Detonation Propagation through a Gradient in Fuel Composition

D.A. Kessler, V.N. Gamezo, E.S. Oran Laboratory for Computational Physics and Fluid Dynamics Naval Research Laboratory, Washington, DC

1 Introduction

In uniform mixtures, detonation velocities increase and detonation cell sizes increase as mixture compostions deviate from stoichiometric [1, 2]. In systems where the concentration of fuel is nonuniform and the local equivalence ratio ϕ varies, the behavior of a detonation is not as simple. Both the propagation speed and the detonation cell size can vary with ϕ throughout the system. This can lead to complex behavior by the multiple shocks and reaction zones that make up a gas-phase detonation.

Consider the propagation of a detonation through two simple nonuniform mixtures, one in which the gradient of fuel concentration is parallel to the direction of propagation and one in which it is perpendicular. The former situation has been studied extensively, particularly for the problem of detonation transmission from a more to a less reactive mixture (see, for example, [3,4]). The latter case has thus far received far less research attention. Calhoon and Sinha [5] calculated the structure of a stable, two-dimensional detonation in the mixing layer of initially nonpremixed coflowing streams of fuel and air. The complex wave structure they computed is the high-speed analog to the triple flame found in low-speed mixing layers. The leading edge of the combustion wave is a curved detonation that varies from fuel-lean near the oxidizer stream to fuel-rich near the fuel stream. The excess reactants not consumed by this detonation combine downstream and react in a laminar diffusion flame. The curvature of the front is caused by the nonuniformity in the propagation speed of the detonation as a function of ϕ . Far from the line of stoichiometry, where the mixture is very fuel-rich or very fuel-lean, the propagation speed is too slow and the induction times are too large to support the detonation, and the reaction zone decouples from the leading oblique shock. Experiments in nonuniform hydrogen-oxygen [6] and ethylene-oxygen [7] mixtures exhibit characteristics similar to these curved detonation fronts.

Calhoon and Sinha [5] calculated the structure of *stable* detonations only and did not consider how transverse instabilities might affect their behavior. Ishii and Kojima found the detonation cells recorded in their experiments were larger and more distorted in less reactive regions of the mixture compared to the regular diamond-shaped cells formed in the near-stoichiometric regions of the mixture [6]. Kessler et al. [8] performed two-dimensional numerical simulations of detonations in various nonuniform methane-air mixtures and found their behavior to be quite complex. The computed cellular structures, however, were too irregular and incomplete to make a comparison with those recorded in experiments [6].

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The regularity of detonation cells computed using a single-step Arhenius reaction model is controlled by the effective activation energy of the mixture. As this quantity increases, the cellular structures become increasingly irregular and smaller secondary cellular structures form [9]. For large enough activation energies, there is no clear distinction between primary and secondary structures, and the large-scale cells that appear are incomplete and chaotic [10]. In this work, we begin to clarify how transverse instabilities affect the structure and dynamics of detonations in gas mixtures in which there is a gradient of mixture composition perpendicular to the direction of propagation by considering relatively low activation energies for which the complications associated with the formation of secondary cellular structures are not an issue.

2 Computational Model

Consider a single-step reaction model, $\nu_F F + \nu_O O \rightarrow \nu_P P$, in which ν_F moles of fuel (F) combine with ν_O moles of oxidizer (O) to form ν_P moles of product (P). The mass fractions of the fuel and oxidizer can be scaled by their maximum values in the unburned mixture, \tilde{Y}_{∞} and \tilde{X}_{∞} , so that the scaled values, Y and X, respectively, range between 0 and 1. These mass fractions are governed by

$$\frac{\partial \left(\rho Y\right)}{\partial t} + \nabla \cdot \left(\rho Y \mathbf{v}\right) - \rho \dot{\Omega} = 0 , \qquad (1)$$

$$\frac{\partial \left(\rho X\right)}{\partial t} + \nabla \cdot \left(\rho X \mathbf{v}\right) - \rho S \dot{\Omega} = 0, \tag{2}$$

