Turbulent catalytically stabilized combustion of hydrogen/air mixtures in entry channel flows

Christoph Appel, John Mantzaras *, Rolf Schaeren, Rolf Bombach, Andreas Inauen

Paul Scherrer Institute, Combustion Research, CH-5232 Villigen PSI, Switzerland

Received 4 May 2004; received in revised form 8 October 2004; accepted 21 October 2004
Available online 30 November 2004

Abstract

The turbulent catalytically stabilized combustion (CST) of fuel-lean hydrogen/air premixtures over platinum was investigated experimentally and numerically in the entrance region of a channel. Experiments were carried out in an optically accessible catalytic channel reactor with incoming Reynolds numbers up to $3 \times 10^4$ and involved particle image velocimetry (PIV) and laser Doppler velocimetry (LDV) for the characterization of the in-channel and the inlet flow, respectively, 1-D spontaneous Raman measurements of major species concentrations over the catalyst boundary layer for the assessment of turbulent scalar transport, and planar laser-induced fluorescence (LIF) of the OH radical for the determination of gaseous combustion. Additional PIV measurements were carried out with room-temperature air flows having Reynolds numbers comparable to those of the reacting cases. The numerical predictions included a Favre-averaged moment closure approach with different low-Reynolds number (LR) near-wall turbulence models. The reacting cases exhibited a strong relaminarization of the turbulent flow due to the intense heating from the catalytic surfaces and this was captured only by a recent LR heat-transfer model. Standard LR turbulence models performed excellently in the nonreacting flows but they overpredicted substantially the turbulence levels of the reacting cases. The suppression of turbulence led to a reduction of the catalytic hydrogen conversion and to a promotion of homogeneous ignition. Parametric numerical studies have delineated the regimes of flow laminarization in CST. It was shown that for a certain range of inlet Reynolds numbers, inlet turbulent kinetic energies and channel wall temperatures, a simpler laminar model was sufficient to capture key CST characteristics such as catalytic fuel conversion and onset of homogeneous ignition, while standard LR turbulence models overpredicted substantially the former and could not reproduce the latter.

Keywords: Turbulent catalytic combustion; Channel-flow combustion; Flow laminarization; PIV, Raman, and LIF experiments

1. Introduction

Turbulent reacting channel flows have received increased attention in numerous chemical engineering systems (such as high-velocity fluidized bed reactors), in rocket propulsion motors, in fire safety research and, more recently, in gas turbines employing the
### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$b$</td>
<td>channel half-height, Fig. 1</td>
</tr>
<tr>
<td>$c_p$</td>
<td>specific heat at constant pressure</td>
</tr>
<tr>
<td>$c_{\mu}, c_{\epsilon_1}, c_{\epsilon_2}, c_{\epsilon_3}$</td>
<td>turbulence constants, Eqs. (2), (9), and (17)</td>
</tr>
<tr>
<td>$c_{\mu_1}, c_{\mu_2}$</td>
<td>turbulence constants, Eqs. (13), (14), (15), and (18)</td>
</tr>
<tr>
<td>$D_k^T$</td>
<td>species thermal diffusion coefficient</td>
</tr>
<tr>
<td>$D_{km}$</td>
<td>species mixture-average diffusion coefficient</td>
</tr>
<tr>
<td>$f_{\mu}, f_3$</td>
<td>damping functions, Eqs. (2), (9), (22), (23), and (25)</td>
</tr>
<tr>
<td>$h$</td>
<td>total enthalpy</td>
</tr>
<tr>
<td>$k$</td>
<td>turbulent kinetic energy</td>
</tr>
<tr>
<td>$K_g, K_s$</td>
<td>total number of gas-phase and surface species</td>
</tr>
<tr>
<td>$\ell_{\mu}, \ell_{\epsilon}$</td>
<td>turbulent length scales, Eq. (21)</td>
</tr>
<tr>
<td>$L$</td>
<td>channel length, Fig. 1</td>
</tr>
<tr>
<td>$Le$</td>
<td>Lewis number (thermal over species diffusivity)</td>
</tr>
<tr>
<td>$N_r$</td>
<td>number of gas-phase reactions, Eq. (11)</td>
</tr>
<tr>
<td>$p$</td>
<td>pressure</td>
</tr>
<tr>
<td>$P_k, G_k$</td>
<td>production terms of turbulent kinetic energy, Eqs. (6)–(8)</td>
</tr>
<tr>
<td>$Pr, Pr_t$</td>
<td>Prandtl number, turbulent Prandtl number</td>
</tr>
<tr>
<td>$R$</td>
<td>universal gas constant</td>
</tr>
<tr>
<td>$R_k$</td>
<td>turbulence Reynolds number, Eq. (21)</td>
</tr>
<tr>
<td>$Re$</td>
<td>Reynolds number</td>
</tr>
<tr>
<td>$r_k$</td>
<td>reaction rate of species $k$, Eq. (11)</td>
</tr>
<tr>
<td>$\dot{s}_m, \dot{s}_k$</td>
<td>species heterogeneous molar production rates, Eqs. (26) and (27)</td>
</tr>
<tr>
<td>$S_\varphi$</td>
<td>source term for variable $\varphi$, Eq. (1)</td>
</tr>
<tr>
<td>$T$</td>
<td>temperature</td>
</tr>
<tr>
<td>$u, v$</td>
<td>streamwise and transverse velocity</td>
</tr>
<tr>
<td>$u', v'$, $w'$</td>
<td>streamwise, transverse, and lateral turbulence intensity</td>
</tr>
<tr>
<td>$u_W$</td>
<td>wall friction velocity</td>
</tr>
<tr>
<td>$U_{IN}$</td>
<td>inlet streamwise velocity</td>
</tr>
<tr>
<td>$V_{k,y}$</td>
<td>transverse component of diffusion velocity for species $k$</td>
</tr>
<tr>
<td>$W$</td>
<td>channel width, Fig. 1</td>
</tr>
<tr>
<td>$W_k$</td>
<td>gas-phase species molecular weight</td>
</tr>
<tr>
<td>$X_k$</td>
<td>gas-phase species mole fraction</td>
</tr>
<tr>
<td>$Y_k$</td>
<td>gas-phase species mass fraction</td>
</tr>
<tr>
<td>$y^+$</td>
<td>nondimensional wall distance ($y^+ = \rho u_w y / \mu$)</td>
</tr>
<tr>
<td>$x, y, z$</td>
<td>streamwise, transverse and lateral coordinate, Fig. 1</td>
</tr>
</tbody>
</table>

### Greek symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Gamma$</td>
<td>surface site density, Eqs. (26) and (27)</td>
</tr>
<tr>
<td>$\Gamma_{\ell}, \Gamma_{\text{eff}}$</td>
<td>laminar and effective turbulent transport coefficients, Eq. (2)</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>dissipation rate of the turbulent kinetic energy</td>
</tr>
<tr>
<td>$\theta_m$</td>
<td>surface species coverage, Eq. (26)</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>von Karman constant, defined after Eq. (21)</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>thermal conductivity of the gas, Eq. (12)</td>
</tr>
<tr>
<td>$\mu, \mu_t$</td>
<td>laminar and turbulent viscosity</td>
</tr>
<tr>
<td>$v_{k,i}^n, v_{k,i}^l$</td>
<td>stoichiometric coefficients of species $k$ in reaction $i$, Eq. (11)</td>
</tr>
<tr>
<td>$\rho$</td>
<td>density</td>
</tr>
<tr>
<td>$\sigma_m$</td>
<td>surface species site occupancy, Eq. (26)</td>
</tr>
<tr>
<td>$\sigma_{\varphi}, \sigma_k, \sigma_\epsilon, \sigma_\rho, \sigma_g$</td>
<td>turbulent Prandtl or Schmidt numbers, Eqs. (2), (13), (14), and (15)</td>
</tr>
<tr>
<td>$\varphi$</td>
<td>scalar variable in Eq. (1), equivalence ratio</td>
</tr>
<tr>
<td>$\dot{\omega}_i$</td>
<td>molar production rate of the $i$th gaseous reaction, Eq. (11)</td>
</tr>
</tbody>
</table>

### Subscripts

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>channel center</td>
</tr>
<tr>
<td>IN</td>
<td>inlet</td>
</tr>
<tr>
<td>$k, m$</td>
<td>index for gas-phase species, index for surface species</td>
</tr>
<tr>
<td>$W$</td>
<td>wall</td>
</tr>
</tbody>
</table>

### Superscripts

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>~, --</td>
<td>Favre and Reynolds averages</td>
</tr>
<tr>
<td>$''$</td>
<td>fluctuations in Favre-averaged decomposition</td>
</tr>
</tbody>
</table>

Low-NO$_x$ catalytically stabilized combustion (CST) technology \[1,2\]. In CST, nearly half of the fuel is converted heterogeneously (catalytically) in reactors having a suitably large surface to volume ratio, and the remaining is combusted in a postcatalyst homogeneous (gaseous) burnout zone. Up to three sequential honeycomb-type catalytic reactor modules can be used \[3\], each consisting of a multitude of channels that are typically straight but can have various crossflow geometries. Under high-pressure operation, the incoming Reynolds numbers (based on the individual channel hydraulic diameter) can reach up to $4 \times 10^4$ \[2\]. Furthermore, for the maximum desired 50% heterogeneous fuel conversion and for the aforementioned range of Reynolds numbers, mass-transport considerations dictate for each catalytic module a length that is typically less than 80 channel hydraulic diameters \[2,3\], leading to spatially developing flows over the entire reactor extent. Multidimensional turbulent CST models used in catalytic combustor de-
sign should, therefore, describe adequately the transitional entry channel flow in the presence of both heterogeneous and homogeneous reaction pathways. The latter is particularly important in gas turbines due to the increased gas-phase reactivity of hydrocarbons at elevated pressures [4].

Elementary heterogeneous chemical reaction schemes for the oxidation of simple fuels (H$_2$, CO, and CH$_4$) over platinum, which is also the catalyst of the present investigation, have advanced considerably in the last years [5–7]. Multidimensional CST numerical models with detailed hetero/homogeneous chemistry and laminar transport have followed suit [8–11], providing a powerful tool for the design of—mainly lower pressure—industrial reactors and for fundamental studies in laboratory-scale burners. In particular, the two-dimensional models of our earlier laminar studies [8,9,12,13] have led, in conjunction with in situ measurements of species compositions and temperature over the catalyst boundary layer, to the validation of heterogeneous and homogeneous chemical reaction schemes in CST of CH$_4$/air and H$_2$/air over Pt, and to the elucidation of the underlying hetero/homogeneous chemistry coupling. Notwithstanding the advances in hetero/homogeneous chemistry and multidimensional laminar simulations, turbulent CST modeling has not yet received proper attention. Previous attempts used either simplified 1-D models with lumped transport coefficients, or 2-D approaches with isothermal turbulence models. Mantzaras et al. [14] and Appel et al. [15] have recently developed a turbulent CST model with the aim of reproducing two key characteristics, the heterogeneous fuel conversion and the onset of homogeneous ignition.

Numerical modeling of the spatially developing entry channel flow at industrially relevant Reynolds numbers is a particularly demanding task even for nonreacting cases. In gaseous channel flows with intense heating, the controlling parameters are the magnitude of the incoming turbulence and the strong flow laminarization induced by the wall heat transfer [16]; the resulting near-wall turbulence damping is much stronger compared to that of the isothermal case, which is driven solely by wall proximity, due to the additional influence of viscosity variation and, potentially, gravity. Direct numerical simulations (DNS) in the thermally developing region of a heated pipe airflow were reported recently [17] (albeit at a relatively low inlet Reynolds number of 4300) and suggested that laminarization was largely controlled by the continuous suppression of the near-wall vortical structures. DNS supplemented with heat-transfer experiments [18] have led to the development and validation of advanced low-Reynolds (LR) number turbulence models with appropriate damping functions suited for strongly heated and developing channel flows [16]. These models were implemented in Reynolds-averaged Navier–Stokes (RANS) approaches, which constitute the main tool for numerical heat transfer at practical Reynolds numbers; large eddy simulations (LES) are still hindered by the lack of proper subgrid scale models for wall-bounded flows [19]. In chemically reacting entry channel flows, on the other hand, DNS have not yet been reported: reacting DNS stress the temporally and not the spatially developing flows, due to the difficulty in applying proper initial conditions for the latter [20]. Many turbulent reacting flow models do not use appropriate near-wall corrections, leading to errors in the calculated wall heat fluxes and reaction rates (see discussion in Refs. [20,21]). The recently developed near-wall LR heat-transfer models have not been hitherto applied to reacting internal flows. The case similarity between heat transfer and CST flows is evident, since the exothermicity of the catalytic pathway induces a strong heating of the flowing gas. It is noted that whereas in pure gaseous internal-flow combustion heat is usually transferred from the reacting gas to the channel wall, thereby causing flame quenching [21,22], in CST heat is supplied from the hot catalytic wall to the reacting gas and this has been shown to extend the flame extinction limits [23].

