Numerical Investigation of the Detonation Cell Size in Hydrogen and Hydrocarbons with Complex Multi-step Kinetics

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1 Introduction

Detonations hold significant promise for enhancing the efficiency of propulsion devices and energy conversion systems when compared to the conventional combustion technologies [1]. A defining characteristic of a multi-dimensional detonation front is its inherent instability. The spatiotemporal evolution of this instability manifests itself as a cellular structure formed on the walls of the channel, in which a detonation propagates. Prior studies showed that numerical models, which use realistic multi-step chemical reaction kinetics, are not able to reproduce the cell sizes in hydrogen mixtures for practically relevant conditions [2]. In this work, detonation structure is analyzed primarily by means of the detonation cell sizes for a range of initial pressures and temperatures and for a wide variety of fuels, from hydrogen to simple hydrocarbons, such as methane, and to heavy hydrocarbons, such as JP10 jet fuel. More specifically, mixtures and thermodynamic conditions are chosen to cover a wide range of effective activation energies and specific heat ratios, which have been previously shown to be the primary thermochemical characteristics determining the cell regularity. Particular focus is placed on exploring the cell sizes at atmospheric conditions to contrast the vast majority of existing studies in the literature, which predominantly consider lower pressures [3]. Finally, numerically obtained cell sizes are compared with experimental values.

2 Numerical Setup

The unsteady reactive Navier-Stokes equations are numerically solved on a structured, adaptively refined grid using the massively parallel code `Athena-RFX++`. This code employs a directionally unsplit Van Leer finite-volume method with piecewise linear reconstruction for spatial discretization [4]. The HLLC-ADC Riemann solver is employed at cell interfaces to mitigate the occurrence of carbuncles. The stiff system of equations governing chemical kinetics is integrated using the semi-implicit YASS solver [5]. The chemical source terms are coupled to the hydrodynamic flow equations through an operator splitting approach with the global time-step control [6]. The numerical scheme is second-order accurate in both space and time. Reduced multi-step chemical kinetics is based on the FFCM-1 and HyChem families of reaction mechanisms [7]. Finally, the physical model includes detailed treatment of the multi-species transport based on the mixture averaging approximation [8].

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Table 1: Summary of mixtures considered and the corresponding parameters.

Case	Mixture	ϕ	Pressure (bar)	Temperature (K)	D_{CJ} (km/s)	ϵ	γ_{vn}	χ
1a	H_2-O_2-0.12036O_3	1.500	0.150	295	3.05	3.66	1.32	0.80
1b	2.965C_2H_2-2.5O_2-7.52N_2	2.965	1.013	293	2.00	3.88	1.23	1.50
2a	2H_2-O_2	1.000	0.159	298	2.74	5.39	1.32	2.53
2b	3C_2H_4-3O_2	3.000	0.100	298	2.59	5.72	1.17	1.89
3a	2H_2-O_2-3.76N_2	1.000	1.000	298	1.98	7.56	1.32	5.24
3b	$C_{10}H_{16}$-14O_2-10.5N_2	1.000	1.000	353	2.11	7.76	1.17	10.50
4a	H_2-O_2-3.76N_2	0.500	0.239	298	1.61	8.78	1.33	7.51
4b	1.4$C_{10}H_{16}$-14O_2-52.4N_2	1.400	1.000	353	1.83	9.69	1.23	16.2
4c	C_2H_4-3O_2-11.28N_2	1.000	1.000	298	1.83	10.51	1.13	8.36
4d	CH_4-2O_2	1.000	1.200	293	2.40	11.35	1.17	45.90
5a	CH_4-2O_2-7.52N_2	1.000	1.013	293	1.81	14.81	1.26	185.00
5b	H_2-O_2-3.76N_2	0.500	1.013	298	1.62	37.71	1.33	155.00

In all calculations, the computational domain has the spatial extent of $4\lambda_{exp} \times 40\lambda_{exp}$, where λ_{exp} is the experimentally measured detonation cell length. Such domain width is found to be sufficient to prevent mode-locking of the detonation structure, while the large domain length allows the front to relax from any initial perturbations associated with the detonation initiation. Detonation is ignited by placing a perturbed ZND detonation profile on the grid at $t = 0$. The ZND induction length, Δ_i, is spatially resolved with a minimum of 40 numerical cells, and the thermal width of the ZND profile, Δ_r, is resolved with a minimum of 5 numerical cells in all calculations discussed here. Simulations are performed in the laboratory reference frame. Boundary conditions are adiabatic, slip walls in the transverse direction, and the outflow (zero-order extrapolation) in the streamwise direction.

