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Detailed transient numerical simulation of H_2/air hetero-/homogeneous combustion in platinum-coated channels with conjugate heat transfer

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ABSTRACT

Transient two-dimensional simulations of fuel-lean H_2/air combustion were performed in a 2-mm-height planar channel coated with platinum, using detailed hetero-/homogeneous chemistry and transport as well as heat conduction in the solid wall. The developed model resolved, for the first time, all relevant spatiotemporal scales in a practical channel-flow reactor configuration. A parametric study was carried out to investigate the effects of wall material, inlet velocity, and inlet temperature on the fundamental catalytic and gas-phase combustion processes. Computational singular perturbation (CSP) analysis identified the key catalytic reactions affecting light-off and homogeneous ignition. Homogeneous ignition crucially depended on the OH desorbing fluxes from the catalyst, while flame propagation and stabilization involved time scales of a few milliseconds. During the short duration of the light-off event, the ensuing Stefan velocity appreciably altered the flow field. Predictions of time accurate numerical simulations were further compared against those of a code relying on the quasisteady state assumption, and the specific conditions under which the latter was invalidated were identified. Finally, CSP analysis unraveled the reasons for the high computational cost of the fully transient 2-D simulations. The surface reaction mechanism exhibited a high stiffness with fastest time scales of the order of 10^{-12} s, pertaining to the hydrogen adsorption and to the $H(s) + O(s) = OH(s) + Pt(s)$ reactions. These time scales were in turn six orders of magnitude shorter than the ones associated with gas-phase chemistry or with a simplified single-step catalytic reaction.

1. Introduction

Combustion in microscale and mesoscale channels has attracted increased attention in recent years due to the growing interest in developing portable power generation systems [1,2]. Although pure gas-phase combustion can be stabilized both in microscale [3] and mesoscale [4] channels, defined respectively as channels with height smaller or larger than 1 mm [1], increasing surface-to-volume ratios render catalytic (heterogeneous) combustion the preferred route compared to pure gaseous (homogeneous) combustion. Catalytic reactions broaden the classical flammability limits and lead to stable combustion even in the presence of high heat losses [4,5]. Recent works [6,7] have further demonstrated the potential of catalytically-coated walls in suppressing intrinsic flame instabilities of micro- and meso-scale channels. Applications of catalytic combustion at the mesoscale also include stationary gas turbines of large power plants, whereby combustion is attained in honeycomb reactors comprising a multitude of catalytically coated channels with individual hydraulic diameters of 1 to 2 mm [8]. Relevant combustion methodologies include the fuel-lean catalytically-stabilized thermal combustion (CST) [9,10] and the fuel-rich catalytic/fuel-lean gaseous combustion [11–13], with the potential to achieve single-digit NO_x emissions ($\leqslant$3 ppmv [9]). Catalytic combustion methodologies crucially depend on advances in catalyst technology (development of active and thermally stable catalysts) as well as on multidimensional modeling needed for reactor design, with the latter being the focus of the present work.

Development of advanced numerical catalytic combustion models poses severe challenges due to the different physicochemical processes with disparate spatio-/temporal scales: surface and gas-phase chemistry, convective and diffusive heat and mass transport within the bulk flow (interphase transport), intraphase transport in the catalyst washcoat, heat conduction in the channel wall and

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radiative heat transfer. The corresponding time scales range from fractions of a millisecond (surface and gaseous chemical times) to seconds (solid axial heat conduction time). Steady and transient models are required for the simulation of an individual catalytic channel during steady operation and light-off, respectively. Furthermore, a spatial dimensionality of at least two is necessary to correctly describe interphase transport and, in particular, homogeneous combustion. The reason is that gaseous combustion, which is in most cases detrimental to the catalyst integrity and hence of interest for safety reasons, is largely controlled by the reactant species and temperature boundary layer profiles [14,15]. Homogeneous chemistry is often neglected in catalytic combustion modeling even though its influence, especially on steady-state reactor operation and at elevated pressures, can be significant [16–18]. Flow laminarization induced by the intense heating from the catalytic surfaces renders laminar modeling appropriate at high-enough inlet Reynolds numbers, where an isothermal flow would have otherwise transitioned to turbulence [19]. Three-dimensional effects do play a role in commercial straight-channel honeycomb reactors [20,21], despite the rounding of the channel corners during catalyst coating [22] that in turn suppresses strong secondary flows.

Elliptic (full Navier–Stokes) 2-D models with detailed hetero-/homogeneous chemistry [8,19,23–33] have nowadays become common research tools for steady simulations, but, to the best of the authors' knowledge, research codes based on such detailed models have not been applied to time-accurate transient simulations. Transient catalytic channel simulations with detailed methane surface chemistry were reported in [34] using a commercial CFD software, however, this study did not address issues like light-off, gas-phase ignition and hetero-/homogeneous chemistry coupling.

