

On the effect of pressure on intrinsic flame instabilities in lean hydrogen-air mixtures – Part I: Detailed chemistry based direct numerical simulation

J. Hasslberger, P. Katzy and T. Sattelmayer
Lehrstuhl für Thermodynamik, Technische Universität München (TUM)
Garching, Germany

1 Introduction

The promoting effect of intrinsic flame instabilities on explosive combustion is generally known. Most detailed computational studies (e.g. from Denev & Bockhorn [1]) are limited to generic configurations however. In the present numerical investigation, direct qualitative and quantitative comparison with experimental data from a laboratory-scale explosion channel is provided. The ultimate goal of this work is to develop a CFD sub-grid model incorporating the effect of mentioned instabilities without resolving them. Such a model is urgently needed in the context of nuclear safety analysis [2]. To obtain a comprehensive picture of the phenomenon, a complementary methodological approach is employed at TUM, cf. Tab. 1. Part I of the study focuses on detailed chemistry based DNS whereas the experimental procedure is shown in Part II.

Characterized by Lewis numbers ($Le = a/D$ with thermal diffusivity $a = \lambda/(\rho c_p)$ and the diffusion coefficient D of the deficient reactant, i.e. hydrogen) clearly smaller than unity, lean hydrogen-air mixtures are particularly prone to intrinsic flame instabilities. An imbalance of heat conduction and species fluxes leads to the development of a cellular flame structure, known as the thermal-diffusive instability. Increased flame surface area and interconnected flame stretch effects significantly affect the flame speed. Experimental evidence suggests a strong influence of pressure on the phenomenon. However, a thorough quantification is currently missing. One difficulty arises from the fact that the thermal-diffusive instability is superimposed by the hydrodynamic Landau-Darrieus instability. Additional insight is thus gained by manipulation of the mathematical model to separate different effects.

2 Numerical model

The computational methodology is based on the unsteady, compressible, reactive Navier-Stokes equations. Encompassing the contribution of all chemical species, the conservation of mass is formulated

Table 1: Complementary explosion analysis methods employed at TUM

	Experiment	Simulation
Macroscopic	Conventional: Pressure transducer, Photo diodes	3D, URANS, Turbulent Flame speed Closure, CFX
Microscopic	Optical: Shadowgraphy, OH-PLIF	2D, DNS, Detailed chemistry, OpenFOAM

as

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_j} (\rho u_j) = 0. \quad (1)$$

Conservation of momentum in each Cartesian direction i is guaranteed by solving

$$\frac{\partial}{\partial t} (\rho u_i) + \frac{\partial}{\partial x_j} (\rho u_i u_j) = \frac{\partial}{\partial x_j} \underbrace{\left(\mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial u_m}{\partial x_m} \right) \right)}_{\tau_{ij}} - \frac{\partial p}{\partial x_i} + \rho g_i, \quad (2)$$

where g_i represents the gravitational acceleration. The viscous stress tensor τ_{ij} is calculated on the basis of Stokes' hypothesis, with δ_{ij} denoting the Kronecker delta. Linking the velocity gradients to the resulting shear stress, the dynamic viscosity μ is obtained from Sutherland's formula and other molecular transport properties like individual species diffusivities $D_k = \mu / (\text{Sc}_k \rho)$ are derived therefrom. Corresponding Schmidt numbers Sc_k are summarized in Tab. 2. Mixture heat conductivity λ is calculated by Eucken's formula. To compute the unsteady local composition of the multi-component mixture, $N - 1$ transport equations

$$\frac{\partial}{\partial t} (\rho Y_k) + \frac{\partial}{\partial x_j} (\rho Y_k u_j) = \frac{\partial}{\partial x_j} \left(\rho D_k \frac{\partial Y_k}{\partial x_j} \right) + \dot{\omega}_k \quad (3)$$

are solved for the mass fractions Y_k of N species. Y_N (excess species nitrogen here) finally results from the fact that all mass fractions must sum up to unity. Because of its wide validation range (pressure from 0.05 to 87 atm, temperature from 298 to 2700 K and equivalence ratio from 0.2 to 6), the chemical source term $\dot{\omega}_k$ is calculated by means of O'Conaire's detailed mechanism [4]. The original scheme, consisting of 9 species and 19 reversible elementary reactions, is additionally extended for the excited hydroxyl molecule OH^* following Kathrotia et al. [5]. In the context of this study, it is important to note that Arrhenius rate coefficients are only depending on temperature. However, pressure implicitly influences the reaction rate via the concentration of involved species. Energy conservation is formulated in terms of the sensible part h^s of the specific enthalpy $h = h^f + h^s$:

$$\frac{\partial}{\partial t} (\rho h^s) + \frac{\partial}{\partial x_j} (\rho h^s u_j) = \frac{Dp}{Dt} + \frac{\partial}{\partial x_j} \left(\lambda \frac{\partial T}{\partial x_j} \right) + \tau_{ij} \frac{\partial u_i}{\partial x_j} - \underbrace{\sum_{k=1}^N h_k^f \dot{\omega}_k}_{\dot{q}}. \quad (4)$$