3 Results and Discussion

Simulations were performed for twelve different mixtures for a range of pressures and temperatures. Details of the mixture compositions, thermodynamic conditions, and other relevant parameters are listed in Table 1, where ϕ is the equivalence ratio, D_{CJ} is the Chapman-Jouguet detonation speed, and

$$\epsilon = \frac{E_a}{RT_{VN}} = \frac{1}{T_{VN}} \frac{\partial \ln \tau}{\partial (1/T)} \tag{1}$$

is the normalized effective activation energy based on the logarithmic temperature sensitivity of the ignition delay time, τ. Here, T_{VN} and γ_{VN} are the temperature and the ratio of specific heats, respectively, at the von Neumann (post-shock) state. The parameter χ, previously suggested to control cell regularity [9], is defined as

$$\chi = \epsilon \frac{\dot{\sigma}_{max}}{u_{CJ}}. \tag{2}$$

All cases considered can be divided into five sub-groups described below and also shown in Fig. 1.

Low ϵ: In this regime (Cases 1a and 1b), ϵ is small, typically $\lesssim 4-5$. For an H_2-O_2-O_3 detonation at low pressure (Case 1a), which is representative of this regime, numerical simulation gives cells with the correct size compared to experiments (see Fig. 2a) [3]. The cells are regular since ϵ is low and at the same time γ_{VN} is high. In contrast, for the atmospheric C_2H_2 case (Case 1b; see Fig. 2b), the numerical cells are $\sim 4-5$ times smaller than the experimental cells, with some degree of irregularity [10]. Case 1b has very similar ϵ to Case 1a (3.88 vs. 3.66), but lower γ_{VN} (1.23 vs. 1.32). While lower γ_{VN} is indeed expected to lead to more irregular cells [11], these two cases suggest that low temperature sensitivity of the mixture does not necessarily guarantee the correct detonation structure.

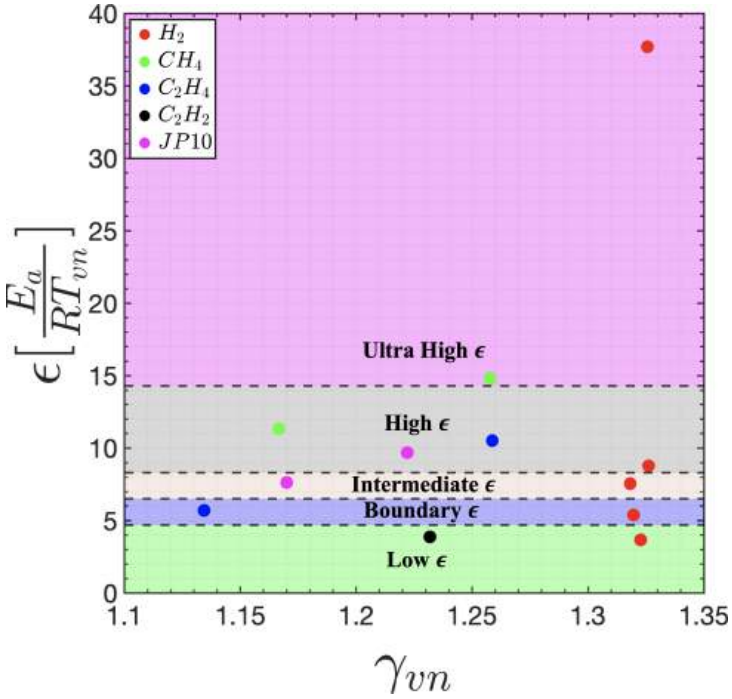

Figure 1: Regime diagram of the detonation conditions surveyed.

