Analysis of Flame Propagation and Detonation Characteristics in Hydrogen-Oxygen and Ammonia-Oxygen Mixtures

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Abstract
This work investigates detonation in a microdevice for ammonia-hydrogen-oxygen mixtures at 10 bar. The ZND model and the Navier-Stokes equations with skeletal mechanisms are solved. The results are in good agreement with experimental data, where available. Temperature, Chapman-Jouguet parameters, and cellular size are correlated to hydrogen concentration, revealing a clear functional relationship. In flames with low hydrogen composition, the cell size increases and the CJ velocity decreases. In the 2D simulation, starting from a hydrogen/oxygen products hotspot, the time and space to detonation are analyzed, showing good agreement with ZND model. For pure ammonia/oxygen composition, flame does not propagate.Ammonia is a promising compound for the chemical storage of renewable energy produced from non-continuous sources. However, the low reactivity of ammonia necessitates the use of ammonia–hydrogen blends as a fuel for combustion applications.

Introduction
Combustion can manifest in various modes, including laminar flames, deflagrations, and detonations. Deflagrations involve subsonic expansion waves that propagate through the diffusion of heat and mass from the reaction zone to ignite the unburned gases. This burning rate leads to increased turbulence in the unburned gases, establishing a positive feedback loop between turbulence and the reaction zone. Additionally, the flame front interacts with walls and obstacles, further enhancing acceleration. At the same time, the reaction zone is quenched due to flame stretch and mixing of cold reactants with hot products, imposing an upper limit on flame acceleration and finding an equilibrium in deflagration. This is why the interest in Deflagration-to-Detonation Transition (DDT) in micro/meso devices has led to the utilization of hydrogen as an essential fuel component. Without the presence of hydrogen, avoiding flame quenching and the cessation of propagation remain undocumented in the literature. Ignition and flame stabilization processes in supersonic combustion microdevices significantly differ from those observed in the subsonic regime [1]. Flames generate acoustic waves that propagate into the unburned mixture at the local speed of sound. These waves may converge and form precursor shocks, thereby pre-compressing the reactant mixture and elevating its temperature. The fundamental arrangement of a stable detonation wave can be elucidated through the Zeldovich-von Neumann-Döring (ZND) model, comprising

a leading shock and a subsequent reaction zone. Within this framework, the lowest attainable detonation velocity is identified as the Chapman-Jouguet (CJ) speed [2]. Despite the ZND model assumption of a stable detonation wave, empirical and computational investigations have demonstrated that actual detonation waves exhibit instability under flow disturbances. Another issue is the selection of an appropriate kinetic model. On one hand, there's the need to retain information about radicals.On the other hand, a highly detailed mechanism would make simulations computationally heavily demanding. This study aims to shed new light on the detonation of mixtures at high pressure with blends of ammonia and hydrogen, an area where the literature on explosions and detonation is largely unexplored. Flame acceleration and transition to detonation in $NH_3/H_2/O_2$ mixture at atmospheric condition was studied by Zhu et al [3]. This research focuses on investigating detonation within a microdevice containing H_2 / O_2 and NH_3-H_2-O_2 mixtures at 10 bar. The primary objectives include correlating temperature, CJ parameters, and cellular size with hydrogen/ammonia concentration to understand their relationships. Additionally, the research will examine the impact of varying blend compositions on cell size and CJ velocity. Preliminary 2D simulations, starting from a hotspot, will analyze the time and space required for H_2 detonation and its 2D cellular structure.

Methodology

To compute the DDT parameters, we employ a two-pronged approach using both 2D and 1D simulations. For the 2D simulations, we utilize OpenFoam 6 with detonationFoam and Adaptive Mesh Refinement (AMR) technology [4], [5]. For 1D computations, we utilize the Shock and Detonation Toolbox, a collection of MATLAB scripts and functions, fo CJ parameter forecasting and 1D computations. The toolbox is used to calculate CJ velocities, pressure, and temperature for the given fuel mixtures, which are crucial for understanding the theoretical detonation characteristics and serve as a benchmark for 2D simulation results. The 1D computations involve solving the ZND equations to obtain detailed profiles of detonation wave structure, including shock front, reaction zone, and energy release rate. The results from 1D computations provide a foundational understanding of the detonation process, which is compared and validated against the more complex 2D simulations performed with Detonationfoam. An adiabatic duct of 0.9 mm width is the test case, already analyzed by Ramachandran et al. [6] with the PeleC code. A symmetry boundary condition is applied to half the computation cost, knowing the symmetrical flame behavior. Pressure is equal to 10 bar, and the unburned gas is at the temperature of 503 K at stoichiometric conditions. At the beginning of the simulation, the flame front is a y = 1 mm with a temperature of 3075 K for pure hydrogen and 2773 for ammonia blends test cases. 2D simulations with pure ammonia/oxygen test case shows no propagation, proving the importance of hydrogen blending in DDT ammonia mixtures. In Table 1, molar fractions for different mixtures simulated in the paper are provided. The duct is divided into 10^4x16 cells. A Courant number of 0.55 is applied.