In Appel et al. [15], the degree of flow laminarization was indirectly deduced from measurements of major species concentrations and temperature. The present investigation extends the earlier work by including direct in-channel measurements of the turbulent flow statistics in CST of H$_2$/air over Pt. For comparison purposes, room-temperature channel airflow were additionally investigated. Experiments have been performed in an optically accessible channel-flow catalytic reactor with incoming Reynolds numbers up to 3 × 10$^4$ and included digital particle image velocimetry (PIV) and laser Doppler velocimetry (LDV) for the characterization of the in-channel and the inlet turbulent flow, respectively, 1-D Raman measurements of major species concentrations and temperature for the assessment of turbulent scalar transport and, finally, planar laser-induced fluorescence (LIF) of the OH radical for the determination of homogeneous ignition. Numerical predictions were carried out with an elliptic 2-D RANS code that included different near-wall LR turbulence models. The main objectives were to quantify the degree of flow laminarization and to delineate the operating regimes over which a turbulence model was truly necessary in channel-flow CST. Particular objectives were to investigate the effect of turbulence on catalytic and gaseous combustion as well as on their respective coupling, to assess the impact of the incoming turbulence and, finally, to provide a numerical model that can reproduce key CST issues such as catalytic fuel
conversion, onset of homogeneous ignition, and flame propagation.

This article is organized as follows. The test rig, the measuring techniques, and the experimental conditions are first introduced, followed by the numerical model. The key issue of flow relaminarization is subsequently discussed with the aid of isothermal and CST experiments. Comparisons between measurements and numerical predictions lead to the validation of different LR models. The influence of turbulence on the catalytic fuel conversion and on the onset of homogeneous ignition is elaborated and the impact of the incoming turbulence is addressed. Finally, the CST operating regimes where flow laminarization prevails are delineated.

2. Experimental

2.1. Reactor geometry and test rig

The reactor was used in earlier laminar-flow CST studies [9] and hence is described briefly below. It consisted of two horizontal Si[SiC] ceramic plates, 300 mm long (L), 110 mm wide, and 10 mm thick, which were placed 7 mm (z) apart (see Fig. 1). Two quartz windows (300 × 12 × 3 mm³) formed the other sides of the reactor; the lateral window separation was 104 mm (W) and the channel combustor volume was delineated by the 300 × 104 × 7 mm³ enclosure. Four 7-mm-high ceramic spacers were affixed at the corners of the lower plate in order to maintain a constant 7-mm channel height. The upper plate was held on the spacers by gravity, thus minimizing thermal-stress-induced deformations that could alter the reactor height, which was a sensitive parameter in the numerical predictions. Optical accessibility was achieved from both 300 × 7 mm² sides, excluding the first 5 mm of the reactor. The inner horizontal surfaces of the Si[SiC] plates were coated with platinum; in the nonreacting test cases a pair of inert ceramic plates was used. The surface temperature was monitored along the x–y symmetry plane (z = 0) with 1-mm-thick S-type thermocouples (14 for each plate) embedded 0.9 mm beneath the catalyst through 1.2-mm diameters in diameter and 9.1-mm deep holes eroded from the outer plate surfaces. The channel reactor assembly was supported on ceramic rings that were, in turn, positioned inside an inconel-steel frame. To facilitate the laser-based measurements, the test rig was mounted on an axially traversable optical table.

In CST of diffusionaly imbalanced fuels with Lewis numbers $Le \ll 1$ (such as $H_2$ with $Le \approx 0.3$) the catalyst attained superadiabatic temperatures, which were much more pronounced at the upstream channel positions [9]. Operation under such conditions was not only detrimental to the catalyst integrity, but also compounded the experiments: tests have shown that upon homogeneous ignition the flame anchored directly at the hotter reactor entrance. To suppress the high catalyst temperatures and hence to control the onset of homogeneous ignition, a cooling/heating arrangement was adopted. The reactor entry was contacted to an 18-mm-long water-cooled metal section of the support frame, whereas the rear 240-mm of the ceramic plates was heated by two resistive heaters as to counteract the end-section heat losses (see Fig. 1). This arrangement resulted in flames anchored farther downstream in the channel, thus providing ample reactor length to study the pure heterogeneous processes that preceded the onset of homogeneous ignition. To further minimize heat losses from the rear part of the reactor, a 5-cm-thick ceramic insulation was placed between the heaters and the inconel-steel frame.

An oil-free compressor supplied air and high-pressure bottles provided hydrogen. The flow rates of the air and hydrogen were measured by four Brooks mass-flow controllers; the mixture equivalence ratio was determined with accuracy better than 1%. The reactants were mixed at room temperature in a 40-mm-long section filled with 2-mm-diameter metallic spheres to straighten the flow and, finally, were brought to the combustor through an inlet section $400 \times 21 \times 104 \text{ mm}^3$ (in $x$–$y$–$z$) with an ending y-contraction ratio of 3:1 (Fig. 1a). A metal grid with a $2 \times 2 \text{ mm}^2$ mesh was positioned 138 mm upstream of the reactor entry to generate turbulence and accelerate the transition of the flow. In the PIV tests, a fraction of the airflow was bypassed into a propeller-stirred tank containing $\text{Al}_2\text{O}_3$ seed particles with a size of 1 to 3 µm; the seeded airflow was subsequently injected into the inlet section through a 4-mm-diameter tube attached to the downstream end of the flow straightening section (Fig. 1a). A retractable sheathed thermocouple positioned 2 mm upstream of the reactor monitored the inlet temperature of the gaseous premixture across the y direction; it was withdrawn during the PIV/LIF/Raman measurements to avoid any flow disturbances. A second fixed thermocouple monitored the surface temperature of the water-cooled section; it was positioned 4 mm upstream of the reactor and embedded 0.9 mm beneath the metal surface. Finally, a 350-mm-long inconel-steel cylindrical exhaust section insulated with a porous fiber ceramic was attached at the reactor exit. A 50-mm diameter and 3-mm-thick quartz window positioned at the rear flange of the exhaust section provided an additional optical access for the LIF and PIV experiments.
2.2. Catalyst preparation

The inner surfaces of the Si[SiC] plates were coated with Pt using plasma vapor deposition (PVD): a 1.5-µm-thick nonporous Al₂O₃ layer was first deposited, followed by a 2.2-µm-thick Pt layer. The very thick Pt coating on top of the nonporous Al₂O₃ layer closely resembled a polycrystalline Pt surface as was also verified with independent surface area and surface composition measurements. The total and active surface areas were measured with BET (Kr physisorption) and CO chemisorption, respectively. The BET measurements confirmed the absence of porous surface structure and further showed that the total and active surface areas were the same (0.26 m²/g Cat), indicating a surface covered only with Pt. The latter was also verified with independent X-ray photoelectron spectroscopy (XPS) surface composition measurements before and after the Raman/LIF experiments; the XPS analyses have shown that the surface was covered with Pt and that Al or Si did not diffuse on the surface even after extended reactor operation. In the PIV experiments, however, which were carried out after the Raman/LIF tests, the postcombustion XPS analysis indicated a small deposition of Al₂O₃ seed (up to 20%) on the catalyst surface. This had no implications to the ensuing data interpretation given the extremely high activity of hydrogen on noble metals: as further discussed in Section 4.3.2, there were always ample free Pt sites to accommodate the catalytic processes. A Pt surface site density of $2.7 \times 10^{-9}$ mol/cm² (that of polycrystalline Pt [6,9]) was, therefore, used in the numerical model.

2.3. Test conditions

Three nonreacting and 11 reacting cases were investigated at atmospheric pressure with nominal incoming mean velocities of 20, 30, and 40 m/s (see Table 1). In the former cases the flow consisted of air (296 K) and in the latter of fuel-lean H₂/air premixtures. PIV/LDV experiments were carried out in the nonreacting Cases 1–3 and in the reacting Cases 4–8 while Raman/LIF measurements were performed in Cases 9–14. The PIV/LDV Cases 4–8 referred to pure heterogeneous combustion without homogeneous ignition; therefore, the maximum fuel-to-air equivalence ratio and the plate heating power were

Table 1

<table>
<thead>
<tr>
<th>Case</th>
<th>Type</th>
<th>Tests</th>
<th>$\varphi$</th>
<th>$U_{IN}$ (m/s)</th>
<th>$T_{IN}$ (K)</th>
<th>$Re_{IN}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Nonreacting</td>
<td>PIV</td>
<td>0.0</td>
<td>20</td>
<td>296</td>
<td>17,840</td>
</tr>
<tr>
<td>2</td>
<td>Nonreacting</td>
<td>PIV</td>
<td>0.0</td>
<td>30</td>
<td>296</td>
<td>26,710</td>
</tr>
<tr>
<td>3</td>
<td>Nonreacting</td>
<td>PIV</td>
<td>0.0</td>
<td>40</td>
<td>296</td>
<td>35,240</td>
</tr>
<tr>
<td>4</td>
<td>Reacting</td>
<td>PIV</td>
<td>0.18</td>
<td>20</td>
<td>300</td>
<td>15,390</td>
</tr>
<tr>
<td>5</td>
<td>Reacting</td>
<td>PIV</td>
<td>0.21</td>
<td>20</td>
<td>300</td>
<td>15,230</td>
</tr>
<tr>
<td>6</td>
<td>Reacting</td>
<td>PIV</td>
<td>0.18</td>
<td>30</td>
<td>300</td>
<td>23,090</td>
</tr>
<tr>
<td>7</td>
<td>Reacting</td>
<td>PIV</td>
<td>0.21</td>
<td>30</td>
<td>300</td>
<td>23,090</td>
</tr>
<tr>
<td>8</td>
<td>Reacting</td>
<td>PIV</td>
<td>0.21</td>
<td>40</td>
<td>300</td>
<td>30,460</td>
</tr>
<tr>
<td>9</td>
<td>Reacting</td>
<td>Raman/LIF</td>
<td>0.18</td>
<td>20</td>
<td>300</td>
<td>30,460</td>
</tr>
<tr>
<td>10</td>
<td>Reacting</td>
<td>Raman/LIF</td>
<td>0.24</td>
<td>20</td>
<td>300</td>
<td>30,080</td>
</tr>
<tr>
<td>11</td>
<td>Reacting</td>
<td>Raman/LIF</td>
<td>0.18</td>
<td>30</td>
<td>300</td>
<td>23,090</td>
</tr>
<tr>
<td>12</td>
<td>Reacting</td>
<td>Raman/LIF</td>
<td>0.24</td>
<td>30</td>
<td>300</td>
<td>22,610</td>
</tr>
<tr>
<td>13</td>
<td>Reacting</td>
<td>Raman/LIF</td>
<td>0.18</td>
<td>40</td>
<td>300</td>
<td>30,780</td>
</tr>
<tr>
<td>14</td>
<td>Reacting</td>
<td>Raman/LIF</td>
<td>0.24</td>
<td>40</td>
<td>300</td>
<td>30,150</td>
</tr>
</tbody>
</table>

* Equivalence ratio, mean inlet velocity and temperature, and inlet Reynolds number.
lower compared to those of the Raman/LIF cases that had combined hetero/homogeneous combustion. The Reynolds numbers shown in Table 1 were based on the inlet mean velocity, the inlet fluid properties, and the channel hydraulic diameter (= 13.1 mm). The relatively low mean inlet temperatures of all reacting cases ($T_{IN} = 300$ K) reflected the efficient water cooling and the very short contact times of the flow with the inlet section. For $T_{IN} = 300$ K, the adiabatic flame temperatures (constant pressure and enthalpy calculations) were 870 K ($\phi = 0.18$), 954 K ($\phi = 0.21$), and 1035 K ($\phi = 0.24$).

