NON-LINEAR EVOLUTION OF THERMODIFFUSIVELY UNSTABLE LEAN PREMIXED HYDROGEN FLAMES

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Abstract

The characteristic patterns of thermodiffusively unstable lean premixed hydrogen flames are studied in several large-scale simulations conducting a parameter variation with respect to the thermal expansion ratio of the flame. This enables a systematic separation of the influence of the thermodiffusive and Darrieus-Landau instabilities on the flame front's evolution. A detailed chemical mechanism is employed and the variation of the thermal expansion ratio is obtained by a modification of the equation of state. For all simulations, the flame front corrugation possesses two characteristic length scales. In particular, flames without thermal expansion also feature both length scales, suggesting that both length scales originate from the thermodiffusive instability. Additionally, the size of the largest characteristic length scale is found to increase with the thermal expansion ratio.

Introduction

Recently, the interest in hydrogen combustion has increased as it represents a promising candidate for carbon-free energy carriers in future energy systems. In order to store an energy excess, hydrogen can be generated by the electrolysis of water and in times of demand, it can be electrified again by burning it in gas turbines. However, hydrogen combustion significantly differs from conventional combustion of hydrocarbons since it is prone to combustion instabilities. In particular, lean, premixed hydrogen combustion is susceptible to thermodiffusive and Darrieus-Landau instabilities. Thermodiffusive instabilities arise from the low Lewis number of hydrogen, which induces a strong disparity between the molecular and thermal fluxes entering and leaving the flame front. Depending on the Lewis number, this disparity can either stabilize or destabilize the flame front. The Darrieus-Landau instability is induced by the thermal expansion of the gas during combustion, which is always destabilizing. These instabilities have been studied by Matalon and Matkowsky [1], Pelce and Clavin [2], and Frankel and Sivashinsky [3], among others. As typical flame configurations always involve thermal expansion, thermodiffusively unstable flames are always accompanied by Darrieus-Landau instabilities. Thus, separately studying the impact of the thermodiffusive instability on the flame front evolution becomes difficult. In this work, we performed large-

scale Direct Numerical Simulations (DNS), for which the equation of state is modified such that a separation of the thermodiffusive and Darrieus-Landau instability is possible, enabling a rigorous analysis of the former.

Configuration

The simulation configuration comprises an initially planar, premixed, lean hydrogen/air flame that burns in a rectangular two-dimensional domain towards an inlet as shown in Fig. 1. The simulation domain is periodic in crosswise direction and the inlet velocity of the unburnt mixture is chosen such that the flame is stabilized sufficiently long within the simulation domain. The initially planar flame is perturbed by a harmonic wave to trigger the flame instabilities. All simulations feature an equivalence ratio of $\phi = 0.4$ and an unburnt temperature of $T_u = 298K$. Five different simulations have been conducted, in which the thermal expansion ratio ρ_u/ρ_b is varied from 1 to 4.42 by modifying the equation of state (c.f. section Governing Equations). All computational domains comprise a length of 200 laminar flame thicknesses in the streamwise and crosswise directions. If not stated differently, all quantities are normalized by the laminar flame thickness l_F or the laminar flame speed s_L of the corresponding unstretched premixed flame. Since the long-term behavior of the various flames is studied, simulations are run for at least 300 flame times. The constant density case is run for 1400 flame times.

Governing Equations & Numerical Framework

The governing equations of the DNS are given by the reacting Navier-Stokes equations in the low-Mach limit [4]. Chemical reactions are modeled by the mechanism of Hong et al. [5], which contains 9 species and 46 reactions. Species diffusivities are determined imposing constant, non-unity Lewis numbers, which are the same for all simulations. A detailed description of the employed numerical methods can be found in Ref. [6] and simulations similar to those conducted in this study are described in Ref. [7]. For all cases, the laminar flame thickness is resolved by ten grid points and timesteps between $3\mu s$ and $6\mu s$ are used. The imposed resolution yields numerical meshes of 2048 x 2048 grid points.