Hence, the heat release rate $\dot{q}$ is obtained by scaling the reaction rate $\dot{\omega}_k$ with the enthalpy of formation h_k^f of each species k . The mathematical model is as slim as possible to incorporate the dominant effects. Soret effect, Dufour effect, radiation etc. are assumed to be less important and therefore neglected. The system of equations is closed by the ideal gas law $p = \rho RT$ as an equation of state. R represents the specific gas constant of the mixture.

The open-source CFD package OpenFOAM is used to solve the set of equations in a finite-volume framework. Since gas-dynamic effects like shock waves do not play a role in this investigation, robust pressure-velocity coupling is realized by the PIMPLE algorithm. Spatial discretization is second-order accurate for both convective and diffusive fluxes. Dynamic time-stepping with a maximum Courant number of 0.1 assures a stable and sufficiently accurate temporal discretization. Chemical source terms are linearized to avoid issues related to stiff Arrhenius chemistry. Turbulence closure of the governing equations is unnecessary since all relevant scales are resolved, cf. Sec. 3.

3 Computational setup

The two-dimensional rectangular domain features a height of 60 mm, i.e. identical to the laboratory-scale GraVent facility. The length of 325 mm is only a fourth of the real channel to save computational

Table 2: Individual species Schmidt numbers [3]

k	H	H ₂	O	O ₂	OH	OH*	H ₂ O	HO ₂	H ₂ O ₂	N ₂
Sc_k	0.17	0.28	0.64	0.99	0.65	0.65	0.77	0.65	0.65	0.87

Table 3: Case overview

No.	X_{H_2} [%]	p^0 [bar]	$n [\cdot 10^6]$	Sc_k	Method
1 (ref.)	13	1.01	12.19	Individual	DNS + PLIF
2	13	1.01	7.8	Individual	DNS
3	13	1.01	21.67	Individual	DNS
4	13	1.01	12.19	Identical ($Sc_k = 1.0$)	DNS
5	13	0.5	12.19	Individual	DNS
6	13	2.0	12.19	Individual	DNS + PLIF
7	13	0.7	12.19	Individual	DNS + PLIF

resources. A sufficiently high length-to-height ratio is still assured. Anyway, accurate reproduction of experimental flame speeds must not be expected since flame surface increases with flame radius r in 2D but r^2 in 3D (assuming spherical propagation). For the evaluation of the flame wrinkling factor Ξ (Eq. 5), this difference does not play a (significant) role though. Comparison with the experiment is primarily based on two-dimensional OH-PLIF images. Line-of-sight integration inherent to shadowgraphy images is avoided by this technique. A detailed description of the experimental setup and quantitative evaluation procedure is provided by Katzy et al. [6].

Thermal spark plug ignition is modeled by patching adiabatic flame temperature and chemical equilibrium mixture composition around the ignition kernel with a radius of 2.5 mm. Initial temperature is assumed to be 293 K. No-slip adiabatic walls are imposed at all boundaries. The default mesh density (12.19 mio. cells) is specified such that the in-flame structure is resolved by approximately 10 cells. Additional cell size variation allows to assess the grid dependency of the solutions. Regarding the neglected third channel dimension, the simulations must be characterized as pseudo-DNS. Turbulent phenomena are incorporated in principle but not necessarily correct in a quantitative way. Fortunately, flame-generated turbulence seems to be of inferior importance. Flame stretch effects (due to front curvature and aerodynamic strain) are naturally included as well. Massively parallelized computations on 1024 cores were executed on the SuperMUC high performance cluster, operated by the Leibniz Supercomputing Center in Garching (Germany).

A case overview with respect to hydrogen volume fraction X_{H_2} , initial pressure p^0 , cell count n and Schmidt numbers Sc_k is presented in Tab. 3. The examined hydrogen concentration of 13 % guarantees a strong influence of the thermal-diffusive instability ($Le \ll 1$) and is close to the safety-relevant lower detonation limit of 12.5 % as measured in the large-scale RUT facility [2].

4 Results and discussion

Variation of the Cartesian mesh cell size (50 μm , 40 μm , 30 μm) reveals nearly grid-independent solutions. A thorough grid convergence analysis will be supplied with the final paper.