2.4. Laser diagnostics

The LIF, Raman, and PIV setup is illustrated in Fig. 2. The Raman/LIF test cases were completed first, followed by the PIV/LDV cases; otherwise, the remnant seeding particles would have complicated the sensitive spectroscopic measurements.

2.4.1. Flow velocity measurements

In the PIV experiments, a Quantel-Twins system was employed, consisting of two pulsed Nd:YAG lasers with an adjustable pulse delay. Each laser yielded a second harmonic radiation at 532 nm with a pulse energy and duration of 150 mJ and 4 ns, respectively. The two orthogonally polarized laser beams were combined into a double-pulsed beam by a polarization plate. A beamsplitter (60% transmission) attenuated the laser beam energy, which was then focused inside the reactor by an $f_1 = 1000$ mm focal length spherical lens (see Fig. 2). A telescopic system consisting of two cylindrical lenses with $f_2 = -150$ mm and $f_3 = -200$ mm, respectively, transformed the double-pulsed beam into two overlapping vertical light sheets that entered the reactor from the exhaust, propagating counterflow along the $x$–$y$ symmetry plane. The laser sheets had a slight divergence in the $y$ direction to ensure light contact to both channel walls. The scattered light was collected through one of the reactor side windows with a PCO-SensiCam CCD camera operated on the fast shutter mode. The camera was equipped with an $f = 70–210$ mm, $f/4$ zoom lens, a 532 nm bandpass interference filter, and a hot mirror to reflect the thermal radiation emitted from the hot catalytic plates.

The two sequential images were recorded on the CCD chip with a maximum resolution of $1280 \times 1024$ pixels per image. The field of view was $44 \times 9.9$ mm$^2$ (in $x$ and $y$, respectively, with the $y$ direction encompassing the entire 7-mm channel height and parts of the two solid walls) and was resolved with $1280 \times 288$ pixels for each image. The resulting optical magnifi-
cation ratio was 0.198. Only data from the relevant 1280 × 288 pixel areas of the CCD chip were stored (with an 8-bit resolution) on a dedicated PC, thus allowing a data acquisition and laser repetition rate of 3 Hz; 600 double images were recorded at each measuring location for the ensuing statistical analysis. The camera was supported on a frame with an x–y traverse. The frame limited the camera axial traverse down to \( x = 218 \text{ mm}\), whereas spurious light reflected from the inlet precluded measurements at \( x < 12 \text{ mm}\). The extent \( 12 \text{ mm} \leq x \leq 218 \text{ mm}\) was mapped with seven successive interrogation windows having a 30% axial overlap. The laser sheets were focused at the center of the measuring domain \((x \approx 110 \text{ mm})\) and had a beam waist of \( \approx 0.6 \text{ mm}\).

The data postprocessing was performed with the LaVision software [24]. The interrogation windows had an initial size of \( 32 \times 32 \text{ pixels} \) \((1.1 \times 1.1 \text{ mm}^2\) in physical space) and the successive windows had a 50% overlap in both \( x \) and \( y \) directions. The typical size of each imaged particle on the CCD chip was \( 2 \times 3 \) pixels, which was optimum for the reduction of velocity bias errors [25]. The time delay between the two PIV images was set to 6 \( \mu \text{s}\) for the highest velocity \((40 \text{ m/s})\) and 10 \( \mu \text{s}\) for the lowest velocity \((20 \text{ m/s})\) cases, resulting in particle displacements \( \approx 25\% \) of the interrogation domain. For accurate PIV velocity measurements, the particle displacement during the cross-correlation had to be determined with subpixel resolution; this entailed the assumption of a certain shape for the correlation peak (e.g., Gaussian), with the associated peak-locking bias errors [25]. The bias errors, along with additional system-specific noise (due to particle movement outside the interrogation window, decrease of the particle number density near the wall in the reacting cases for reasons explained below, and inclusion of the illuminated solid walls in the visualized image), limited the applicability of the standard FFT-based correlation algorithm and necessitated the use of the second-order correlation function of Hart [26]. To increase the signal-to-noise ratio, an adaptive multipass [27] (with decreasingly smaller interrogation windows of 32, 24, and 16 pixels) and a deformed interrogation windows approach [28] (depending on the local velocity gradient) were used. Both features were useful in the near-wall data analysis. The reacting PIV experiments were particularly demanding: the seeding particle number density scaled with the flow density, leading to lower particle number densities near the hot catalytic walls compared to those at the colder channel center. In addition, thermophoretic forces drove the seed particles away from the hot catalyst surfaces [29], further contributing to an inhomogeneous seed distribution. In the nonreacting cases there were, on the average, 8.5 particle pairs in the original \( 32 \times 32 \text{ pixel} \) interrogation window and this number was largely independent of \( y \) position. In the reacting cases, however, the average number of particle pairs was 7.5 at the channel center, 4.0 at \( y = 1.5 \text{ mm}\), and 2 at \( y = 0.50 \text{ mm}\). Velocity vectors as close as 0.25 \( \text{ mm}\) to both channel walls were evaluated in the nonreacting cases; in the reacting cases the corresponding distance was increased to 0.65 \( \text{ mm}\) due to the reduction in the near-wall particle density.

Validation algorithms were employed to remove spurious velocity vectors: a minimum threshold value was set for the acceptance of the peak correlation and the standard median filter [24] was applied to the velocity vectors. The noise-free root mean square (rms) velocity measurement errors calculated from the optical parameters and the pulse delay (Adrian [30]), were 0.18 and 0.31 \( \text{ m/s}\) for the lowest and highest velocities cases of Table 1, respectively. This entailed a velocity bandwidth (maximum-to-minimum resolvable velocity ratio) better than 110. Such high bandwidths were necessary in turbulent channel flows, which were characterized by outer zones near the channel center with high mean velocities and low relative turbulence intensities and near-wall inner regions with high relative turbulence intensities and low mean velocities. To ensure that the turbulence statistics were not contaminated by system-induced deterministic flow motions, time-series analyses of the velocity field near the entry were performed over the 600 successive realizations; the analyses showed that the flow did not contain any organized motion with frequencies slower than the data acquisition frequency (3 Hz).

The inlet axial velocity \((x = 0, z = 0)\) of all cases was mapped with LDV using an analogue plexiglass inlet/channel assembly (PIV and LDV in the test rig of Fig. 1) were not feasible at \( x \approx 0\). The absence of combustion in the analogue configuration had practically no impact on the assessed inlet velocities of the reacting Cases 4–14. The reason was the very efficient water cooling of the reactor frame which, in conjunction with the high flow velocities, resulted in average inlet bulk gas temperatures of 300 K (measured with the traversable thermocouple, see Fig. 1a) and in wall temperatures of the cooled metal section of 345 K (measured with the fixed thermocouple) for all reacting cases. The LDV setup consisted of a Dantec 57N20BSA system with a BB-W Coherent Innova 300 Argon-Ion 1.8-W continuous-wave laser and a photomultiplier tube in the backscatter mode. The axial velocity component was evaluated using the principal 514.5 nm radiation: a Bragg cell created two frequency-shifted beams that entered the reactor from the side. Both beams propagated on the \( x–z \) plane and were crossed at \( x = z = 0\) by an \( f = 310 \text{ mm} \) lens; the long axis of the formed LDV probe volume...
formed an acute $9^\circ$ angle with the $z$ axis, which was considered to correct for the magnitude of the axial velocity.

2.4.2. Spectroscopic measurements

The LIF and Raman setup is depicted in Fig. 2; the setup was the same used in earlier laminar studies [9] and, therefore, a short description is given below. In the planar LIF, a frequency-doubled Nd:YAG pulsed laser (Quantel YG781C20, 20 Hz repetition rate) pumped a tunable dye laser (Quantel TDL50) with a frequency-doubled radiation at 285.09 nm and a pulse duration of 15 ns. The laser beam was transformed into a vertical laser sheet by a cylindrical lens telescope and a 1-mm slit mask. The laser sheet propagated counterflow along the $x-y$ symmetry plane in a fashion similar to that of the PIV laser sheets. To ensure optical accessibility to both walls, the laser sheet had a small divergence in the $y$ direction. The pulse energy was low enough (0.5 mJ) to avoid saturation of the $A(v = 1) \rightarrow X(v' = 0)$ OH transition. The fluorescence of both $(1-1)$ and $(0-0)$ transitions at 308 and 314 nm, respectively, was collected at $90^\circ$ through one of the side quartz windows with an intensified CCD camera (LaVision FlameStar 2F, 576 x 384 pixels) gated to 100 ns; the camera was equipped with an $f = 105$ mm focal length $f/4.5$ lens (Nikon UV Nikkor) and an interference band-pass filter centered at 310 nm. A $180 \times 7$ mm$^2$ section of the combustor was imaged on a 576 x 24 pixel area of the CCD. One hundred images were averaged to increase the signal-to-noise ratio. In that, the LIF data provided the average flame position. The LIF was calibrated with absorption measurements performed with the vertical laser sheet crossing laterally ($z$) the combustion zone through both quartz windows, as in previous laminar studies [4,9]. The determination of absolute OH concentrations in a turbulent flow entailed inaccuracies; however, the LIF was used for the assessment of the flame position rather than for the extraction of absolute OH levels.

In the Raman experiments, the 248-nm radiation of a two-stage tunable narrowband KrF excimer laser (Lambda Physik Compex 150 T) was used. The pulse energy and duration were 230 mJ and 20 ns, respectively, and the repetition rate was 10 Hz. The $25 \times 9$ mm$^2$ rectangular excimer beam profile was focused to a vertical line ($\sim0.3$ mm thick) inside the reactor by an $f_z = 202$ mm cylindrical quartz lens (see Fig. 2). The focal line spanned the entire 7-mm transverse reactor extent and was 15 mm offset from the symmetry plane ($z = 15$ mm) in order to increase the collection angle and minimize beam steering. This offset had no impact to the forthcoming data evaluation, given the very large cross-flow aspect ratio (15:1) of the channel. Two $f_x = 200$ mm and $f_y = 300$ mm spherical lenses collected the scattered light (at a $50^\circ$ angle with respect to the sending optical path) and focused it at the entrance slit of a 25-cm imaging spectrograph (Chromex 250i). The spectrograph was equipped with a second intensified CCD camera identical to that of the LIF setup; the 576 and 384 pixel dimensions corresponded to spectral range and channel height, respectively. The spectral dispersion on the CCD camera ranged from 1250 to 5000 cm$^{-1}$, allowing observation of all major species ($O_2$, $N_2$, $H_2O$, and $H_2$); the Raman shift ranged from 1555 cm$^{-1}$ ($O_2$) to 4155 cm$^{-1}$ ($H_2$). For the evaluation of thermoscalar statistics, 600 single shot images were acquired at every axial position. The 7-mm channel height was resolved with 250 pixels. Raman data were acquired at different streamwise positions (starting at $x = 15$ mm) by traversing axially the combustor. Given the statistically steady nature of the tests, simultaneous acquisition of LIF and Raman data was not necessary; LIF images were acquired at regular time intervals in-between the Raman measurements.

The Raman data were referenced to a room temperature distribution of nitrogen inside the combustion channel. Determination of the effective Raman cross sections, which included transmission efficiencies of the detection system (i.e., windows, lenses, filter, spectrometer, and camera), was achieved by recording the signals of pure $H_2$, air, and completely burnt exhaust gases of known composition (the latter for assessing the cross section of $H_2O$). The $N_2$ line was used to assess the temperature. The 250 pixel transverse extent was binned to 27 pixels, providing a $y$ resolution of 0.26 mm. Raman data points closer than 0.6 mm to both walls were discarded due to low signal-to-noise ratio.