The equation of state has been modified by a scaling factor α with the objective to vary the flames' thermal expansion ratio. The resulting equation of state yields

$$\rho = \alpha \left(\frac{p}{RT} - \rho_u \right) + \rho_u, \tag{1}$$

where ρ, p, R, T represent the density, the pressure, the ideal gas constant divided by the mean molar mass of the fluid, and the temperature, respectively. ρ_u refers to the density of the unburnt mixture. The coefficient α is used to control the thermal expansion. It is varied among the different simulations between $\alpha = 0$, for which the density is constant, and $\alpha = 1$, for which the equation of state of an ideal gas is obtained. The case $\alpha = 1$ is referred to as reference case and is characterized by a thermal expansion ratio of $\rho_u/\rho_b = 4.42$.

Results and Discussion
In Fig. 1, characteristic snapshots of the flames are shown for four of the five conducted simulations. The simulation domains are cropped in streamwise direction to fit all flames, but their full width is shown. For the reference case, Berger et al. [7] proved that the flame dynamics become independent of the domain size if the width of the computation domain comprises at least 100 l_F. Similarly to their study, all flames in Fig. 1 reveal two characteristic length scales. First, a small wrinkling scale that periodically recurs along the flame front is identified, which, for the reference case, has been shown by Berger et al. [7] to be equal to the most unstable wave length of the dispersion relation. Second, a characteristic large structure always appears, which is marked in Fig. 1 and will be referred to as flame finger. These flame fingers periodically arise from the flame front and propagate towards the inlet. Except for the constant density case, these fingers are found to feature a tilting behavior in their propagation movement towards the inlet such that they tilt laterally and eventually collide with a different part of the flame. According to Berger et al. [7], this tilting behavior is a consequence of the deflection of the streamlines at a flame finger's tip. The displacement speed of the flame finger is found to be symmetric with respect to its tip, but as soon as a finger tilts laterally the flow field normal to the flame front becomes asymmetric such that the tilting behavior is amplified. In the constant density case the velocity is constant in the entire domain, so this mechanism does not apply and consequently no tilting behavior is observed. However, in the constant density case, the flame fingers can still move laterally just without tilting, which still enables an interaction among themselves, e.g., merging of two flame fingers. Thus, flame fingers are found to exist also for the constant density case, where Darrieus-Landau instabilities are suppressed. Consequently, their existence needs to originate from the thermodiffusive instability. A visual inspection of Fig. 1 suggests that an increasing thermal expansion ratio of the flame amplifies the size of the flame fingers. Movies of the temporal evolution of all cases will be shown at the conference.

In the following, the impact of the instabilities on the flame front corrugations is assessed by means of the consumption speed. It is defined as

$$s_{cons} = \frac{1}{L \rho_u Y_{H_2,u}} \int \dot{\omega}_{H_2} \, dx \, dy, \tag{2}$$