Figure 1 shows the development of the cellular flame structure for varying initial pressure p^0 . Note that pressure build-up in the closed vessel is insignificant during the initial stage of flame propagation which is of prior interest here. The visualized flame surface is defined as the $T = 800$ K contour, i.e. roughly the average between initial temperature and adiabatic flame temperature. The cell size

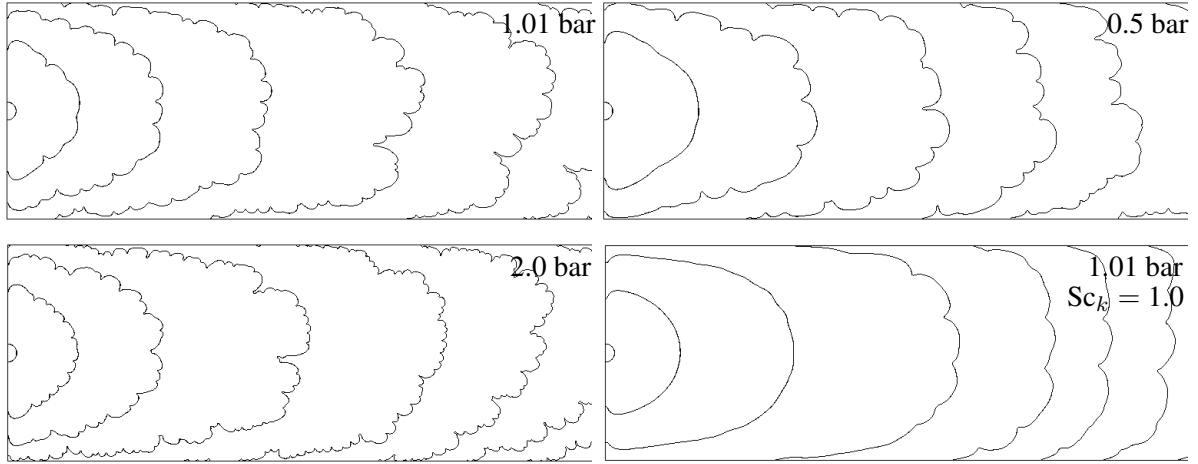


Figure 1: $T = 800$ K contour lines of case 1 (reference, $p^0 = 1.01$ bar, $\Delta t = 0.015$ s, top left), case 5 ($p^0 = 0.5$ bar, $\Delta t = 0.015$ s, top right), case 6 ($p^0 = 2.0$ bar, $\Delta t = 0.02$ s, bottom left) and case 4 (identical diffusivities, $p^0 = 1.01$ bar, $\Delta t = 0.015$ s, bottom right); Visualized sub-domain corresponds to the optically accessible part of the GraVent facility

obviously decreases with increasing pressure. Especially in the high pressure case, primary as well as secondary cells can be identified. In the bottom right frame, occurrence of the thermal-diffusive instability is intentionally extenuated by imposing identical species diffusivities ($Sc_k = 1.0$). Since the Prandtl number Pr is not directly specified (but rather indirectly given by NASA polynomials for c_p as well as Sutherland's and Eucken's formula), $Le_k = Sc_k/Pr = 1$ is not necessarily guaranteed in case 4. Nevertheless, visual inspection of the temperature and species fields suggests no straight occurrence of thermal-diffusive instability. Flame wrinkling starting at later stages might also be attributed to the hydrodynamic Landau-Darrieus instability.

Satisfying qualitative agreement with the experiment is evident from Fig. 2. Partial extinction in concave sections (due to hydrogen deficiency) is well reproduced by the simulation. This behavior manifests in the characteristic lower-temperature funnels originating from the concave sections. Temperature in convex sections is clearly above the adiabatic flame temperature. Due to the negligence of heat losses, wall quenching is not included in the simulation. Qualitative and quantitative comparison of computed OH and OH* fields is further insightful. The latter quantity can be interpreted as an indirect measure of heat release.

Related to large-scale explosion simulations, the idea is to incorporate the effect of unresolved flame instabilities via the three-dimensional flame surface wrinkling factor $\Xi_{3D} = A_{\text{wrinkled}}/A_{\text{smooth}} \approx s_t/s_l$. As required in the context of the Turbulent Flame speed Closure (TFC) approach, the effective burning velocity s_t can then be calculated from the well-known laminar burning velocity s_l . The proposed ansatz

$$\Xi_{3D} = \Xi_{2D}^2 = f(Le, p) = f(Le) \cdot f(p) = f(Le) \Big|_{p_{\text{ref}}} \cdot \left(\frac{p}{p_{\text{ref}}} \right)^{\beta_{\Xi}} \quad (5)$$

assumes direction-independent flame wrinkling and that there is no cross-correlation between the Lewis number and pressure influence. The effect of turbulence is not considered here. Evaluation of the pressure exponent $\beta_{\Xi} = 0.18$ (using $p_{\text{ref}} = 1.01$ bar) from both DNS and OH-PLIF data is presented in Part II of the study.

