and the flow-laminarization weaken the turbulent transport, resulting in a shift of the means μ of the PDFs to lower values with increasing z -distance. Moreover, as high H_2 -concentration islands at the walls are suppressed along the streamwise direction, the PDF standard deviations σ decrease with increasing z . The comparison of mean values at the same streamwise position shows that the averages of $Y_{H_2,w}/Y_{H_2,IN}$ are lower for Case II compared to Case I due to the higher wall temperature and equivalence ratio of the former case, which lead to stronger flow laminarization and faster catalytic chemistry, respectively.

Figure 4a provides the instantaneous streamwise profiles of surface coverage for $H(s)$, $Pt(s)$, $OH(s)$, and $O(s)$ at the spanwise location $x = -0.7$ marked in Fig. 2a. Regions of high $Y_{H_2,w}$ are associated with higher $H(s)$, $Pt(s)$ and $OH(s)$, and lower $O(s)$ surface coverage, as indicated by the two vertical dashed lines in Fig. 4a corresponding to the center of two hydrogen islands in Fig. 2a. Figure 4b provides PDFs of $H(s)$, $Pt(s)$, $OH(s)$ and $O(s)$ at $z = 20$. The broad PDFs in Figs. 3 and 4b have implications for turbulence modeling. A laminar-like closure for surface reactions was investigated in [7], considering that the major non-linearity due to the Arrhenius exponential is removed for prescribed surface temperature (all reaction-rate coefficients k are then constant). For example, the average adsorption rate of the i -th gaseous species is $\overline{\dot{s}_{i,ads}} = k_{ads,i} \overline{[C_i]^a} \overline{[\Gamma\theta_{Pt}]^b}$ (overbars denote averaging, $k_{ads,k}$ the adsorption rate coefficient, $[C_i]$ the instantaneous concentration of the i -th gaseous species at the gas/wall interface, $[\Gamma\theta_{Pt}]$ the surface concentration of the free platinum sites with θ_{Pt} the free-site coverage, and a and b appropriate reaction orders). In the case of weak fluctuations, this rate can be approximated as $\overline{\dot{s}_{k,ads}} \approx k_{ads,k} \overline{[C_k]^a} \times \overline{[\Gamma\theta_{Pt}]^b}$. However, as the fluctuations of both gas and surface species become appreciable, this approach is

invalidated (e.g. $\pm 2\sigma$ fluctuations for H_2 and $H(s)$ correspond to up to 34% and 20% of their mean values, respectively; see Figs. 3 and 4b).

The same also applies for reactions between surface species (e.g. $2OH(s) \rightarrow H_2O(s) + O(s)$, $OH(s) + H(s) \rightarrow H_2O(s) + Pt(s)$), given the appreciable fluctuations in $OH(s)$ and $H(s)$ in Fig. 4. Hence, a turbulent surface chemistry closure is needed similar to that used for gaseous reactions (e.g. via a presumed-shape PDF for the surface reaction rates). Such modeling is required not only for the present detailed surface chemistry but also for global catalytic steps of the type $\dot{s}_{H_2} = k[C_{H_2}]^a$, due to the non-linearity induced by the concentration exponent (for example, $a = 1.5$ for lean H_2 /air combustion over Pt [22]).

Sensitivity analysis (SA) was carried out using the surface perfectly stirred reactor (SPSR) package of Chemkin [23] to identify the limiting reaction(s) responsible for the aforementioned finite-rate surface chemistry. The surface-to-volume ratio was set to 4 cm^{-1} (as in the coated channel section of Fig. 1), while the residence time was varied from 0.1 to 1 ms to mimic the fastest flow time scales. The SA indicated that the surface and desorption reactions were always fast enough and that kinetic limitations originated from the adsorption reactions.

3.2. Homogeneous ignition

Statistically-steady homogeneous ignition is attained only for Case II. The PDF of the homogeneous ignition locations (z_{ig} , to be properly defined in Section 3.3) presented in Fig. 5 shows that about 90% of z_{ig} falls within $24.4 < z < 34.2$. The average ignition position is $\overline{z_{ig}} = 29.04$ and its standard deviation $\sigma_{z_{ig}} = 2.95$. The homogeneous ignition location computed with a laminar model (having the same uniform inlet properties as the present DNS simulations) is also indicated in Fig. 5, whereby $z_{ig,lam} = 27.2$. Since $z_{ig,lam} < \overline{z_{ig}}$, turbulence, on the average, inhibits

homogeneous ignition. This inhibition is also evident in Case I, where in the corresponding laminar simulation $z_{ig,lam} = 30.8$, while no stationary turbulent flame was established.