where L is the domain size in crosswise direction, $Y_{H_2,u}$ is the unburnt mass fraction of hydrogen and $\dot{\omega}_{H_2}$ represents the source term of hydrogen due to chemical reactions. Fig. 2 shows the time-averaged consumption speed with respect to the thermal expansion ratio ρ_u / ρ_b. The standard deviation of the temporal fluctuations is described by the respective error bar. If neglecting the constant density case, an increasing thermal expansion ratio always leads to an increasing consumption speed. Since the consumption speed depends on the ratio of the largest to the smallest characteristic length scale of the flame front corrugations [8], the range of length
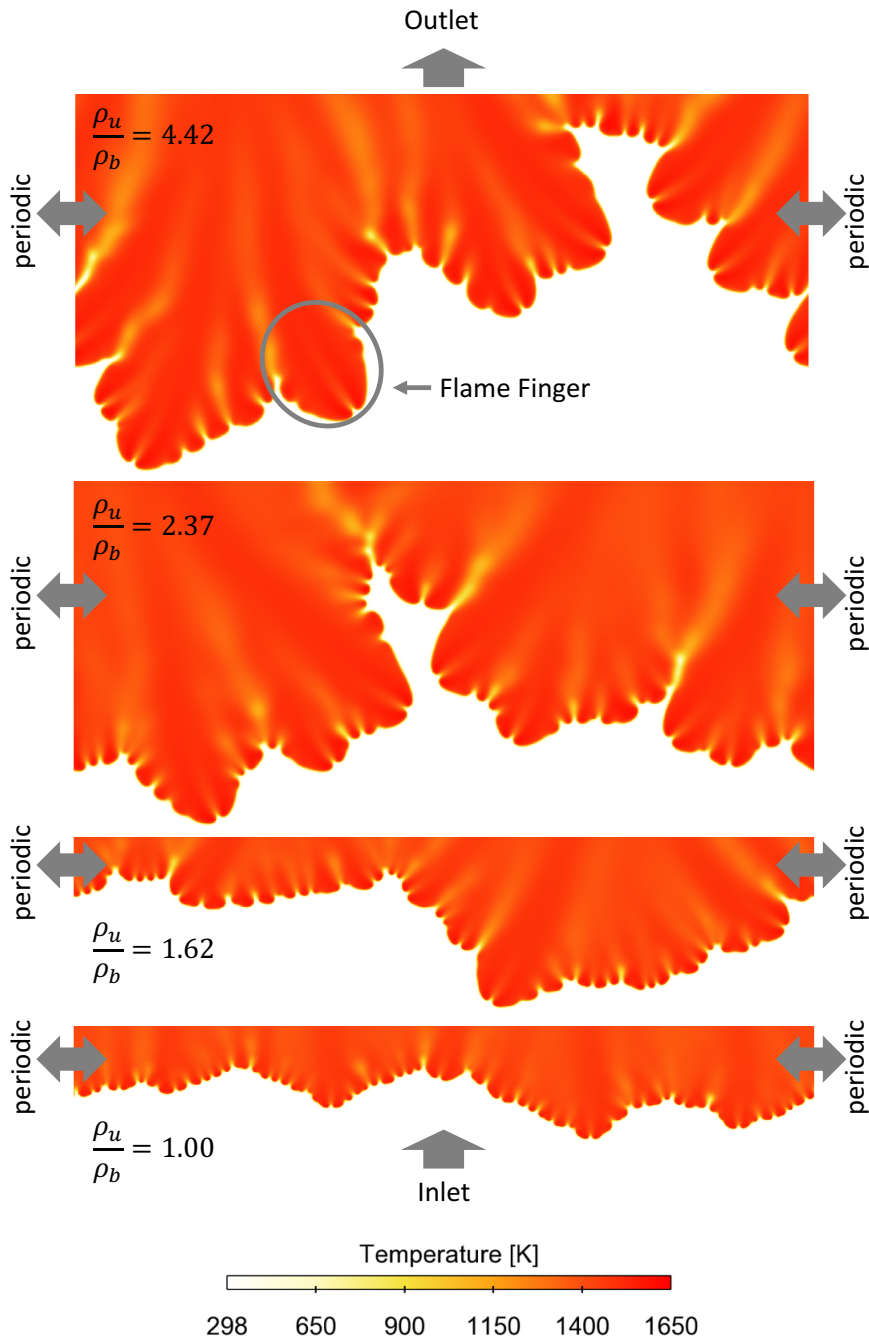
Figure 1. Thermodiffusively unstable premixed, lean hydrogen/air flames at $\phi = 0.4$ and $T_u = 298K$ for varying thermal expansion ratios. The simulation domains are cropped in streamwise direction to fit four of the five flames.

scales of the flame front wrinkles is found to increase with increasing thermal expansion ratio. Assuming the smallest length scale remains constant with respect to the laminar flame thickness, Fig. 2 quantifies that the size of the flame fingers is amplified by the thermal expansion ratio. However, the assumption of a constant smallest length scale still needs to be proven as well as the differing behavior of the constant density case requires further investigation.
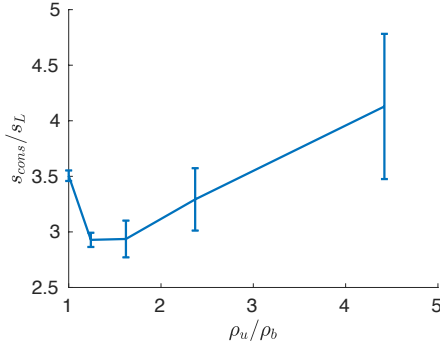