The flame surface length L in 2D simulations for varying initial pressure p^0 and identical species diffusivities is depicted in Fig. 3. Whereas the general shape of the curves is similar, the absolute level is

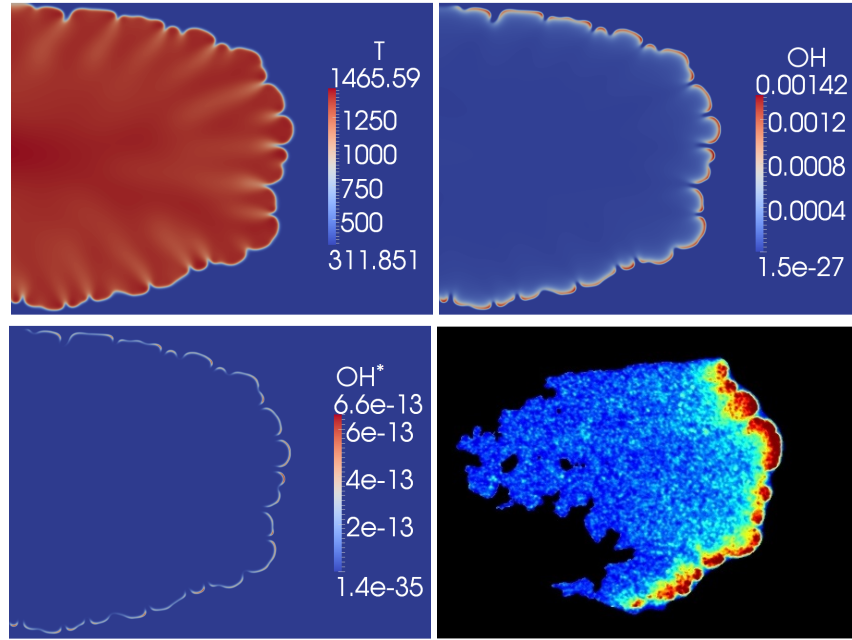


Figure 2: Case 1 (reference, $p^0 = 1.01$ bar): Simulated fields of temperature (top left), OH mass fraction (top right) and OH* mass fraction (bottom left) at 45 ms after ignition, compared to experimental OH-PLIF image (bottom right) at a similar stage

clearly shifted. On that basis, the two-dimensional flame wrinkling factor can directly be approximated from $\Xi_{2D} = L/L_{\text{identical}}$. The difference between both curves, L and $L_{\text{identical}}$, is attributed to mentioned flame instabilities in a first approximation. According to Fig. 4, flame wrinkling increases with increasing pressure. To exclude the initial development phase, the evaluation is restricted to the quasi-stationary regime which ranges approximately from 80 ms to 150 ms after ignition. Note that Ξ_{2D} , not Ξ_{3D} , is also measured in the experiment. The tendency with respect to pressure is in good agreement.

5 Conclusion and outlook

Pressure variation does not only affect the burning velocity but also the cellular structure of the flame in sufficiently lean hydrogen-air mixtures. The higher the pressure, the smaller the cell size. Flame surface area increases accordingly. The diminishing effect of elevated pressure on (laminar) burning velocity is a standard in modeling whereas the promoting effect of enhanced flame wrinkling via intrinsic instabilities is usually neglected. Burning laws without such correction might lead to an underestimation of flame speed for under-resolved explosion simulations.

Further investigation is required on the role of the hydrodynamic Landau-Darrieus instability which superimposes the thermal-diffusive instability. It might be advisable to separate both effects in terms of sub-grid modeling. Finally, the newly developed pressure correction has to be validated by means of under-resolved URANS simulations.

References

- [1] Denev J and Bockhorn H. (2013). Turbulent Burning Velocity in Premixed Combustion. 26th Deutscher Flammentag, Duisburg, Germany.