Table 1. Molar fractions for different test cases. P = 10 bar, T = 503 K

Test case	*NH_3*	*H_2*	*O_2*	*Test case*	*NH_3*	*H_2*	*O_2*
m1	0.4706	0.1176	0.41	*m5*	0.16	0.48	0.36
m2	0.4	0.2	0.4	*m6*	0.0816	0.5714	0.3469
m3	0.3077	0.3077	0.3846	*m7*	0	0.667	0.333
m4	0.2105	0.4211	0.368				

Bykov et al. mechanism has been used [7]. Ignition delay time and CJ velocity were predicted and compared with experiments respectively in [8] and in [9] in Figure 1.

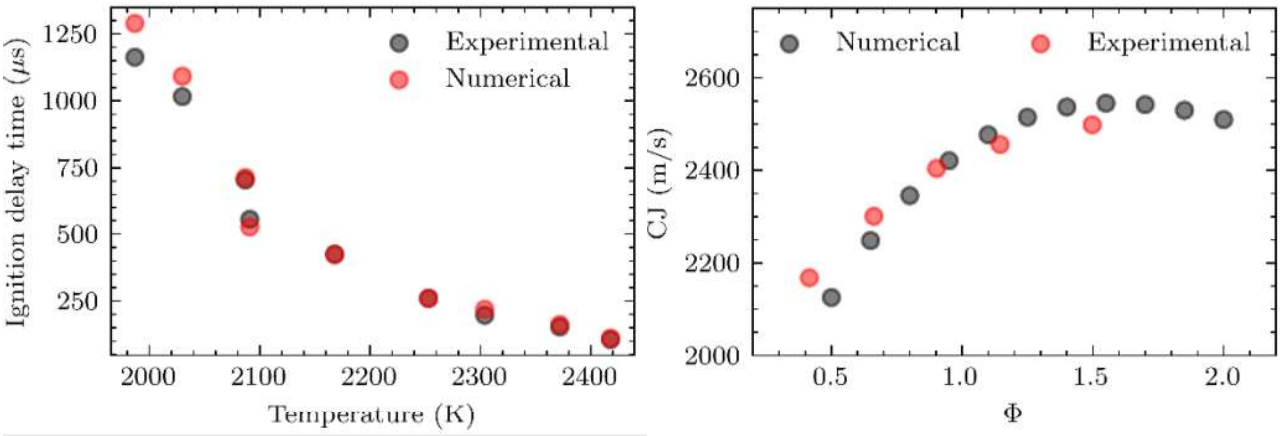
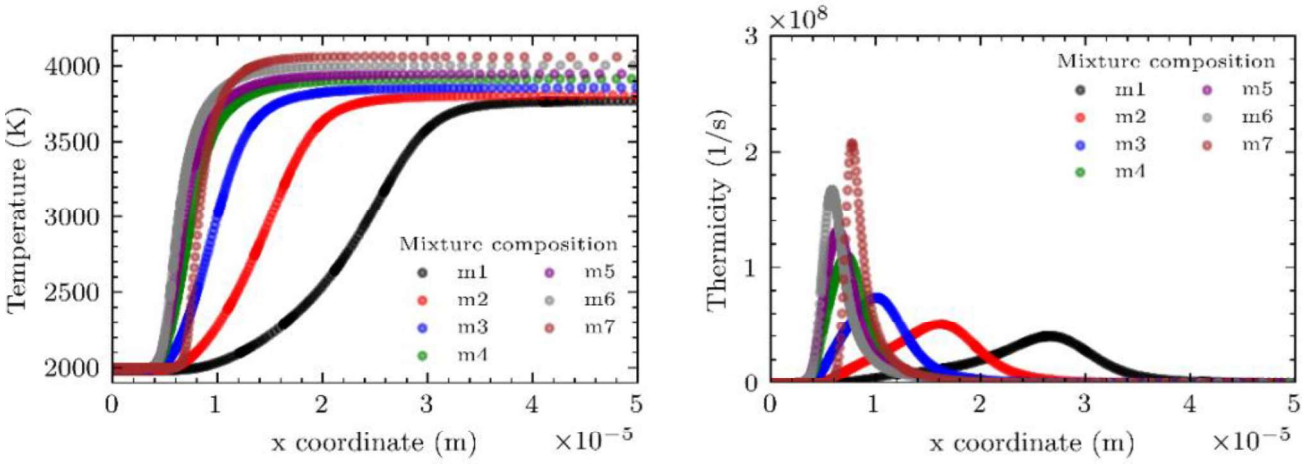