3. Numerical

3.1. Governing equations

Favre-averaged modeled transport equations for the first- and second-order moments of the gas-phase variables were constructed. For a statistically steady turbulent flow, the transport equation of any variable $\phi$ in its 2-D elliptic form becomes

$$\frac{\partial(\bar{\rho}\bar{\omega})}{\partial x} + \frac{\partial(\bar{\rho} \bar{\omega}_y)}{\partial y} = \frac{\partial}{\partial x} \left( \Gamma_{\text{eff}} \frac{\partial \bar{\omega}}{\partial x} \right) - \frac{\partial}{\partial y} \left( \Gamma_{\text{eff}} \frac{\partial \bar{\omega}}{\partial y} \right) = \bar{S}_\phi, \quad (1)$$

with

$$\Gamma_{\text{eff}} = \Gamma + \mu_t / \sigma_\phi \quad \text{and} \quad \mu_t = c_\mu f_{\mu} \bar{k}^2 / \bar{\varepsilon}. \quad (2)$$
\( \Gamma_\ell \) and \( \bar{S}_\varphi \) were the relevant laminar transport coefficients and the source terms, respectively. The governing equations were derived from Eq. (1) as follows [14,31].

Continuity:
\[
\varphi = 1, \quad \Gamma_{\text{eff}} = 0, \quad \bar{S}_\varphi = 0.
\]  

x momentum:
\[
\varphi = \tilde{u}, \quad \Gamma_\ell = \mu,
\]
\[
\bar{S}_\varphi = -\frac{\partial \tilde{p}}{\partial x} + \frac{\partial}{\partial x} \left( \Gamma_{\text{eff}} \frac{\partial \tilde{u}}{\partial x} \right) + \frac{\partial}{\partial y} \left( \Gamma_{\text{eff}} \frac{\partial \tilde{v}}{\partial x} \right) - \frac{\partial}{\partial x} \left[ \frac{2}{3} \Gamma_{\text{eff}} \left( \frac{\partial \tilde{u}}{\partial x} + \frac{\partial \tilde{v}}{\partial y} \right) \right] - \frac{\partial}{\partial y} \left( \frac{2}{3} \tilde{\rho} \tilde{k} \right). \tag{4}
\]

y momentum:
\[
\varphi = \tilde{v}, \quad \Gamma_\ell = \mu,
\]
\[
\bar{S}_\varphi = -\frac{\partial \tilde{p}}{\partial y} + \frac{\partial}{\partial x} \left( \Gamma_{\text{eff}} \frac{\partial \tilde{u}}{\partial y} \right) + \frac{\partial}{\partial y} \left( \Gamma_{\text{eff}} \frac{\partial \tilde{v}}{\partial y} \right) - \frac{\partial}{\partial y} \left[ \frac{2}{3} \Gamma_{\text{eff}} \left( \frac{\partial \tilde{u}}{\partial x} + \frac{\partial \tilde{v}}{\partial y} \right) \right] - \frac{\partial}{\partial y} \left( \frac{2}{3} \tilde{\rho} \tilde{k} \right). \tag{5}
\]

Turbulent kinetic energy:
\[
\varphi = \tilde{k}, \quad \Gamma_\ell = \mu, \quad \bar{S}_\varphi = P_k + G_k - \tilde{\rho} \tilde{\varepsilon}, \tag{6}
\]

with
\[
P_k = \mu_t \left[ \frac{2}{3} \left( \frac{\partial \tilde{u}}{\partial x} \right)^2 + \frac{2}{3} \left( \frac{\partial \tilde{v}}{\partial y} \right)^2 + \frac{2}{3} \left( \frac{\partial \tilde{w}}{\partial z} \right)^2 \right] - \frac{2}{3} \tilde{\rho} \tilde{k} \left( \frac{\partial \tilde{u}}{\partial x} + \frac{\partial \tilde{v}}{\partial y} \right)^2 - \frac{2}{3} \mu_t \left( \frac{\partial \tilde{u}}{\partial x} + \frac{\partial \tilde{v}}{\partial y} \right)^2. \tag{7}
\]

\[
G_k = -\frac{\mu_t}{\sigma_p \tilde{\rho}^2} \left[ \frac{\partial \tilde{p}}{\partial x} \frac{\partial \tilde{\rho}}{\partial x} + \frac{\partial \tilde{p}}{\partial y} \frac{\partial \tilde{\rho}}{\partial y} \right]. \tag{8}
\]

Dissipation rate of turbulent kinetic energy:
\[
\varphi = \tilde{\varepsilon}, \quad \Gamma_\ell = \mu,
\]
\[
\bar{S}_\varphi = (c_{\varepsilon_1} P_k + c_{\varepsilon_2} G_k - c_{\varepsilon_3} f_3 \tilde{\rho} \tilde{\varepsilon}) (\tilde{\varepsilon} / \tilde{k}). \tag{9}
\]

Species mass fractions:
\[
\varphi = \tilde{Y}_k, \quad k = 1, 2, \ldots, K_g.
\]

The molecular diffusion was treated in the source term (\( \Gamma_\ell \) was set to zero in Eq. (1)):
\[
\bar{S}_\varphi = \tilde{\rho} \tilde{r}_k + \frac{\partial}{\partial x} \left( \tilde{\rho} D_{km} \frac{\partial \tilde{Y}_k}{\partial x} \right) + \frac{\partial}{\partial y} \left( \tilde{\rho} D_{km} \frac{\partial \tilde{Y}_k}{\partial y} \right). \tag{10}
\]
The Reynolds-averaged gradients in Eq. (10) accounted for preferential diffusion effects and were modeled as in Ref. [31]. The reaction rate term in Eq. (10) was
\[
\tilde{r}_k = W_k \sum_{i=1}^{N_T} \left( \tilde{Y}_{k,i} - \tilde{Y}_{k,i}^\prime \right) (\tilde{\omega}_{i} / \tilde{\rho}). \tag{11}
\]

Total enthalpy:
\[
\varphi = \tilde{h}, \quad \Gamma_\ell = 0,
\]
with the molecular contribution treated again in the source term:
\[
\bar{S}_\varphi = \frac{\partial}{\partial x} \left[ \sum_{k=1}^{K_g} \left( \tilde{\rho} \tilde{D}_{km} \tilde{h}_k \frac{\partial \tilde{Y}_k}{\partial x} \right) \right] + \frac{\partial}{\partial y} \left[ \sum_{k=1}^{K_g} \left( \tilde{\rho} \tilde{D}_{km} \tilde{h}_k \frac{\partial \tilde{Y}_k}{\partial y} \right) \right]. \tag{12}
\]

The contribution of the modeled molecular terms in Eq. (12) was particularly small, justifying the commonly employed \( \bar{S}_\varphi = 0 \) for nonradiative flames.

Species variances and covariances:
\[
\varphi = Y_k'' Y_l'', \quad k = 1, 2, \ldots, K_g \quad \text{and} \quad \ell = 1, 2, \ldots, K_g,
\]
with \( \Gamma_\ell = 0 \) and
\[
\bar{S}_\varphi = c_{g_1} \frac{\mu_t}{\sigma_g} \left[ \frac{\partial \tilde{Y}_k}{\partial x} \frac{\partial \tilde{Y}_l}{\partial x} + \frac{\partial \tilde{Y}_k}{\partial y} \frac{\partial \tilde{Y}_l}{\partial y} \right] - c_{g_2} \tilde{h}_k \tilde{Y}_l'' + \tilde{h}_l \tilde{Y}_k'' + \tilde{h}_k \tilde{r}_l + \tilde{Y}_k'' \tilde{r}_l. \tag{13}
\]

Total enthalpy variance:
\[
\varphi = \tilde{h}''^2, \quad \Gamma_\ell = 0,
\]
\[
\bar{S}_\varphi = c_{g_1} \frac{\mu_t}{\sigma_g} \left[ \left( \frac{\partial \tilde{h}}{\partial x} \right)^2 + \left( \frac{\partial \tilde{h}}{\partial y} \right)^2 \right] - c_{g_2} \tilde{h} \tilde{h}'' + \tilde{h}'' \tilde{h}. \tag{14}
\]

Enthalpy-species covariances:
\[
\varphi = Y_k'' h'', \quad k = 1, 2, \ldots, K_g,
\]
with \( \Gamma_\ell = 0 \) and
\[
\bar{S}_\varphi = c_{g_1} \frac{\mu_t}{\sigma_g} \left[ \frac{\partial \tilde{Y}_k}{\partial x} \frac{\partial \tilde{h}}{\partial x} + \frac{\partial \tilde{Y}_k}{\partial y} \frac{\partial \tilde{h}}{\partial y} \right] - c_{g_2} \tilde{h} \tilde{Y}_k'' \tilde{h}''. \tag{15}
\]

Finally, the ideal gas equation was
\[
\tilde{\rho} = \tilde{\rho} R \sum_{k=1}^{K_g} \left( \tilde{Y}_k + \tilde{Y}_k'' \tilde{T}'' \right) / W_k. \tag{16}
\]

The temperature-related variables \( \tilde{T}, \tilde{T}'' \), and \( \tilde{Y}_k'' T'' \), were calculated from the dependent variables \( \tilde{h}, h''^2 \), and \( \tilde{Y}_k'' h'' \) as in [31].
The form of the damping functions $f_\mu$ and $f_3$ ($0 < f_\mu, f_3 \leq 1$) in Eqs. (2) and (9) was particular to each LR model. Their purpose was to render the models overdissipative compared to the standard high Reynolds number model: both functions approached unity far away from the wall. Standard values were used for the turbulence constants [32],

$$c_\mu = 0.09, \quad c_\epsilon = 1.44,$$
$$c_{\epsilon 2} = 1.0, \quad c_{\epsilon 3} = 1.92,$$  \hfill (17)
and the thermochemical constants [31],

$$c_{g1} = 2.8, \quad c_{g2} = 2.0, \quad \sigma_k = 1.0,$$
$$\sigma_\epsilon = 1.3, \quad \sigma_\rho = \sigma_g = 0.85.$$  \hfill (18)

Variations in some of the constant values for each particular LR model will be given in Section 3.2. Finally, the turbulent Prandtl number of Kays and Crawford [33] was used,

$$Pr_t = \left[ \frac{1}{2 Pr_{t,\infty}} + \frac{c Pe_\ell}{\sqrt{Pr_{t,\infty}}} \right]^{-1} - (c Pe_\ell)^2 \left( 1 - \exp \left( \frac{-1}{c Pe_\ell \sqrt{Pr_{t,\infty}}} \right) \right)^{-1},$$  \hfill (19)

with $Pe_\ell = (\mu_\ell / \mu) Pr, Pr_{t,\infty} = 0.85$, and $c = 0.3$.

For comparison purposes, laminar computations were also carried out; the corresponding governing equations for laminar CST have been provided elsewhere [9].

3.2. Low-Reynolds number turbulence models

Three different models were used to achieve turbulence closure; they included a two-layer LR model and two full LR models. The two-layer model of Chen and Patel [34] (further denoted as two-layer) has been tested in flows without heat transfer. It presented a computationally efficient approach by splitting the flow domain into two zones, thereby avoiding integration of the $\bar{e}$ equation directly to the wall. In the inner zone, which included the viscous sublayer, the buffer layer, and part of the fully turbulent layer, only the $\bar{e}$ equation was solved, while $\bar{\epsilon}$ and $\mu_\ell$ were calculated algebraically:

$$\bar{\epsilon} = \bar{R}^{3/2} / \ell_\epsilon \quad \text{and} \quad \mu_\ell = \bar{\rho} C_\mu \bar{R}^{1/2} / \ell_\mu.$$  \hfill (20)

The length scales $\ell_\mu$ and $\ell_\epsilon$ accounted for near-wall damping in terms of a local turbulent Reynolds number $R_\ell = k^{1/2} / y$, where $y$ was the distance from the wall [34],

$$\ell_\mu = c_\ell y \left[ 1 - \exp \left( -R_\ell / A_\mu \right) \right],$$
$$\ell_\epsilon = c_\ell y \left[ 1 - \exp \left( -R_\ell / A_\epsilon \right) \right].$$  \hfill (21)

with $c_\ell = \kappa c_\mu^{3/4}, \kappa = 0.42, A_\mu = 2c_\ell$, and $A_\epsilon = 70$. In the outer zone, the standard variable-density $\bar{k}-\bar{\epsilon}$ model (Eqs. (6) and (9)) was used, and the two zones were matched at $R_\ell \approx 200$. The model was typical to the isothermal models presently used in CST, hence allowing for performance comparisons with the following heat-transfer LR models.