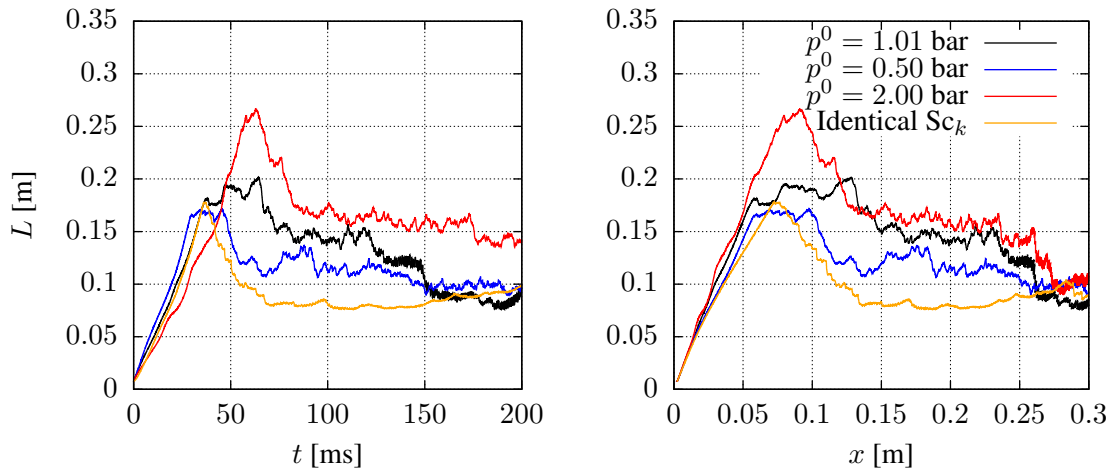


Figure 3: Temporal (left) and spatial (right) evolution of the flame surface length L in the simulation

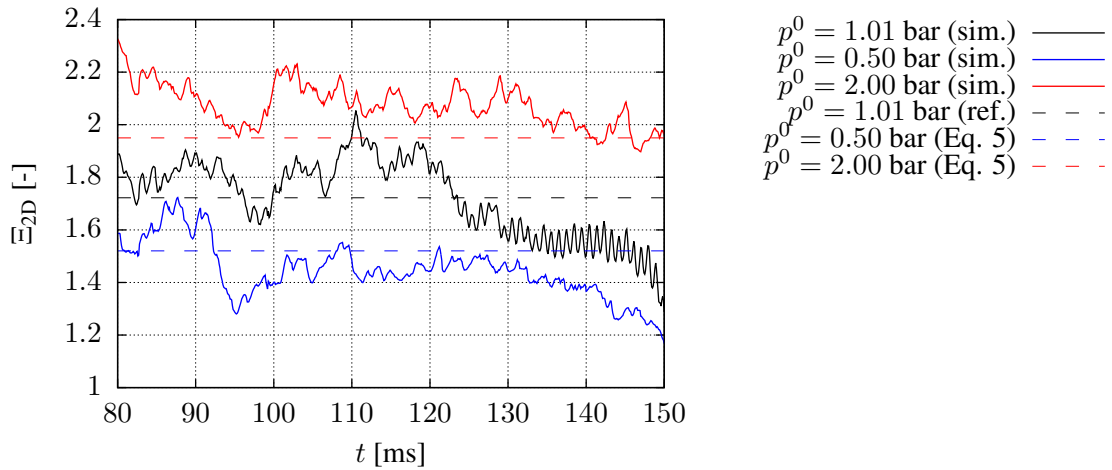


Figure 4: Temporal evolution of the two-dimensional flame wrinkling factor Ξ_{2D} in the simulation, compared to experimentally correlated values (Eq. 5)

- [2] Breitung W, Chan C, Dorofeev S, Eder A, Gerland B, Heitsch M, Klein R, Malliakos A, Shepherd J, Studer E and Thibault P. (2000). Flame Acceleration and Deflagration-to-Detonation Transition in Nuclear Safety. Tech. rep. OECD/NEA/CSNI/R 7.
- [3] Makowka K. (2015). Numerically Efficient Hybrid RANS/LES of Supersonic Combustion. PhD thesis. Technical University of Munich.
- [4] O’Conaire M, Curran H, Simmie J, Pitz W and Westbrook C. (2004). A Comprehensive Modeling Study of Hydrogen Oxidation. International Journal of Chemical Kinetics 36(11): 603–622.
- [5] Kathrotia T, Fikri M, Bozkurt M, Hartmann M, Riedel U and Schulz C. (2010). Study of the H+O+M Reaction Forming OH*: Kinetics of OH* Chemiluminescence in Hydrogen Combustion Systems. Combustion and Flame 157(7): 1261–1273.
- [6] Katzy P, Boeck L, Hasslberger J and Sattelmayer T. (2016). Application of High-Speed OH-PLIF Technique for Improvement of Lean Hydrogen-Air Combustion Modeling. 24th International Conference on Nuclear Engineering. Charlotte, NC, USA.