Figure 1. a) IDTs of 0.005715 NH_3/ 0.004285 O_2 / 0.99Ar mixture with p=11 atm [8], b) CJ velocity for NH_3/ O_2, Φ = 1, T=295 K [9].

Results

The Shock & Detonation Toolbox is used to predict the $NH_3/H_2/O_2$ ZND structures at p_0 = 10 atm consistent with the initial condition in Table 1. Figures 2a and 2b illustrate the spatial distribution of temperature and thermicity (heat release beyond the shock) from the ZDN structure (the shock is positioned at x=0 with a velocity corresponding to the CJ value for each mixture). Across the shock wave, the temperature sharply jumps from 2000 K to over 4000 for the pure H_2/O_2 mixture, and to 3700 K for the mixture with the lowest H2 fraction (m1). The heat release is primarily located around 10 μm for most conditions. For NH_3 molar concentrations greater than 0.3077, the peak of thermicity shifts (see Figures 2a-2b. Specifically, for the m1 mixture, this peak occurs at x=28 μm.



a) b)

Figure 2. Distributions of a) temperature, b) thermicity for different mixture compositions (read Table 1).

In Figure 3, CJ velocity and cellular size (λ) with Ng et al. model correlation [10] are computed. CJ increases from 2495 m/s to 2835 m/s. The λ value decreases from 1 cm to 2.4 mm for m6 to the final value of 3.2 mm for pure H_2 case.

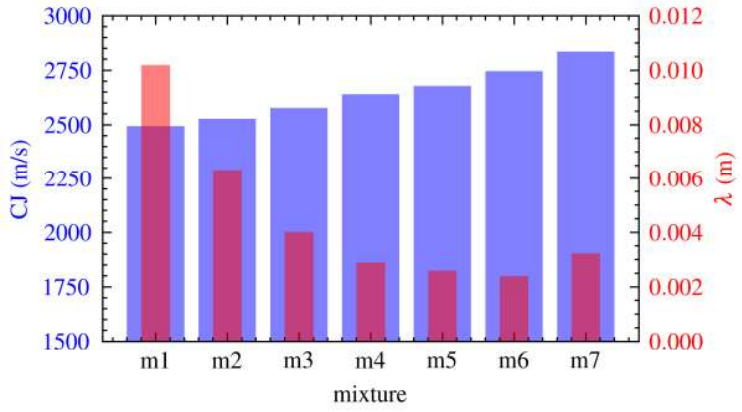
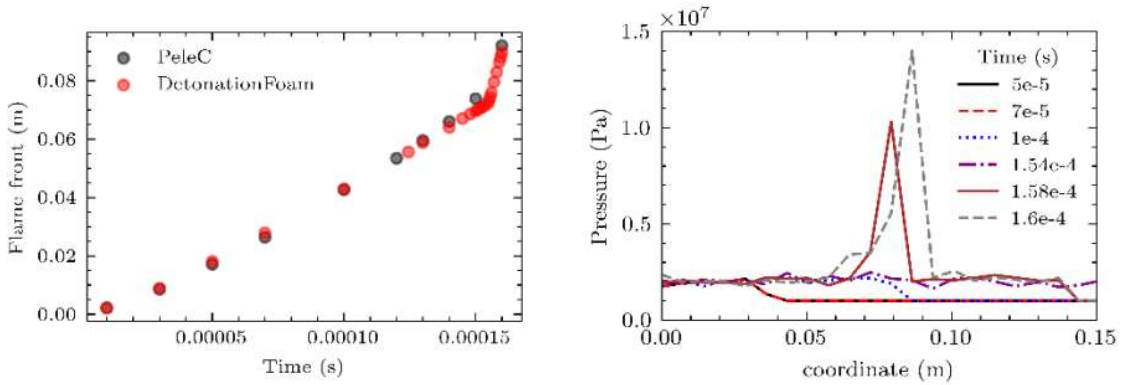


Figure 3. Computed CJ velocity and cell size as functions of NH_3/H_2 molar ratio a in stoichiometric NH3/H2/ O2 mixtures.