The full LR models involved integration of both $\bar{k}$ and $\bar{\epsilon}$ equations directly to the wall. The first full LR model, further denoted as Ezato et al. [16], was a recent improvement of previous LR thermal entry-flow models [35] and has been validated in strongly laminarizing heated channel flows. Therein, the damping functions were

$$f_\mu = \left[ 1 - \exp \left( -y^* / 14 \right) \right]^2 \times \left[ 1 + (5/3) \exp \left( -R_\ell / 200 \right)^2 \right],$$
$$f_3 = \left[ 1 - 0.3 \exp \left( -R_\ell / 6.5 \right)^2 \right] \times \left[ 1 - \exp \left( -y^* / 3.1 \right)^2 \right],$$  \hfill (22, 23)

with $R_\ell = \bar{R}^{3/2} / (\nu e)$. The nondimensional velocity $y^* = u_\epsilon y / \nu$ was calculated in terms of the Kolmogorov velocity $u_\epsilon = (\nu e)^{1/4}$ rather than the wall friction velocity $u_\nu$, as to avoid singularities in separating flows. The constants $c_{\epsilon 1}, \sigma_k$, and $\sigma_\epsilon$ of Eqs. (17) and (18) were 1.5, 1.9, and 1.4, respectively.

The second full LR model (further denoted as Lin) was developed and tested in channel flows with and without heat transfer [36]. The independent variable $\bar{\epsilon}_\ell$ was used, with $\bar{\epsilon} = \bar{\epsilon}_\ell + \hat{\epsilon}$ and $\hat{\epsilon} = 2\nu (\partial \sqrt{\bar{k}} / \partial y)^2$ such that the wall boundary condition was $\langle \bar{\epsilon}_\ell \rangle w = 0$. Furthermore, the $\bar{k}$ and $\bar{\epsilon}_\ell$ transport equations contained the additional terms

$$\Pi_{\bar{k}} = -\frac{1}{2} \frac{\partial}{\partial x} \left( \frac{\mu_\ell}{\bar{k}} \frac{\partial \bar{k}}{\partial x} \right) - \frac{1}{2} \frac{\partial}{\partial y} \left( \frac{\mu_\ell}{\bar{k}} \frac{\partial \bar{k}}{\partial y} \right),$$
$$\Pi_{\bar{\epsilon}_\ell} = -\frac{\partial}{\partial x} \left( \frac{\mu_\ell}{\bar{k}} \frac{\partial \bar{k}}{\partial x} \right) - \frac{\partial}{\partial y} \left( \frac{\mu_\ell}{\bar{k}} \frac{\partial \bar{k}}{\partial y} \right)$$  \hfill (24)

that represented the pressure diffusion contribution separated from the molecular diffusion and the turbulent transport. The damping functions were

$$f_\mu = 1 - \exp \left( -y_{\gamma} / 100 - 8y_{\gamma}^3 / 1000 \right) \quad \text{and} \quad f_3 = 1.0,$$  \hfill (25)

with $y_{\gamma} = y (\bar{\epsilon}_\ell / \nu k)^{0.5}$ the transverse distance normalized by the Taylor microscale. This formulation reproduced the proper asymptotic limit toward the wall. The constants $\sigma_k$ and $\sigma_\epsilon$ were replaced by the functions $\sigma_k = 1.4 - 1.1 \exp \left( -y_{\gamma} / 10 \right)$ and $\sigma_\epsilon = 1.3 - \exp \left( -y_{\gamma} / 10 \right)$.
3.3. Thermochemistry modeling

3.3.1. Gas-phase modeling

The thermochemistry included modeled transport equations for the Favre-averaged mean values, variances, and covariances of the total enthalpy and the species mass fractions (Eqs. (3)–(15)). The approach of solving transport equations for all second-order scalar moments was advanced in Bockhorn [31]. In Eqs. (10) and (12)–(15) gradient-type modeling was used for the scalar fluxes and the dissipation hypothesis in a manner analogous to the turbulent flow model. The gradient scalar flux modeling was particularly justified in CST, due to its low heat-release rate [37]. To evaluate the mean chemical reaction rates (the terms $\tilde{\sigma}_m$ in Eq. (11)), a presumed Gaussian joint probability density function (pdf) coupling submodel was used for each elementary gaseous reaction. In the absence of reduced hetero/homogeneous chemical reaction schemes, this approach had the advantage of retaining an elementary level description for chemistry. It is emphasized that gaseous schemes reduced with pure homogeneous combustion investigations could not be used in CST due to the hetero/homogeneous coupling of intermediate species [9]. Measurements in noncatalytic turbulent reacting H$_2$/air boundary layers [38] and in nonreacting turbulent heated channel air flows [39] have shown nearly symmetric scalar pdfs, consistent with the Gaussian approach. In this respect, the Gaussian shape was less restrictive compared to open turbulent combustion applications [31]. The joint pdf for a bimolecular reaction was trivariate: $P(Y_1, Y_2, T)$ depended on the nine first- and second-order moments (established by the temperature and the two species mass fractions), which were computed with the modeled transport equations of Section 3.1. To expedite the computations, polynomial functions (in terms of the controlling nine moments) were fitted to the mean reaction rates. This approach was computationally more efficient compared to the look-up tables used previously [14]. A similar approach was undertaken for the $\tilde{Y}_k^{\mu}\tilde{r}_\ell$ source terms in Eq. (13).

3.3.2. Surface chemistry modeling

The instantaneous surface species coverage equations were the following:

$$\frac{\partial \sigma_m}{\partial t} = \frac{\partial \sigma_m}{\partial y}, \quad m = 1, 2, \ldots, K_s.$$  (26)

The instantaneous interfacial boundary conditions for the gas-phase species were

$$\rho_{k,s} \frac{\partial Y_k}{\partial t} = -[(D_{km} / Y_k)(\partial Y_k / \partial y) + \frac{\partial T}{\partial y} / (\rho Y_k T)]_{W} + \tilde{\sigma}_k W_k,$$

$$k = 1, 2, \ldots, K_g$$  (27)

with $\rho_{k,s}$ a characteristic surface density and $V_{k,y}$ the y component of the diffusion velocity, $(V_{k,y})_{W} = -[(D_{km} / Y_k)(\partial Y_k / \partial y) + \frac{\partial T}{\partial y} / (\rho Y_k T)]_{W}$ (accounting also for thermal diffusion). The left-hand side in Eq. (27) was the mass accumulation at the surface; on the right-hand side, the bracketed term was the combined Stefan flux, $(\rho Y_k v)_{W}$, and diffusive flux, $(\rho Y_k V_k,v)_{W}$, at the wall and the second term was the surface reaction. For a statistically-steady turbulent flow, averaging of Eqs. (26) and (27) removes the transient terms as well as the Stefan flux. On the other hand, the averaging introduced the same difficulties with the surface chemical source terms $\tilde{\sigma}_k$ and $\tilde{\sigma}_m$ as with the gaseous reaction rates discussed previously. However, the large thermal inertia of the Si[SiC] ceramic plates suppressed the surface temperature fluctuations and removed the major reaction nonlinearity due to the Arrhenius exponential term. In addition, the adsorption reactions were first order with respect to the adsorbing species [6] and hence they were a linear function of the relevant concentration; thus, averaging of the reaction rates reproduced exactly the mean species concentration. In any case, even for reaction rates with nonlinear concentration dependence, the resulting impact of the nonlinearity on the mean reaction rate was, for a constant reaction temperature, small (see Ref. [40]). The mean heterogeneous reaction rates were then treated with a “laminar-like” closure, evaluated at the wall temperature and the corresponding mean gas concentrations; in adsorption reactions, for example, $\tilde{\sigma}_k = \tilde{\sigma}_k(T_W, \tilde{Y}_k, \theta_{pi})$, and in reactions involving only surface species, $\tilde{\sigma}_m = \tilde{\sigma}_m(T_W, \theta_i)$ with $i$ the participating surface species.

3.4. Chemical kinetics

The heterogeneous scheme of Deutschmann et al. [6] (5 surface species, 6 gaseous species, and 16 reactions) was employed to describe the oxidation of H$_2$ over Pt. As stated in Section 2.2 the surface site density was $\Gamma = 2.7 \times 10^{-9}$ mol/cm$^2$, simulating polycrystalline platinum. For gaseous combustion, the scheme of Warnatz et al. [41] (38 reactions and 9 species including N$_2$) was used. The above schemes have been validated in their capacity to reproduce measured catalytic fuel conversion and onset of homogeneous ignition in previous laminar CST studies [9]. Surface reaction rates and laminar gaseous reaction rates were evaluated with Surface-CHEMKIN [42] and CHEMKIN [43], respectively.

3.5. Solution algorithm and boundary conditions

A finite volume procedure with a SIMPLER [44] approach for the pressure-velocity field was used.
Computations were carried out down to $x = 250$ mm, over half the channel domain ($250 \times 3.5$ mm$^2$). The discretized algebraic gas-phase transport equations were solved iteratively. In the two-layer model, an orthogonal staggered grid of $300 \times 24$ points (in $x$ and $y$ respectively, $250 \times 3.5$ mm$^2$) was used, whereas in the full LR models a $y$ grid of 72 points was sufficient to produce a grid-independent solution (the near-wall node was positioned at $y^+ = 0.5$, as opposed to $y^+ = 15$ in the two-layer model). The inlet conditions were uniform properties for the species mass fractions and temperature, and variable profiles for $\bar{U}$ and $\bar{k}$; the latter was deduced from the LDV-measured axial turbulence intensity $u'$ as $\bar{k} = 3/2u'^2$. The inlet $\bar{e}$ was calculated according to Ref. [45],

$$\bar{e}_{IN} = \bar{k}^{3/2}/\ell_e \quad \text{with} \quad \ell_e = 2.44\sqrt{1 - \exp(-R_k/3.8)},$$

which yielded the same $\bar{e}$ as the algebraic expression for the inner layer of the two-layer model (Eq. (20)).

The inlet temperature was uniform and equal to 296 K for the isothermal and the reacting cases, respectively. In the reacting cases, the retractable thermocouple revealed a thin ($\sim 0.5$ mm) thermal boundary layer 2 mm upstream of the reactor, which was neglected: its influence on the in-channel processes was insignificant as also discussed in earlier laminar studies [9]. At the wall ($y = 0$), the measured temperature profile—averaged between both plate distributions—was used as energy boundary condition: as the temperature differences between the two plates were less than $\pm 8$ K, this had no implications to the ensuing comparisons with the measurements. In the full LR models $\bar{e} = 2v(\partial \sqrt{k}/\partial y)^2$ at $y = 0$. Finally, zero-Neumann conditions were applied at the plane of symmetry ($y = 3.5$ mm) and at the end of the computational domain ($x = 250$ mm).

The surface and gas phases were coupled via the interfacial boundary conditions (Eqs. (27)). After completing a gas-phase iteration, the averaged Eqs. (26) and (27) were solved with a modified Newton’s method. The CHEMKIN database was used for thermodynamic [46] and laminar transport properties [47], and Warnatz et al. [48] for surface thermodynamic data. The computational time was $\sim 3$ days for two-layer model and $\sim 8$ days for the full LR models on a 3-processor (each running at 599 MHz) alpha cluster.