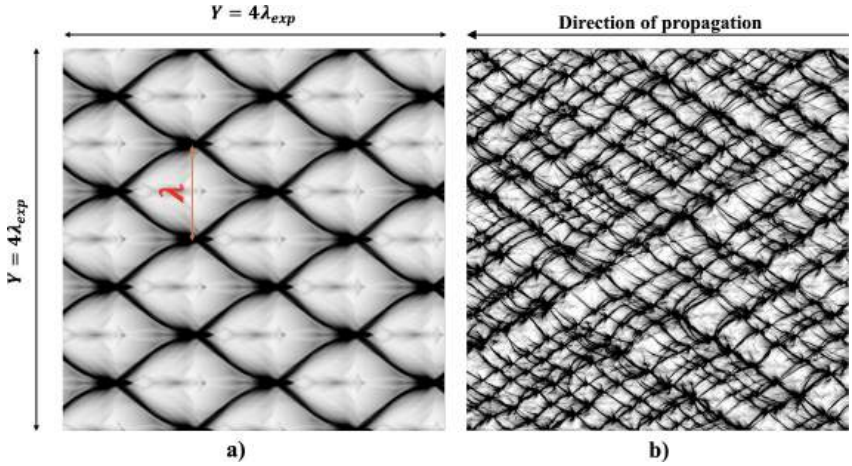

Figure 2: Numerical soot foils for a) Case 1a and b) Case 1b in the low-ϵ regime.

Boundary ϵ: In this regime (mixtures of H_2 and C_2H_4 with O_2 at low pressures; Cases 2a and 2b), ϵ is in the range $\approx 5-7$. Similar to the low-ϵ regime, numerical simulations can reproduce the experimental cell sizes for high γ_{VN} [12], though cells become more irregular (Case 2a). Also, similar to the Case 1b, for low γ_{VN} (C_2H_4, Case 2b), detonation cells are $\approx 8-10$ times smaller than the experimental values [13], and they are irregular.

Intermediate ϵ: This regime (atmospheric H_2-air and JP10; Cases 3a and 3b) is defined by ϵ in the range $\sim 7-8.5$. Here, detonation cells in the simulations are always smaller than the experimental values for all γ_{VN}. Figure 3a (Case 3a, stoichiometric, atmospheric H_2-air) shows that cells are irregular and $\sim 3-4$ times smaller than the experimental cells [14], in agreement with the previous observations by [15]. There can be seen $\sim 12-14$ detonation cells in the transverse direction instead of 4. For lower γ_{VN}, (Case 3b, JP10, Fig. 3b) the disagreement is much more pronounced with cells being smaller by a

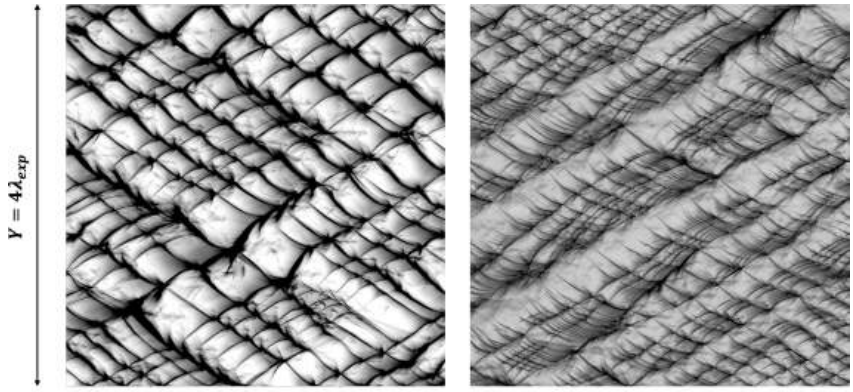

Figure 3: Numerical soot foils for a) Case 3a and b) Case 3b in the intermediate-ϵ regime.

factor of $\sim 8 - 10$ compared to experiments [16], and they are significantly more irregular.