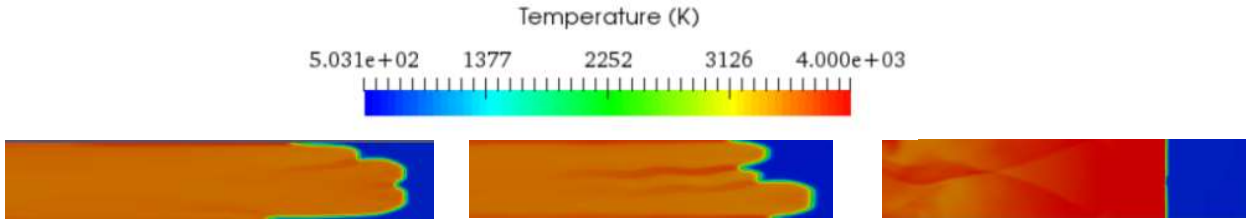
DDT 2-D results for m7 mixture are presented compared with PeleC solver simulation results performed in [6]. For hydrogen-oxygen DNS, Li et al. mechanism is used, already widely validated in literature for different pressure conditions [8]. The flame propagates at a velocity of 550 m/s, and the DDT initiates around 155 μs.Figure 4a illustrates the flame front propagation, which aligns well with previously reported results. Figure 4b depicts the axial pressure time sequence, showing the distribution and evolution of shock waves. The pressure in the detonation increases from 10 to 14 bar within $1.6e^{-4}$ s. The temperature increases from 3700 K to over 4000 K during detonation.



a) b)

Figure 4. 2D-time sequence of a) flame front position, b) pressure in the midline for m7 mixture.

Figure 5 depicts the shape evolution of the flame front from ignition to DDT.



t = 1.4e-4 s t = 1.5 e-4 s t = 1.555 e-4 s

Figure 5. DDT time sequence of temperature distributions at flame front for m7 mixture.

Figure 6 illustrates the detonation and the cellular structure of the phenomenon at t=1.555e^{-4} s. The cell size λ at the completed transition (t = 1.57e-4 s) measures 3.05 mm, which is in good agreement with the results shown in Figure 3.

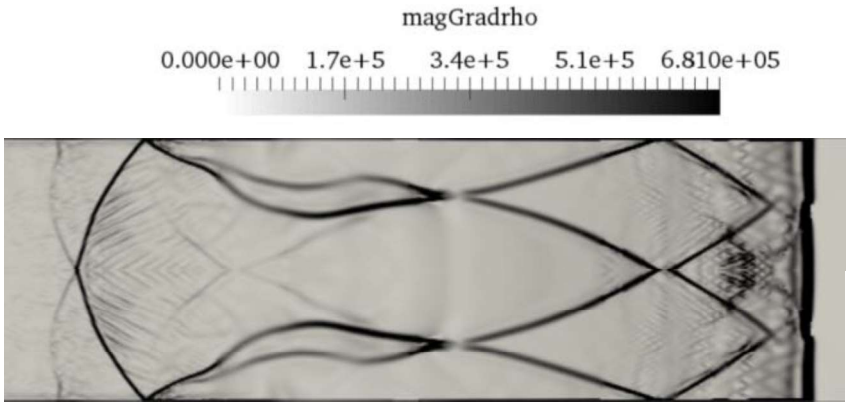


Figure 6. Numerical Schlieren visualization for m7 mixture at t = 1.555 e-4 s.

Conclusions

In this work, the characteristics of flame deflagration and detonation in a channel

filled with pure H_2/O_2 and $NH_3/H_2/O_2$ at 10 bar have been investigated. The fully compressible Navier-Stokes equations, coupled with the perfect gas equation of state, are solved using detonationFoam. The ZND model is analyzed using the Shock & Detonation Toolbox for seven different mixture compositions, providing Chapman-Jouguet (CJ) parameters, cellular sizes, and shock thermal characteristics. As H_2 concentration increases, the temperature and thermicity rise, and the shock width shifts to the left, shortening to a few micrometers. For the H_2/O_2 condition, a 2D DDT analysis reveals a detonation initiation time of 1.6e-4 seconds and a detonation distance of 0.089 meters. The cellular structure is confirmed with a size of 3 mm, which is in good agreement with ZND model predictions.

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