4. Results and discussion

4.1. Isothermal channel flows

The LDV-measured flows of mean axial velocity and axial turbulence intensity are presented in Fig. 3 for all isothermal cases. Therein, all Favre-averaged quantities were equivalent to the Reynolds-averaged ones. Comparisons between PIV-measured and predicted transverse profiles of $\bar{U}$ and $\bar{k}$ are depicted in Fig. 4 for Cases 1 and 3 at four selected axial positions. The measured $\bar{k}$ in Fig. 4 was deduced as $0.5(u'^2 + 2v'^2)$ by approximating the lateral intensity $w'$ to the measured transverse intensity $v'$: this assumption entailed a small underestimation of $\bar{k}$ in the near-wall region. The measurements of Lauffer [49] in fully developed 2-D channel flows have shown (for moderate Reynolds numbers $Re_{IN} \sim 3 \times 10^4$) that the near-wall peak intensities scaled as $w'/v' \sim 1.15$ and $u'/v' > 3$. This resulted in an underestimate of $\bar{k}$ by less than 3% when considering $w'/v' \sim 1$; the above underestimate was also expected to hold in the developing flows of Cases 1–3. Measured and predicted $\bar{U}_C$ and $\bar{k}_C$ along the symmetry axis ($y = 3.5$ mm, $z = 0$) are further provided in Fig. 5. For the large crossflow aspect ratio of the channel ($\sim 15:1$), the 2-D computations resulted in an underprediction of the true $\bar{U}$ by less than 4.5%, which was the theoretical (using analytical solutions) deviation between the corresponding $\bar{U}_C$ of two laminar fully developed flows: the 3-D channel-flow of Fig. 1 and the ideal 2-D flow between two infinitely wide parallel plates. The developing and turbulent character of the flow, however, implied that the 3-D effects introduced deviations in $\bar{U}$ that were considerably smaller than the aforesaid upper bound of 4.5%.

The comparisons in Figs. 4 and 5 indicated that the two-layer model provided very good agreement to the experiments, underpredicting in Case 1 the measured $\bar{U}$ at the channel core by less than 2.0% for all $x$. In Case 3, the two-layer model overpredicted $\bar{U}$ in the channel core by less than 1.5% for $x > 100$ mm. Moreover, the model captured excellently the axial position of the peak in $\bar{U}_C$ (see Figs. 5a and 5c). The
Fig. 4. Transverse profiles of mean axial velocities $\bar{U}$ and turbulent kinetic energies $\bar{k}$ for the nonreacting Cases 1 and 3 of Table 1. Symbols, PIV measurements; lines, numerical predictions (dashed lines, two-layer model; dashed-dotted lines, Lin model; solid lines, Ezato model). Four axial positions are shown: (a, e) $x = 15$ mm, (b, f) $x = 75$ mm, (c, g) $x = 135$ mm, and (d, h) $x = 195$ mm. The horizontal bars in (1h) and (3h) indicate the experimental uncertainty in $\bar{k}$. Note the change in the scale of the $\bar{U}$ axis in all figures.

Fig. 5. Streamwise profiles of the mean axial velocity $\bar{U}_C$ and turbulent kinetic energy $\bar{k}_C$ along the center of the channel ($z = 0, y = 3.5$ mm), for the nonreacting Cases 1 and 3 of Table 1. Solid lines, PIV measurements; dashed-lines, two-layer model predictions; dashed-dotted lines, Lin model predictions; dotted lines, Ezato model predictions.

The presence of a $\bar{U}_C$ maximum in entry-channel flows (also reported in earlier studies [50]) was controlled by the magnitude of $\bar{k}_IN$. For moderate $\bar{k}_IN$, $\bar{U}$ developed initially in a laminar fashion and farther downstream, upon appreciable wall-generated turbulence, $\bar{U}$ approached a flatter turbulent profile with a corresponding reduction in $\bar{U}_C$. As also evidenced in Figs. 5b and 5d, the peak in $\bar{U}_C$ was associated with the sharper rise in $\bar{k}_C$. The 2-D color maps of Fig. 6 provide measured and predicted (using the two-layer model) $\bar{U}$ and $\bar{k}$ for Case 3 and clearly show the peak of $\bar{U}$ at the channel core.

The two-layer model has also provided good agreement to the measured $\bar{k}_C$. It captured the continuous increase of $\bar{k}_C$ with increasing $x$ in the higher Reynolds number Case 3, and the very slow increase of $\bar{k}_C$ at $x > 150$ mm in the lower Reynolds number Case 1 (see Figs. 5b, 5d and 6). The dark brown areas along the wall and across the entrance in (3a) and (3c) denote zones without PIV data.

Fig. 6. Two-dimensional color maps of PIV-measured and numerically predicted (two-layer model) mean axial velocity $\bar{U}$ (3a, 3b) and turbulent kinetic energy $\bar{k}$ (3c, 3d) for Case 3 of Table 1. Measurements, (3a, 3c); predictions, (3b, 3d). The dark brown areas along the wall and across the entrance in (3a) and (3c) denote zones without PIV data.

The typical accuracy in the measured $\bar{k}$ is depicted with the error bars of Figs. 4 (1h, 3h). The error bars extend $\pm 2\sigma$ (standard deviations) and combine the sampling error due to the finite ensembles and the bandwidth error discussed in Section 2.4.1. The absolute errors in $\bar{k}$ increased near the wall appreciably; nonetheless, the measured near-wall peak $\bar{k}$ values were in good agree-
Fig. 7. (a) Thermocouple-measured axial profiles of the wall temperature for selected reacting cases of Table 1 (PIV cases, open symbols; Raman/LIF cases, filled symbols), (b) LDV-measured transverse profiles of inlet axial mean velocity (open symbols) and axial turbulence intensity (filled symbols) for the reacting cases.

ment to the two-layer model predictions. The error in $\tilde{U}$ (not shown in Fig. 4) was less than 2% of the corresponding mean value at the channel core and less than 6% at the measuring point closest to the wall.

The models of Lin and Ezato—the latter to a greater degree—underpredicted substantially the turbulence levels. As seen in Fig. 5d, Lin’s model yielded a much later rise in $\tilde{k}$, whereas Ezato’s model exhibited a nearly continuous mild drop of $\tilde{k}$ with increasing axial distance. Moreover, the near-wall peak $\tilde{k}$ levels were underpredicted significantly by both models, particularly in the higher Reynolds number Case 3 (see Fig. 4 (3e–3h)). It was, therefore, evident that both Lin and Ezato models were oversensitive under isothermal flow conditions. This was also attested in the predicted mean axial velocities profiles of Fig. 5; the dampening of turbulence led to appreciably higher $\tilde{U}$ farther downstream (see the Lin and Ezato model predictions in Fig. 4 (1c, 1d, 3d)), due to the reduced transport of energetic fluid from the core of the channel toward the wall.

The previous comparisons have clearly shown the applicability of the two-layer model and the inapplicability of the Lin and Ezato models in isothermal channel-flows. Implications of the above findings will be discussed in Section 4.4.

4.2. Flow field of reacting cases

The flow field under reacting conditions is discussed with the PIV experiments of Cases 4–8. The measured surface temperatures (averaged between both plates) are depicted in Fig. 7a for Cases 4 and 8. The suppression of the high entry temperatures with the employed water cooling was evident, especially for the leaner cases. Nonetheless, the surface temperatures exceeded the corresponding adiabatic flame temperatures over the streamwise extent $0 \leq x \leq 250$ mm by as much as 363 K (272 K) for

Fig. 8. Regimes of flow laminarization in channel flows due to heat transfer. The lines proposed by various authors in the $Re_{IN}$ versus $q^+_IN = q_W/(\rho U_c T)_{IN}$ plot delineate an upper laminarizing regime and a lower regime where turbulence is sustained. The vertical bars position the reacting Cases 4–14 of Table 1 (A, cases with nominal velocity 20 m/s; B, cases with nominal velocity 30 m/s; C, cases with nominal velocity 40 m/s). The shaded area is an estimate of the CST operating regimes.

$\varphi = 0.21$ ($\varphi = 0.18$). This degree of superadiabaticity was controlled primarily by the nature of the H$_2$ catalytic combustion (as discussed in Section 2.1) and to a lesser degree by the provided external heating. The LDV-measured inlet mean velocity and axial turbulence intensity profiles are illustrated in Fig. 7b; for a given mean velocity, the turbulence intensities were practically independent of $\varphi$, over the range $0.18 \leq \varphi \leq 0.21$.

The laminarization in channel flows is discussed with the aid of Fig. 8, which was adapted from pure heat-transfer studies. Laminarization is controlled by two competing parameters: the incoming Reynolds number $Re_{IN}$ and the nondimensional heat-transfer parameter $q^+_IN = q_W/(\rho U_c T)_{IN}$, with $q_W$ the wall
heat flux. The lines in Fig. 8 are literature estimates [51–53] and delineate the regimes of prevailing laminarization. In the regions above the lines, which are characterized by strong heat transfer, the flow laminarizes (turbulence is dampened) whereas in the regions below the lines turbulence is sustained. The shaded area in Fig. 8 is an estimate of the $Re_{IN} - q_{IN}$ domain relevant to CST. It is seen that CST spans both the laminarizing and the nonlaminarizing regimes. The vertical bars in Fig. 8 refer to the reacting cases, which extend to both regimes.

Comparisons between PIV-measured and predicted transverse profiles of $\tilde{U}$ and $\tilde{k}$ for Cases 4 and 8 are depicted in Fig. 9, while streamwise profiles of $\tilde{U}_C$ and $\tilde{k}_C$ are given in Fig. 10. To facilitate the ensuing comparisons, the nature of all measured statistical quantities will be considered as Favre averaged. In most laser-based techniques, an inherent density weighting of the measurements was conceivable. Moreover, for the low-density fluctuations typical of internal flows, the difference between Favre- and Reynolds-averaged values was small [54]. The comparisons in Figs. 9 and 10 indicated that Ezato’s model provided good agreement to the measured $\tilde{U}$ and captured well the continuous increase of $\tilde{U}_C$ with increasing $x$. The model of Lin, and to a greater extent the two-layer model, overpredicted $\tilde{U}$ farther downstream (see Fig. 9 (4c, 4d, 8c, 8d) and Figs. 10a and 10c). The measured $\tilde{k}_C$ decreased with increasing $x$ and this was captured well by both Ezato and Lin models. On the other hand, the two-layer model provided a continuous increase of $\tilde{k}_C$ with increasing $x$. The measured near-wall peak of $\tilde{k}$ decreased with increasing $x$; only Ezato’s model captured this trend, although the absolute value of the near-wall $\tilde{k}$ was underpredicted. The model of Lin, and particularly the two-layer model, resulted in a considerably higher near-wall $\tilde{k}$, which increased with increasing axial distance (Fig. 9). The predicted $\tilde{k}$ levels had also a direct impact on the mean velocities: a higher computed near-wall $\tilde{k}$ resulted in an enhanced transport of heat from the wall to the channel core, thereby decreasing the mean flow density and accelerating the flow. The escalating overpredictions in $\tilde{U}$ obtained with the Lin and two-layer model, respectively, were consistent with their significant $\tilde{k}$ overpredictions.
The previous discussion has shown that the model of Ezato reproduced well the turbulent flow field of the reacting cases—even though it overpredicted somewhat the degree of laminarization—the model of Lin exhibited only a mild laminarization as manifested by the drop in $\tilde{k}_C$, and the two-layer model provided a continuous increase of $\tilde{k}$ without laminarization. It was, therefore, concluded that the isothermal turbulence models (such as the two-layer) were inapplicable in CST. From the tested heat-transfer models, only Ezato’s model provided a turbulence damping that was realistically strong. It should be noted, however, that a distinction between reacting and pure heat-transfer flows is the combustion-induced volumetric expansion (reduction) and the associated flow acceleration (deceleration), an effect that deserves future attention in reacting-flow LR-model development. Nonetheless, the volumetric change in CST was particularly small, as it referred to combustion in air with ultralean fuel stoichiometries.