High ϵ: In this regime (Cases 4a - 4d), ϵ ranges between ~ 8.5 and 14.5. At such high temperature sensitivity, the disagreement between the experimental and numerical cells becomes even more pronounced. The detonation front undergoes periodic quasi-failures and re-ignitions, which result in a wide range of cell sizes. For the high γ_{VN} (Case 4a, lean, low-pressure H_2-air), the cells are 8-10 times smaller than in the experiments [17], and for low γ_{VN} (Case 4c, ethylene) cells are $10 - 12$ times smaller.

Ultra-high ϵ: Finally, at the highest values of $\epsilon \gtrsim 14.5$ (see Fig. 4, atmospheric H_2 and CH_4 with air; Cases 5a and 5b), cells are not only extremely small but also they become difficult to identify in large regions of the soot foil. For both high and low γ_{VN}, the cells can be $\gtrsim 20$ times smaller than the experimental values [18, 19]. In this regime, in particular in the atmospheric CH_4-air case, quasi-failures and re-ignitions become pervasive, dominating the front dynamics (Fig. 4b) and making identification of cells and their size possible only in select regions of the flow.

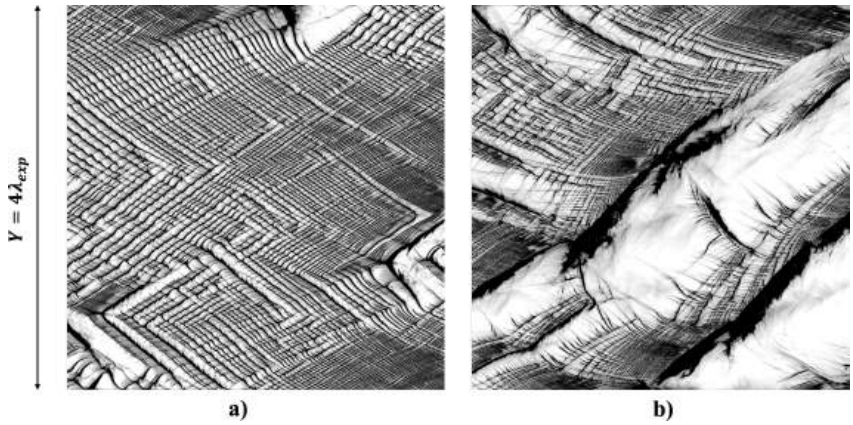

Figure 4: Numerical soot foils for a) Case 5a and b) Case 5b in the ultra-high ϵ regime.

4 Conclusions

We presented a series of numerical two-dimensional simulations of planar, freely propagating detonations in rectangular channels with adiabatic, slip walls. All simulations use multi-step chemistry with full transport for different initial conditions and fuel types. We confirm prior observations that numerical simulations can reproduce the experimental cell size for hydrogen only under very specific conditions,

i.e., when the effective activation energy ϵ is small and simultaneously γ_{VN} is high, in particular in low-pressure H_2-O_2 or H_2-O_2-O_3 mixtures. For all other fuels surveyed, including H_2-air mixtures at atmospheric conditions and all hydrocarbon mixtures both at low and atmospheric pressures, simulations are not able to reproduce experimental cell sizes, even when the effective activation energy is low. In particular, the errors in cell sizes range from a factor of $3-4$ up to more than an order of magnitude. The cause of these discrepancies could potentially be due to the uncertainties in the chemical model, thermal non-equilibrium, effects of wall losses, or insufficient numerical resolution. These effects are analyzed in a separate paper.

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References

[1] K. Kailasanath. "Review of propulsion applications of detonation waves". In: *AIAA journal* 38.9 (2000), pp. 1698–1708.

[2] B. Taylor, D. Kessler, V. Gamezo, and E. Oran. "The Influence of Chemical Kinetics on the Structure of Hydrogen-Air Detonations". In: *50th AIAA Aerospace Sciences Meeting including the New Horizons Forum and Aerospace Exposition*. Vol. 23. 2012, p. 470.

[3] J. Crane, X. Shi, A.V. Singh, Y. Tao, and H. Wang. "Isolating the effect of induction length on detonation structure: Hydrogen–oxygen detonation promoted by ozone". In: *Combustion and Flame* 200 (2019), pp. 44–52.