Figure 6 provides a scatter plot of the normalized instantaneous transverse scalar fluxes $v'T'$ and $v'Y'_{H_2}$ at the average ignition position $\overline{z_{ig}} = 29.04$ and $y = -0.9$, where “prime” refers to fluctuations above the mean value and v is the transverse velocity component. In quadrant II, which is characterized by instantaneous positive temperature fluxes and negative hydrogen mass fraction fluxes (indicating mass transport towards the wall $y = -1$ and heat transport away from the wall), homogeneous ignition is observed for the majority of events. In all other quadrants, however, ignition events are rare. This suggests that ignition is mostly associated with vortices bringing fuel towards the wall and transferring heat away from it.

It is finally pointed out that despite the spatio/temporal fluctuations of z_{ig} , the catalytic pathway still provides a viable stabilization for gaseous combustion. In non-catalytic channels, DNS has usually employed an artificial stabilization mechanism to anchor the flame (e.g. inclusion of a hot zone as in the H_2 /air DNS by Gruber et al. [24]).

3.3. Post-ignition combustion processes

Following homogeneous ignition, combined heterogeneous and homogeneous combustion is attained in Case II as will be shown next. Figure 7 provides color maps of the instantaneous homogeneous hydrogen reaction rate and mass fraction of the OH radical on the xz -plane at $y = -0.95$. “Islands” of high and low homogeneous reaction rates, elongated in the streamwise direction, are seen in Fig. 7a. The differences in instantaneous z_{ig} at different spanwise x -locations is evident, encompassing the range $27 \leq z_{ig} \leq 36$. In Fig. 7b the 2-D distribution of OH mass fraction is depicted, which is a common indicator of homogeneous combustion in experiments. Regions with high concentration of OH form elongated strips along the streamwise z -direction.

Figure 8 illustrates color maps of instantaneous gas-phase reaction rate on the yz -plane at three spanwise locations ($x = -0.5, 0, 0.5$) superimposed by vorticity isolines $\omega_z = \pm 0.5$. It is seen that gaseous combustion is confined close to the catalytic surface, as also observed in the turbulent hetero-/homogeneous combustion experiments of Appel et al. [7]. Turbulent vortical structures push the gaseous reaction zone towards the wall and eventually lead to extinction of gaseous combustion as seen by the disruption of the reaction rate contiguity in Fig. 8.

The local flame extinction and the hetero-/homogeneous chemistry interactions can also be seen in the streamwise profiles of the instantaneous gaseous (G) and catalytic (C) hydrogen conversion rates at two spanwise locations $x = \pm 0.7$ shown in Figs. 9a and 9b. The catalytic C rates refer to the lower wall ($y = -1$), while the volumetric G rates have been integrated over the channel half-height $-1 \leq y \leq 0$ so as to be directly comparable to the surface C rates. Furthermore, Fig. 9c provides a 2-D map of the gaseous fuel conversion over the lower wall, superimposed by ω_z vorticity isolines ($\omega_z = \pm 0.5$). As seen in Fig. 9a and 9b, catalytic and gas-phase combustion occur in parallel (C and G rates may coexist at a given z -position). The instantaneous and x -dependent homogeneous ignition position, z_{ig} , has been computed from plots similar to Fig. 9c. At every spanwise x -location and time, z_{ig} is defined as the far upstream streamwise distance where the local G rate reaches 5% of the peak value on the entire xz -plane in Fig. 9c.

The instantaneous catalytic conversion C in Figs. 9a and 9b, drops with increasing streamwise distance, albeit non-monotonically, due to reduced mass transport and upstream fuel depletion. Homogeneous ignition occurs at $z \approx 27$ for $x = 0.7$ and $z \approx 35$ for $x = -0.7$. Farther downstream, the G rate increases, again non-monotonically, and at some z -locations it drops and even approaches zero (local extinctions marked by arrows in Figs. 9a and 9b). These nearly zero gaseous conversion regions create streamwise-elongated stripes in Fig. 9c. There is a direct correlation between counter-rotating vortices and regions of low gaseous conversion rates. The intense turbulent vortical structures create locally and instantaneously high transverse transport

towards the wall, which in turn leads to incomplete combustion of hydrogen through the gaseous reaction zone. The leaking fuel is subsequently converted catalytically on the wall leading to coexistence of C and G in Figs. 9a and 9b. With a further rise in transverse transport, the flame extinguishes locally and hydrogen is only converted by the catalytic pathway.

The aforementioned process has a direct analogue in the combustion of fuels with Lewis numbers less than unity in laminar stagnation-point flows against catalytic walls. Law and Sivashinsky [25] reported that an increase in the strain rate pushes the flame against the catalytic wall, leading to incomplete gas-phase combustion and to a subsequent catalytic conversion of the leaked fuel, while a further increase of the strain rate leads to flame extinction. The turbulent transport in the present work plays a role analogous to that of the strain rate in laminar stagnation flows.