The laminarization was further investigated by comparing typical instantaneous spanwise vorticity maps for the isothermal and reacting Cases 3 and 8, respectively. The maps of Fig. 11 were constructed by subtracting the measured mean from the measured instantaneous velocity and then computing the vorticity of the resulting velocity field. High-vorticity-elongated structures inclined at 40–50° with respect to the flow direction were typical to the isothermal cases (see circled areas in Fig. 11a); such structures have been also identified in fully developed isothermal turbulent channel flows [55]. In the reacting case of Fig. 11b, however, the vortical structures were not only attenuated in magnitude but also lost their elongated topological features. This picture of laminarization was consistent with DNS observations in heated channel flows [17].

4.3. Combustion characteristics

The heterogeneous and homogeneous combustion processes are elaborated with the Raman/LIF data of Cases 9–14. Typical measured surface temperatures are illustrated in Fig. 7a for four selected Raman/LIF cases; the inlet velocity and turbulence intensities in Cases 9–14 (Fig. 7b) were practically the same as those of Cases 4–8. The surface temperatures of Cases 9–14 were up to 80 K higher than those of the PIV cases, to always assure homogeneous ignition within the reactor. The catalytic processes preceding homogeneous ignition are discussed using the Raman measurements and then the follow-up gaseous combustion is examined with the aid of both LIF and Raman data.

4.3.1. Heterogeneous combustion

Computed streamwise profiles of the local catalytic ($C$: black lines) and gaseous ($G$: gray lines) average hydrogen conversion rates are provided in Figs. 12a and 12c for Cases 10 and 14, respectively. The shaded areas encompass the reactor extent downstream of the predicted homogeneous ignition ($x_{ig}$), which is defined in the forthcoming Fig. 15. Laminar simulations are also provided in Fig. 12, whereby the mean inlet velocities and surface temperatures were the same as those in Cases 10 and 14; however, turbulence was turned off (for details of the laminar modeling, see Appel et al. [9]). In Figs. 12a and 12c, the volumetric gaseous reaction rates have been integrated across the 7-mm channel height so that they can be directly compared to the surface catalytic rates. The streamwise profiles of the predicted percentage hydrogen conversion (combined catalytic and gaseous) are further given in Figs. 12b and 12d. The symbols with the error bars in Figs. 12a and 12c designate the measured total (catalytic and gaseous) local hydrogen conversion rates. Those measurements entailed the determination of the local hydrogen fluxes across the channel and could not be deduced solely by the Raman-measured transverse $H_2$ profiles (see Fig. 13) but they also required the knowledge of the velocity $\tilde{U}(y)$; the $\tilde{U}(y)$ profiles of Ezato’s model predictions were used, as they were shown in Section 4.2 to reproduce well the PIV data of the reacting cases. The error bars in the measurements of Figs. 12a and 12c extend ±2σ and were calculated.
Fig. 12. Streamwise profiles of H\textsubscript{2} conversion for Cases 10 and 14. In (a) and (c) the Raman-measured (circles with error bars) and predicted (lines) local H\textsubscript{2} conversion rates are shown in black lines and the gaseous conversions (G) in gray lines; the measurements refer to the combined (catalytic and gaseous) local conversions. The shaded areas denote the reactor extent downstream of the onset of homogeneous ignition (as predicted with the model of Ezato and indicated by the vertical arrow marked $x_{\text{ig}}$). The measured homogeneous ignition is indicated by $x_{\text{ig},m}$ and that predicted with laminar modeling by $x_{\text{ig},\ell}$. In (b) and (d) the predicted percentage H\textsubscript{2} conversion (catalytic and gaseous) is given along the channel. Dashed lines, two-layer model; dashed-dotted lines, Lin model; solid lines, Ezato model; dashed-double-dotted lines, laminar model predictions.

using the estimated accuracy of the Raman data. In Figs. 12a and 12c, the measured homogeneous ignition position (deduced from the OH-LIF data of Fig. 15) is denoted by $x_{\text{ig},m}$ and the predicted one from laminar modeling by $x_{\text{ig},\ell}$; as will be further discussed in the next section, the two-layer model failed to provide homogeneous ignition in all cases, whereas Lin’s model yielded a weak ignition only in Case 10. Comparisons between Raman-measured and predicted (using the model of Ezato) transverse profiles of the average H\textsubscript{2} and H\textsubscript{2}O mole fractions and of the temperature are provided in Fig. 13 at four selected streamwise positions for Cases 9, 10, and 14. The first two axial positions in Fig. 13 preceded homogeneous ignition. Over the reactor extent $0 \leq x \leq x_{\text{ig}}$ hydrogen was converted solely by the catalytic pathway as seen by comparing the corresponding C and G profiles of Figs. 12a and 12c. The model of Ezato provided good agreement to the measured catalytic conversion rates while the models of Lin and, in particular, the two-layer model overpredicted appreciably the corresponding rates. The model of Lin yielded integrated catalytic H\textsubscript{2} conversions down to $x_{\text{ig}}$ (see Figs. 12b and 12d by 8 and 15% higher compared to those of Ezato’s model for Cases 10 and 14, respectively; the corresponding numbers for the two-layer model were 23 and 66%. The reason for the above discrepancies was that the Lin and two-layer models did not capture the near-wall drop in $\tilde{k}$ (in accordance with the volumetric compositions as low as 1%. The temperature measurements were based on the excess nitrogen and had a typical for Raman experiments accuracy of $\pm 50$ K. As seen in Fig. 13, the predicted concentrations of hydrogen (the deficient reactant) were practically zero at the catalytic walls, indicating a mass-transport-limited operation, i.e., a hydrogen conversion controlled by the rate of turbulent transport. This was also supported by the Raman measurements of Fig. 13, notwithstanding the near-wall experimental limitations. The mass-transport-limited operation was highly desirable as it rendered the validation of turbulent models free from potential inaccuracies in the surface kinetics.

Over the reactor extent $0 \leq x \leq x_{\text{ig}}$, hydrogen was converted solely by the catalytic pathway as seen by comparing the corresponding C and G profiles of Figs. 12a and 12c. The model of Ezato provided good agreement to the measured catalytic conversion rates while the models of Lin and, in particular, the two-layer model overpredicted appreciably the corresponding rates. The model of Lin yielded integrated catalytic H\textsubscript{2} conversions down to $x_{\text{ig}}$ (see Figs. 12b and 12d by 8 and 15% higher compared to those of Ezato’s model for Cases 10 and 14, respectively; the corresponding numbers for the two-layer model were 23 and 66%. The reason for the above discrepancies was that the Lin and two-layer models did not capture the near-wall drop in $\tilde{k}$ (in accordance with the
Fig. 14. Raman-measured (symbols) and predicted (lines) transverse profiles of average hydrogen mole fraction for Case 14 at $x = 130$ mm. The horizontal bars indicate the measurement uncertainty. Dashed lines, two-layer model; dashed-dotted lines, Lin model; solid lines, Ezato model predictions.

findings of Fig. 9) and hence overpredicted the effective transport coefficient $\Gamma_{\text{eff}}$ (Eq. (2)); this, in turn, resulted in an increased transverse scalar transport toward the wall and in a higher $\text{H}_2$ catalytic consumption. It is emphasized that the catalytic chemistry was fast enough to cope with the enhanced transport of the Lin and two-layer models as further illustrated in Fig. 14 (corresponding to Case 14 at $x = 130$ mm, just upstream of $\text{x}_{\text{ig}}$): the $\text{H}_2$ concentration at the channel walls was practically zero in all models, demonstrating again a mass-transport-limited $\text{H}_2$ conversion.

The model of Ezato provided a good agreement to the Raman-measured average $\text{H}_2$ and $\text{H}_2\text{O}$ concentrations and temperature over the preignition zones (Fig. 13). The deviations of Ezato’s model predictions to the measurements shown in Figs. 12 and 13 were within the experimental uncertainty. Nonetheless, this model showed a systematic slight underestimation of the measured catalytic $\text{H}_2$ conversion rates of Fig. 12 (or, equivalently, slight overestimation of the measured $\text{H}_2$ levels in Fig. 13 (10b, 14b)), signifying a somewhat stronger laminarization compared to the one suggested by the measurements. This outcome was also supported by the slight underprediction of the average temperature (Fig. 13) and was also consistent with the PIV studies of Section 4.2.

The previous discussion has clearly shown that standard isothermal (cold-flow) models, such as the two-layer model, were inapplicable in turbulent CST as they resulted in considerable overprediction of the turbulent transport and hence of the fuel catalytic conversion rates. Interestingly, the laminar predictions of Fig. 12 were much closer to the measurements compared to the two-layer model predictions or even to insufficiently laminarizing heat-transfer models (Lin); the laminar $\text{H}_2$ conversions at $\text{x}_{\text{ig}}$ were by 6.5% (Fig. 12b) and 10% (Fig. 12d) lower than those of the Ezato model. This suggested that, under certain operating conditions with moderate Reynolds numbers, a laminar model was well suited for CST. This issue will be further discussed in Section 4.4.

4.3.2. Homogeneous combustion

Comparisons between LIF-measured and predicted 2-D maps of the OH radical are illustrated in Fig. 15 for four selected cases. The predictions in Fig. 15 refer only to those models that captured homogeneous ignition (shown by the yellow arrows in Fig. 15) and flame formation. The stability of the measured flame shapes and their anchoring positions ($\text{x}_{\text{ig}}$) was excellent over extended time periods. The anchoring of a flame inside a straight channel at such high flow velocities—without any additional means of aerodynamic stabilization—was a worth-moting feature of catalytically supported combustion. The model of Ezato captured well the onset of homogeneous ignition (underpredicting $\text{x}_{\text{ig}}$ by only $\sim 15\%$) and the flame shape in all cases. The two-layer model could not reproduce homogeneous ignition in any case and, finally, the model of Lin yielded ignition only for Case 10—albeit with a very short resulting flame length (see Fig. 15 (10c)). The above model dis-

Fig. 15. LIF-measured (a) and numerically predicted (b, c) maps of the OH radical for Cases 9, 10, 11, and 14: (b) Ezato model predictions, (c) Lin model predictions for Case 10. The yellow arrows indicate the onset of homogeneous ignition, measured and predicted. The color bars provide the predicted OH levels in ppm vol.
crepencies were directly linked to the laminarization of the turbulent flow. The two-layer and Lin models provided higher $k$ levels and increased turbulent scalar transport (as discussed in Section 4.2), which had a direct impact on homogeneous ignition. The enhanced transport of those models resulted in increased upstream catalytic conversion (see Fig. 12) that, in turn, deprived hydrogen from the gaseous reaction pathway and at the same time removed efficiently heat from the near-wall hot ignitable mixture toward the colder channel core; both factors inhibited the onset of homogeneous ignition. Furthermore, the increased transport rates had a profound effect on the confined flames of Fig. 15 by reducing the gaseous combustion residence times. This effect was first investigated in laminar stagnation-flow catalytic combustion with fuels having $Le < 1$ [23]. An increase in the strain rate was shown [23] to push the flame against the catalytic wall, leading to incomplete combustion through the gaseous reaction zone and to a subsequent catalytic conversion of the leaked fuel; a further increase of the strain rate led to flame extinction. The turbulent transport played a role analogous to that of the strain rate in laminar stagnation flows. The enhanced turbulent transport led to lack of gaseous ignition for the two-layer and the Lin models, except in Case 10 where the latter model predicted the formation of a short flame (Fig. 15 (10c)). Case 10 had the strongest possible laminarization due to its lowest $Re_{IN}$ (see Table 1) and its relatively high wall temperatures over the preignition zone (see Fig. 7a); moreover, it was particularly resilient to potential flame extinction due to its highest $\varphi$. Those factors allowed for an initial flame development in the model of Lin; however, since there was no laminarization and $k$ increased farther downstream, the flame extinguished after a short distance.