[4] T.A. Gardiner and J.M. Stone. "An unsplit Godunov method for ideal MHD via constrained transport in three dimensions". In: *Journal of Computational Physics* 227 (2008), pp. 4123–4141.

[5] A. Khokhlov, J. Austin, F. Pintgen, and J. Shepherd. "Numerical study of the detonation wave structure in ethylene-oxygen mixtures". In: *42nd AIAA Aerospace Sciences Meeting and Exhibit*. 2004, p. 792.

[6] G. Strang. "On the construction and comparison of difference schemes". In: *SIAM Journal on Numerical Analysis* 5.3 (1968), pp. 506–517.

[7] G.P. Smith, Y. Tao, and H. Wang. "Foundational fuel chemistry model version 1.0 (FFCM-1)". In: *epub, accessed July* 26 (2016), p. 2018.

[8] A. Ern and V. Giovangigli. *Multicomponent transport algorithms*. Vol. 24. Springer Science & Business Media, 1994.

[9] H.D. Ng, M.I. Radulescu, A.J. Higgins, N. Nikiforakis, and J.H.S. Lee. "Numerical investigation of the instability for one-dimensional Chapman–Jouguet detonations with chain-branching kinetics". In: *Combustion Theory and Modelling* 9.3 (2005), pp. 385–401.

[10] R Knystautas, C Guirao, J.H. Lee, and A. Sulmistras. "Measurement of cell size in hydrocarbon-air mixtures and predictions of critical tube diameter, critical initiation energy and detonability limits". In: *Progress in Astronautics and Aeronautics* 94 (1984), pp. 23–37.

[11] P.A. Meagher, X. Shi, J.P. Santos, N.K. Muraleedharan, J. Crane, A.Y. Poludnenko, H. Wang, and X. Zhao. "Isolating gasdynamic and chemical effects on the detonation cellular structure: A combined experimental and computational study". In: *Proceedings of the Combustion Institute* 39.3 (2023), pp. 2865–2873.

[12] H.O. Barthel. "Predicted spacings in hydrogen-oxygen-argon detonations". In: *The Physics of Fluids* 17.8 (1974), pp. 1547–1553.

[13] D.H. Lieberman and J.E. Shepherd. "Shock Wave Induced Mixing and Reaction". In: *20th International Colloquium on the Dynamics of Explosive and Reactive Systems, Montreal, Canada, July*. Vol. 19. 2015.

[14] G. Ciccarelli, T. Ginsberg, and J.L. Boccio. "Detonation cell size measurements in high-temperature hydrogen-air-steam mixtures at the bnl high-temperature combustion facility". In: *Technical report US Nuclear Regulatory Commission (NRC)* (1997).

[15] B. Taylor, D.A. Kessler, and E.S. Oran. "Estimates of vibrational nonequilibrium time scales in hydrogen-air detonation waves". In: *24th International Colloquium on the Dynamics of Explosive and Reactive Systems, Taipei, Taiwan, July*. Vol. 23. 2013, p. 44.

[16] J.M. Austin and J.E. Shepherd. "Detonations in hydrocarbon fuel blends". In: *Combustion and flame* 132.1-2 (2003), pp. 73–90.

[17] S.R. Tieszen, D.W. Stamps, C.K. Westbrook, and W.J. Pitz. "Gaseous hydrocarbon-air detonations". In: *Combustion and Flame* 84.3-4 (1991), pp. 376–390.

[18] S.R. Tieszen, M.P. Sherman, W.B. Benedick, J.E. Shepherd, and R. Knystautas. "Detonation cell size measurements in hydrogen-air-steam mixtures". In: *Dynamics of explosions* (1986), pp. 205–219.

[19] R.K. Zipf Jr, V.N. Gamezo, M.J. Sapko, W.P. Marchewka, K.M. Mohamed, E.S. Oran, D.A. Kessler, E.S. Weiss, J.D. Addis, F.A. Karnack, and D.D. Sellers. "Methane–air detonation experiments at NIOSH Lake Lynn Laboratory". In: *Journal of Loss Prevention in the Process Industries* 26.2 (2013), pp. 295–301.