where \mathbf{v} is the velocity and ρ is the density of the gas mixture. In the above equations,

$$\frac{\mathrm{d}Y}{\mathrm{d}t} = \dot{\Omega} = -YX\rho A^* e^{-E_a/RT} \tag{3}$$

is the reaction rate (in units of s⁻¹), E_a is the activation energy, R is the ideal gas constant, and A^* is the scaled pre-exponential factor in units of cm³/g s. The factor $S = s\tilde{Y}_{\infty}/\tilde{X}_{\infty}$ is a global equivalence ratio based on a mixture containing the maximum amounts of fuel and oxidizer in the unburned mixture, and $s = \nu_O W_O/\nu_F W_F$ is the stoichiometric coefficient of the reaction. Note that for a uniform mixture, S is exactly equal to the equivalence ratio of the unburned mixture.

Equations (1–2) are coupled with the reactive Euler equations, using the equations of state for an ideal, calorically-perfect gas. Energy is released by combustion according to $dE/dt = q^* \rho \dot{\Omega}$, where q^* is the scaled chemical energy release per unit mass of fuel, and E is the total energy density.

The system is partially premixed, and the local ϕ in the unburned mixture varies both spatially and temporally. For a two-component mixture, we define a local mixture fraction Z, that is, a conserved quantity across the reaction zone that can be related to the fuel and oxidizer mass fractions in the unburned mixture just upstream of the reaction zone, Y_{unb} and X_{unb} , respectively [11]. The local ϕ is proportional to the ratio of these unburned mass fractions and is computed using $\phi = SY_{unb}/X_{unb}$ [8].

The reaction parameters q^* and A^* are calibrated so that the computed Chapman-Jouget detonation velocities, D_{CJ} , and the variation of detonation cell sizes, λ , with equivalence ratio are similar to those of methane-air mixtures over a wide range of ϕ . As will be shown, the actual sizes of the detonation cells computed for this model mixture are an order of magnitude smaller than those measured in methane-air mixtures. To do so, we first assume the ratio of specific heats, $\gamma = 1.33$, is constant over the entire domain. We then find the values of q^* for which the computed D_{CJ} equals experimental measurements at five different equivalence ratios, $\phi = 0.6, 0.72, 1.0, 1.2$, and 1.43. Next, we construct least squares curve fits based on these five calibration points. The piecewise continuous function,

$$q^*M/RT_0 = 40.36 \begin{cases} -0.5766\phi^2 + 0.3991\phi + 1.1775 & \phi \le 1, \\ -0.4244\phi^2 + 1.03\phi + 0.3943 & \phi > 1, \end{cases}$$
(4)

gives approximate values of q^* as a function of ϕ , where $T_0 = 298$ K is the unburned gas temperature and M = 27 g/mol is the molecular weight of the mixture, which we take to be constant. In contrast to our past work [8], we also use a constant E_a chosen so that $E_a/RT_{ZND} = 5$ when $\phi = 1$, which is small enough to ensure the formation of regular detonation cells. Here T_{ZND} is the post-shock temperature computed using the one-dimensional ZND model. For $\gamma = 1.33$ and q^* obtained from eqn. (4) for $\phi = 1$, $T_{ZND} = 1592.7$ K and $E_a/RT_0 = 26.72$. A^* was then calibrated for the aforementioned five values of ϕ using the approximated values of q^* from eqn. (4), $\gamma = 1.33$, and $E_a/RT_0 = 26.72$. For each value of ϕ , A^* was systematically varied until the half-reaction length computed using the onedimensional ZND model, x_d , matched values inferred from experimental measurements of methane-air detonation cell sizes [1, 2] based on the correlation $\lambda/x_d = 50$ [12]. (The actual ratio of computed λ to x_d for this particular reaction model is smaller than this approximation.) A least squares method was then used to fit a smooth function to the values of A^* computed for the five calibration mixtures,

$$A^* = 1.0787 \times 10^9 \exp\left[-8.6011\phi^2 + 17.594\phi - 9.0\right] \,\mathrm{cm}^3/g \,s,\tag{5}$$

which we assume to be valid over the range of ϕ used in this study.