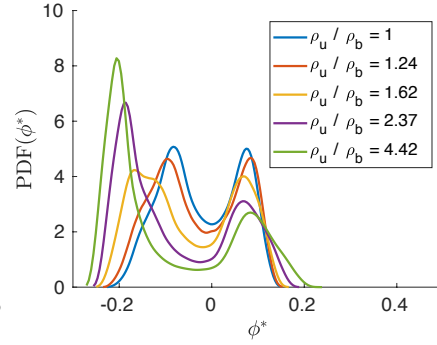**Figure 2.** Consumptions speed versus thermal expansion ratio.

Figure 3. PDF of the local mixture fraction variation expressed by ϕ^*.

In a second step, the intrinsic flame instabilities are assessed by means of the local variations of the composition of the mixture along the flame front. For thermodiffusively unstable flames, a flame front segment that is convexly curved towards the unburnt contains richer mixtures due to the differential diffusion of hydrogen. This induces a local acceleration of the flame and vice versa for concavely curved flame front segments. The local stoichiometry of the flame is determined by the local mixture fraction [9,10]

$$Z = \left(\frac{Z_H}{\nu_{H_2} M_{H_2}} - \frac{Z_O - Y_{O_2,Air}}{\nu_{O_2} M_{O_2}}\right) \Big/ \left(\frac{1}{\nu_{H_2} M_{H_2}} + \frac{Y_{O_2,Air}}{\nu_{O_2} M_{O_2}}\right), \tag{3}$$

where Z_k refers to the element mass fraction of element k, $Y_{O_2,Air}$ is the mass fraction of O_2 in air, the molecular weight of species k is denoted by M_k, and $\nu_{H_2} = 1$ and $\nu_{O_2} = 0.5$ are the stoichiometric coefficients. For convenience, the mixture fraction is expressed in terms of a local equivalence ratio ϕ_{loc} according to

$$\phi_{loc} = \frac{Z}{1-Z} \cdot \frac{1-Z_{st}}{Z_{st}}, \tag{4}$$

where $Z_{st} = 0.028$ is the stoichiometric mixture fraction. Fig. 3 shows the time-averaged PDF of the normalized local equivalence ratio $\phi^* = (\phi_{loc} - \phi_{ref})/\phi_{ref}$ along the flame front. The flame front is defined by the isoline of 1000 K and ϕ_{ref} refers to ϕ_{loc} of an unstretched premixed flame at 1000 K. The PDFs of all cases possess two distinct peaks, which reflect the differential diffusion of hydrogen that leads to leaner and richer mixtures along the flame front. The local equivalence ratio fluctuates up to 25% along the flame front. The variations of local stoichiometry are found to become more pronounced with increasing thermal expansion ratio. For

increasing thermal expansion ratio, the lean peak moves to even leaner mixtures and less flame front segments exist that possess a stoichiometry characteristic of the unstretched flame ($\phi^* = 0$). Thus, differential diffusion and hence thermodiffusive instabilities are enhanced by the thermal expansion of the flame.

Conclusion
Five large-scale DNS have been conducted for a reference flame at an equivalence ratio of $\phi = 0.4$ and an unburnt temperature of $T_u = 298K$. A parameter variation with respect to the thermal expansion ratio of the flame enables a rigorous separation of the influence of the thermodiffusive and Darrieus-Landau instabilities on the flame front's evolution. In agreement with previous studies, two characteristic length scales of the flame front corrugations have been identified, which consistently appear throughout all simulations. Both scales are found to originate from the thermodiffusive instability. Additionally, the largest scale appears to grow when the thermal expansion ratio increases.

Acknowledgement

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