The frequent local flame extinctions are undesirable in practical power generation systems. This is because the catalytic combustion of the diffusionally imbalanced hydrogen fuel creates superadiabatic surface temperature at fuel-lean stoichiometries [22], while the presence of gas-phase reactions moderates this superadiabaticity by shielding the catalyst from the hydrogen-rich channel core [26]. Hence, local flame extinctions may cause detrimental hot spots on the catalyst.

The simulations exemplify the need of a turbulence closure in RANS approaches for the surface reaction rates, similar to the closure used for gas-phase reactions. Simple models using “laminar-like” closure for surface reaction rates were found inadequate. Regarding gas-phase combustion, advanced chemical closures are needed to describe the frequent local flame extinction.

4. Conclusions

The hetero-/homogeneous combustion of fuel lean H_2 /air mixtures (equivalence ratios $\varphi = 0.16$ and 0.18) over platinum was investigated in a planar channel by means of

three-dimensional direct numerical simulations with detailed catalytic and gas-phase chemistry and transport. The wall temperature was prescribed to either 1250 or 1270 K, while the incoming Reynolds number was 5700.

In the $\varphi = 0.16$ case, only catalytic combustion occurred at the statistically stationary state, while for $\varphi = 0.18$ both catalytic and gas-phase combustion were present. Over the channel lengths where only catalytic reactions were important, appreciable fluctuations of the hydrogen concentration on the wall (up to 34%) were observed, while surface species concentration fluctuations were up to 20%. Such appreciable fluctuations exemplify the importance of appropriate turbulent closures for surface chemistry. Islands of high hydrogen concentrations on the wall formed at different time instances and correlated to high local streamwise vorticity, manifesting finite-rate catalytic chemistry effects. The kinetic limitations originate from the hydrogen adsorption reaction and not from desorption or pure surface reactions.

In the case with homogeneous combustion, the streamwise ignition locations exhibited spatio-temporal fluctuations with a standard deviation of about three channel half-heights. Nonetheless, catalytic combustion provided a relatively good flame stabilization mechanism. Gaseous combustion was confined close to the catalytic walls in the form of elongated streamwise structures. Gaseous and catalytic combustion coexisted because of incomplete combustion through the gaseous reaction zones. In regions of high streamwise vorticity, the associated high turbulent transport caused local flame extinction. However, in the flame extinguished zones, hydrogen was still converted by the catalytic reaction pathway.

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Figure captions

Fig. 1. Schematic of the computational domain and wall temperature profile.

Fig. 2. Instantaneous snapshot of $Y_{H_2,w}/Y_{H_2,IN}$ on the upper wall and of the quasi-streamwise streak vorticity structures demarcated by $\omega_z = \pm 0.75$ (black and white isolines) on the zx -plane at $y = 0.9$: (a) Case I ($\phi = 0.16, T_H = 1250$ K), (b) Case II ($\phi = 0.18, T_H = 1270$ K). Yellow line at $x = -0.7$ in (a) refers to the profile of Fig. 4.

Fig. 3. PDFs of $Y_{H_2,w}/Y_{H_2,IN}$ at different streamwise positions for Cases I and II; μ denotes mean values and σ standard deviations.

Fig. 4. (a) Instantaneous surface coverage of Pt(s), OH(s), H(s), and O(s) at $x = -0.7$ (line in Fig. 2a), (b) PDFs of surface coverage of Pt(s), OH(s), H(s), and O(s) at $z = 20$ and Case I. Surface coverages of OH(s) and H(s) are multiplied by 100 and 37000, respectively.

Fig. 5. PDF of the homogeneous ignition streamwise location.

Fig. 6. Scatter plot of normalized transverse scalar fluxes of temperature and hydrogen mass fraction at $\overline{z_{ig}} = 29.04$ and $y = -0.9$.

Fig. 7. Instantaneous snapshot of: (a) the homogeneous hydrogen reaction rate [kg/m^3s], (b) Y_{OH} , on the zx -plane at $y = -0.95$.

Fig. 8. Instantaneous snapshot of the homogeneous conversion rate of H_2 [kg/m^3s] and of the quasi-streamwise streak vorticity demarcated by $\omega_z = \pm 0.5$ (black and white lines) on the yz -plane at: (a) $x = -0.5$, (b) $x = 0$, and (c) $x = 0.5$.

Fig. 9. (a, b) Instantaneous profiles of catalytic (C) and gaseous (G) fuel conversion rates on the lower wall at (a) $x = 0.7$, (b) $x = -0.7$. (c) Instantaneous map of gaseous fuel conversion rates [kg/m^2s] and of quasi-streamwise streak vorticity structures $\omega_z = \pm 0.4$ (black and white isolines) on the zx -plane at $y = 0.95$. The volumetric gaseous G rate has been over the channel half-height ($-1 \leq y \leq 0$).

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9