Given the long characteristic solid wall heat-up time compared to the chemical, convective and diffusive time scales of a catalytic reactor, the quasisteady-state (QSS) assumption for the flow, gas-phase chemistry, and surface chemistry is often invoked. It is nonetheless understood that the QSS assumption may not hold at all spatial locations and/or all times during transient reactor operation [17,35]. Two-dimensional transient CFD models for a single channel invoking the QSS approximation and using single-step surface chemistry were reported in earlier works [35,36]. More recently, a QSS transient 2-D channel model including detailed surface and gas-phase kinetics, 1-D heat conduction in the solid wall and surface radiation heat transfer, was able to reproduce ignition and extinction experiments in catalytic partial oxidation (CPO) of methane over a rhodium-coated reactor [37]. The same code was subsequently applied to study the light-off and hetero-/homogeneous combustion of lean syngas/air mixtures over platinum [38] and of lean methane/air mixtures in Pt-coated microreactors [17]. The quasisteady approximation imposes in many cases a 1-D (axial) model for the solid wall heat conduction [17,37] (a 2-D description would render the transverse heat conduction in the solid faster than the gas-phase equilibration process and would thus invalidate the QSS approximation). Fully transient 2-D models with inclusion of intraphase washcoat diffusion but simplified catalytic chemistry were presented in [39]. Fundamental studies have also been carried out using transient codes with detailed chemistry for 1-D stagnation flows [40,41], which are characterized by an affordable computational cost, although this geometry is not particularly relevant to practical systems.

The present work undertakes a fully transient (without invoking the quasiteady-state assumption) 2-D numerical study of fuel-lean hydrogen/air hetero-/homogeneous combustion in a planar mesoscale (2 mm height) Pt-coated channel. Detailed transport, gas-phase and surface chemistry were used together with 2-D heat conduction in the solid wall. Fast processes such as light-off, homogeneous ignition and hetero-/homogeneous combustion interactions during dynamic operation, with time scales ranging from fractions of a millisecond to a few milliseconds, were studied for the first time – given the model's capacity to resolve all relevant time scales. The code further assessed the regimes of validity for the QSS assumption by comparing predictions with those obtained by the QSS code presented in [37,38].

The paper is organized as follows. The numerical model, the employed chemical reaction schemes, and the validation of the implemented conjugate heat transfer submodel are presented in Section 2, while main results follow in Section 3. The Computational Singular Perturbation method is introduced in Section 3.1 and applied in Section 3.2 to analyze the data of a transient surface perfectly stirred reactor in order to unravel the origin of the stiffness that characterizes the surface reaction mechanism. Section 3.3 presents the main results of 2-D transient simulations. The physicochemical processes of light-off, wall heat-up and gas-phase ignition are discussed in Sections 3.3.1, 3.3.2 and 3.3.3 whereby CSP is applied in the channel, while comparisons between time-accurate detailed numerical simulations and QSS predictions are elaborated in Section 3.3.4.

2. Numerical approach

2.1. Governing equations

The gas-phase is described by the conservation equations for mass, momentum, energy and species in the low-Mach-number limit [42,43]. Therein, acoustic waves are neglected, allowing for longer integration times steps, while compressibility effects due to heat release and density variations are fully accounted for.

Continuity

$$\frac{\partial \rho}{\partial t} + \underline{\nabla} \cdot (\rho \underline{u}) = 0 \tag{1}$$

Momentum

$$\rho\left(\frac{\partial \underline{u}}{\partial t} + \underline{u} \cdot \underline{\nabla}\,\underline{u}\right) = -\underline{\nabla} p_1 + \underline{\nabla} \cdot (\mu \underline{\underline{S}}) \tag{2}$$

$$\underline{\underline{S}} = \underline{\nabla}\,\underline{u} + (\underline{\nabla}\,\underline{u})^T - \frac{2}{3}(\underline{\nabla} \cdot \underline{u})\underline{\underline{I}} \tag{3}$$

Energy

$$\rho c_p\left(\frac{\partial T}{\partial t} + \underline{u} \cdot \underline{\nabla} T\right) = \underline{\nabla} \cdot (\lambda \underline{\nabla} T) - \sum_{i=1}^{N_g} h_i \dot{\omega}_i - \rho\left(\sum_{i=1}^{N_g} c_{p,i} Y_i \underline{V_i}\right) \cdot \underline{\nabla} T \tag{4}$$

$$c_p = \sum_{i=1}^{N_g} c_{p,i} Y_i \tag{5}$$

Gas-phase species

$$\rho\left(\frac{\partial Y_i}{\partial t} + \underline{u} \cdot \underline{\nabla} Y_i\right) = -\underline{\nabla} \cdot (\rho Y_i \underline{V_i}) + \dot{\omega}_i \quad i = 1, \ldots, N_g \tag{6}$$

with N_g the total number of gaseous species.

Equation of state (ideal gas law)

$$p_0 = \frac{\rho R T}{\overline{W}} \tag{7}$$

with $\overline{W}$ the mean molecular weight of the gas mixture:

$$\overline{W} = \left(\sum_{i=1}^{N_g} \frac{Y_i}{W_i}\right)^{-1} \tag{8}$$

The leading order term p_0 is the thermodynamic pressure, while the first order term p_1 appearing in the momentum equation is the hydrodynamic pressure.