In the model of Ezato, the continuous flow laminarization allowed for longer flame residence times that led to sustained near-wall gaseous combustion, in agreement with the LIF experiments (Fig. 15). The predicted transverse hydrogen profiles of Fig. 13 at the last postignition axial positions (d) indicated that, even well downstream homogeneous ignition, the catalytic pathway converted hydrogen in parallel to the gaseous pathway. In the higher Reynolds number Case 14 or the lowest equivalence ratio Case 9, for example, the predicted transverse gradients of hydrogen at the wall (shown with the thick solid lines in Fig. 13 (9d, 14d)) were nonzero, indicating fuel leakage through the flame zone. This was also supported by the hydrogen measurements of Fig. 13, despite the near-wall limitations of the Raman experiments. The fuel leakage was insignificant in Case 10 since (as discussed above) this case exhibited the strongest flow laminarization; over the postignition zone of Case 10, practically all fuel was converted via the gaseous pathway. Fig. 16 provides computed (using the model of Ezato) transverse profiles of the average gaseous hydrogen reaction rates; the push of the reaction zone toward the catalytic wall (leading to incomplete combustion) with increasing Reynolds number (Case 14) or leaning of the mixture (Case 9) was evident. It is further noted that in CST of fuels with $Le \ll 1$ the onset of homogeneous ignition was actually advantageous for catalyst thermal stability reasons. In reactors with a nearly adiabatic operation (without, for example, passive catalyst cooling [56]), a pure catalytic combustion mode led to superadiabatic surface temperatures (as discussed in Section 2.1) that endangered the catalyst integrity, while the presence of a gaseous combustion zone moderated the surface temperatures by shielding the catalyst and depriving it from fuel. The current experiments have also shown that during the transient processes of ignition and flame stabilization, the onset of homogeneous ignition was always accompanied by a drop in the thermocouple-measured surface temperatures. In that, the aforesaid fuel leakage through the gaseous reaction zone, which was augmented considerably by the turbulent transport, was not beneficial as it increased the relative contribution of the catalytic pathway to the total fuel conversion.

As discussed previously, turbulence inhibited the onset of homogeneous ignition. This was also evident from Fig. 12: the laminar homogeneous ignition...
The nondimensionalization of the temperature at the wall in Eq. (29) implied that homogeneous ignition was favored at short characteristic chemical times and/or at long diffusion times (channels with large half-height $b$, i.e., low surface-to-volume ratio). Although Eq. (29) was developed for laminar flows, it was informative to examine its dependence under turbulent flow conditions. In the presence of turbulence, the wall transverse temperature gradient $\theta_W'$ increased (and hence $F(\zeta_{ig})$ decreased) and at the same time the diffusion time decreased due to the increase in the effective (turbulent) diffusivity $a_{th}$. Both effects led to an increase in $\zeta_{ig}$ and hence to inhibition of homogeneous ignition. Physically, the first effect ($\theta_W'$) signified the more efficient transport of heat away from the near-wall hot ignitable mixture, and the second effect ($a_{th}$) the enhanced contribution of the catalytic pathway in fuel conversion. The previous analysis considered the chemical time unaffected by turbulence, a reasonable assumption for the moderate turbulence intensities of wall-bounded flows.

To complete the picture of the in-channel hetero/homogeneous processes, the predicted surface coverage is illustrated in Fig. 17 for Case 10; the dominant coverage was free platinum sites. For the high wall temperatures of the present study ($T_W > 965$ K, see Fig. 7a), the surface oxygen could not block effectively the adsorption of $H_2$ (see also [9]). Therefore, there were always ample Pt sites to accommodate the heterogeneous processes. Additional computations showed that a reduction of the active surface area even by a factor of 2 had no influence on the model predictions. In that, the small (20%) reduction of the active area due to $Al_2O_3$ deposition in the reacting PIV experiments did not impact the combustion processes of Cases 4–8. The Lin and Ezato models provided slightly lower Pt coverage over the extent $0 \leq x \leq 150$ mm due to their increased turbulent transport that resulted in increased loading of the surface. The onset of homogeneous ignition in the model of Ezato was manifested in Fig. 17 by the sudden drop in $\theta_H$ due to the deprivation of gaseous hydrogen above the catalytic surface.

4.4. Computation in practical systems

Having established the applicability of the LR model of Ezato, additional computations of fuel-lean ($\phi = 0.24$) $H_2$/air catalytic combustion over Pt with $T_{IN} = 300$ K were performed in commercial channel geometries. The channel separation ($2b$) was reduced to 1.2 mm while the pressure was increased to 15 bar as to maintain high inlet Reynolds numbers. Plug-flow inlet conditions were considered for $U_{IN}$ and $k_{IN}$, while the surface temperature was taken constant along the channel. Extensive parametric studies were carried out by varying the surface temperature from 1000 to 1200 K, the incoming turbulence $k_{IN}$ from 0.005$U_{IN}^2$ to 0.05$U_{IN}^2$, and the incoming Reynolds number from $10^4$ to $4 \times 10^4$. The gas-phase chemistry was removed in order to investigate the influence of
ence of laminarization on the pure catalytic processes. The inlet uniform flow conditions for the turbulent kinetic energy and its dissipation rate were approximated as [59]

\[ \tilde{k}_\text{IN} = \alpha_1 \tilde{U}_\text{IN}^2 \quad \text{and} \quad \tilde{\epsilon}_\text{IN} = (\epsilon_\mu/\alpha_2) k_{\text{IN}}^{3/2} / r_h, \]  

with \( r_h \) the hydraulic radius and \( \alpha_2 = 0.03 \). In plug-flow turbulence, a typical value for \( \alpha_1 \) was 0.005. In the present study very high values (up to 0.05) were also investigated: for isotropic turbulence \( \alpha_1 = 0.05 \) corresponded to a high relative turbulence intensity \( u'/\tilde{U}_\text{IN} = 0.18 \). The incoming turbulence levels in practical gas turbine systems were determined by the compressor discharge and the downstream fuel/air mixing. Moreover, only the part of the turbulence spectrum with length scales smaller than the hydraulic diameter of the channel could enter the reactor. However, spectral energy considerations have shown [60] that those small eddies had a profound impact on combustion.

Computed streamwise profiles of the percentage hydrogen catalytic conversion rates are provided in Fig. 18 (Ezato and laminar models) for a wall temperature \( T_W = 1200 \) K, three \( Re_\text{IN} \), and four different values of \( \alpha_1 \). The channel length was sufficient (300 mm) to achieve catalytic fuel conversions in excess of 80%, which is considerably higher than the 50% requirements of CST [1,2]. In the lower Reynolds number case \( (Re_\text{IN} = 2 \times 10^4) \), the laminar conversion was by as much as 17 and 24% lower than the turbulent conversions with \( \alpha_1 = 0.005 \) and 0.05, respectively. This was due to the continuous drop of the turbulence intensity along the channel. Therefore, under intensely laminarizing conditions, a simple laminar model is adequate and certainly more accurate that an isothermal LR turbulence model; in the case of Fig. 18a, for example, the two-layer model (not shown) predicted an H\(_2\) conversion by as much as 52% larger than the laminar one when \( \alpha_1 = 0.005 \).

In the \( Re_\text{IN} = 4 \times 10^4 \) case of Fig. 18c, the turbulent hydrogen conversions were, irrespective of \( \alpha_1 \), considerably higher (by as much as 45%) than the laminar conversion. For \( Re_\text{IN} = 4 \times 10^4 \) and irrespective of \( \alpha_1 \), the turbulent kinetic energy increased along the channel. In the intermediate \( Re_\text{IN} = 3 \times 10^4 \) case of Fig. 18b, turbulence was dampened for \( \alpha_1 = 0.005 \) (leading to \( H_2 \) conversions differing by as much as 14% from the laminar conversion) and it increased for \( \alpha_1 \geq 0.01 \). The domains of flow laminarization are shown with the shaded areas in Fig. 19. Therein, turbulence is dampened and the \( H_2 \) conversion differs by less than 10% of the corresponding laminar conversion; this difference is considerably reduced farther to the left of the demarcating line(s). For high-temperature catalysts with \( T_W = 1200 \) K (and \( T_\text{IN} = 300 \) K), turbulence modeling is truly needed for \( Re_\text{IN} > 2.7 \times 10^4 \) when \( \alpha_1 = 0.005 \) and for \( Re_\text{IN} > 2 \times 10^4 \) when \( \alpha_1 = 0.05 \). It is also understood that a reactant preheat would narrow the laminarization domains of Fig. 19, which pertain to \( T_\text{IN} = 300 \) K.

The turbulent model of this study was developed for 2-D straight channels and as such was also applicable to 2-D axisymmetric configurations. Honey-
comb channels do not have a square but rather a circular cross-section due to the rounding of the corners during the coating process. In that, secondary flows typical to sharp-edged cross sections are avoided and hence the developed model is also applicable to practical systems. The present model is valid for steady-state, high-temperature reactor performance. Transient behavior, such as the light off of a catalytic reactor, would necessitate the development of LR models that perform adequately over a much wider surface temperature range. The previous analysis has shown that a single LR turbulent model cannot perform satisfactorily from 300 to 1200 K and future research should emphasize the development of such models.

5. Conclusions

The turbulent catalytically stabilized combustion of hydrogen/air mixtures over Pt was investigated experimentally and numerically in channel-flow configurations. Additional experiments and predictions were carried out with room-temperature airflows, at incoming Reynolds numbers close to those of the reacting cases. The following are the key conclusions of this study.

(1) The turbulent flow field of the nonreacting cases was captured well by a standard two-layer LR near-wall turbulence model. In the reacting cases, good flow predictions were achieved by the heat-transfer LR model of Ezato; this model captured the dampening of turbulence along the catalytic channel, overpredicting only mildly the degree of flow laminarization. The two-layer model overpredicted substantially the levels of turbulence in the reacting cases whereas the model of Ezato was overdissipative in all nonreacting cases. A second heat-transfer LR model (Lin model) could not reproduce the strong flow laminarization of the reacting cases. Advanced LR near-wall turbulent models adopted from recent heat-transfer studies appear to be the best candidates for channel-flow combustion applications.

(2) The heterogeneous processes were strongly influenced by the particular LR model. Turbulence models that failed to capture the intense flow laminarization induced by the heat transfer from the hot catalytic walls led to an increased turbulent transport of fuel toward the catalytic surface and hence to a substantial overprediction of the heterogeneous conversion. The model of Ezato reproduced well the catalytic processes, underpredicting only mildly the catalytic fuel conversion, in agreement with its somewhat stronger laminarization discussed before. At moderate inlet Reynolds numbers, a simple laminar model yielded much better agreement to the measurements compared to the standard two-layer turbulence model.

(3) The onset of homogeneous ignition in CST was inhibited by turbulence, due to the increased heat transfer from the near-wall hot ignitable mixture toward the colder channel core and due to the enhanced catalytic fuel consumption preceding homogeneous ignition. Therefore, turbulence models that did not reproduce the CST-relevant flow laminarization could not capture homogeneous ignition. The model of Ezato underpredicted mildly the onset of homogeneous ignition, which was again consistent with its somewhat stronger flow laminarization.

(4) Following homogeneous ignition and flame formation, a combined heterogeneous and homogeneous fuel conversion could be achieved, depending on the particular turbulence levels. An increase in the turbulent transport resulted in a successive push of the reaction zone toward the catalytic wall, in fuel-leakage through the near-wall gaseous reaction zone and, finally, in subsequent catalytic conversion of the leaked fuel. In practical systems operating with strongly diffusionally imbalanced fuels having \( Le \ll 1 \) (such as hydrogen) the fuel leakage was detrimental to the catalyst as it augmented the surface temperatures through the superadiabatic combustion of the heterogeneous pathway.

(5) Computations with the validated model of Ezato in channels of practical systems have delineated the regimes of surface temperatures \( T_W \), incoming turbulent kinetic energies, and incoming Reynolds numbers \( Re_{IN} \) where the flow laminarizes. The regimes of laminarization encompass a sizeable part of the operating conditions of practical systems and can be computed with good accuracy using a laminar model.

(6) No single LR model performed adequately under isothermal (nonreacting) and high-temperature (reacting) conditions. The modeling of transient light-off processes would, therefore, require the development of LR models appropriate for the temperature range \( 300 \, \text{K} \lesssim T_W \lesssim 1200 \, \text{K} \).

Acknowledgments

Support was provided by the Swiss Federal Office of Energy (BFE) and Alstom Power of Switzerland. We thank Dr. F. Raimondi and Ms. F. Geiger for the XPS and BET analyses.

References