We consider two-dimensional, adiabatic channels of height h and length L with boundary conditions $\mathbf{v} = 0$, $\partial T/\partial n = \partial Y/\partial n = \partial X/\partial n = 0$ where n is the direction normal to the wall surface and \mathbf{v} is the velocity vector. The channels are filled with methane-air mixtures that vary in mixture composition from an equivalence ratio of $\phi = 1.95$ at the top wall to $\phi = 0.05$ at the bottom wall according to the distributions shown in Fig. 1. The mixture is ignited directly by a strong shock placed initially 2 cm to the right of the closed end of the channel.

The reactive Euler equations and Equations (1–2) are solved concurrently using the explicit operatorsplitting technique described in [8]. Equations (4–5) are used to determine A^* and q^* everywhere in the domain at the beginning of each time step based on local values of ϕ . A structured adaptive grid is used to achieve spatial resolution of 1/32 cm, corresponding to about 7 computational cells per half reaction zone thickness of a ZND detonation, which is sufficient for low-activation-energy mixtures [9].

3 Results and Discussion

We consider the behavior of a detonation propagating through three channels of varying sizes, h = 128, 256, and 512 cm, with concentration gradients shown in Fig. 1. In each case, the propagation is unstable, and multiple triple-shock configurations (triple points) form on the detonation front. We track the motions of these triple points by recording the time history of maximum pressure in the channels. This creates a series of numerical smoke foils that show the types of cellular structures typically found on experimental smoke foils. Numerical smoke foils and contours of reaction rate and temperature taken at a representative instant in time are shown in Fig. 2 for the three non-uniform mixtures described above and a uniform, stoichiometric ($\phi = 1$) mixture in the smallest, h = 128 cm, channel.

The detonation in the uniform stoichiometric mixture is, on average, planar and propagates at the CJ velocity. Closer inspection of the detonation shows the complex behavior of the reaction front. A number of triple points are visible that separate locally overdriven detonations from regions where the shock and reaction front are decoupled. The trajectories of the triple points are shown in the numerical



Figure 1: Distribution of the local equivalence ratio in the unburned mixtures across channels with h = 128 cm (red line), 256 cm (green line), and 512 cm (blue line).

smoke foil. The cellular pattern is fairly regular, with each individual cell differing in size by no more than a factor of two, so that approximately 15 detonation cells span the width of the channel (Fig. 2a).

The detonation propagating through the nonuniform mixture in the 128 cm channel is significantly more complex. Shortly after detonation initiation, the triple point trajectories, shown in Fig. 2b, are regular. As the detonation slows down, the distance between adjacent triple points grows as do the sizes of the cellular structures that are formed. Eventually, only two triple points remain in the channel, and the detonation nearly dies. The collision of these two remaining triple points ignites a new detonation. The sizes of the cellular structures formed by this detonation are quite large, and there is no clear gradation in size at any particular location in the channel. A typical reaction zone structure is shown in Fig. 2f. In some ways, it resembles the structure described by Calhoon and Sinha [5]. The detonation front is curved, and in the lean region (near the bottom of the channel) the reaction zone and shock are decoupled. In the rich mixture (near the top of the channel), the detonation survives almost all the way up to the wall. The reaction zone behind the shock near the top and bottom of the channel is quite large, but the energy release rate is small compared to that in the detonation front. Near the center of the channel, excess fuel (from the rich portion of the channel) and excess oxidizer (from the lean portion of the channel) combine and react in a turbulent diffusion flame.

The curvature of the detonation that forms in the 256 cm channel (Fig. 2g) is greater than that of the detonation in the 128 cm channel (Fig. 2f), and several additional triple points exist along the leading edge of the reaction zone. Again, the shock and reaction zone decouple near the bottom of the channel. The inert Mach reflection that forms near the bottom wall leaves behind a wide path in the numerical smoke foil (Fig. 2c). Triple points that reflect from the wall in this region are relatively weak and are barely visible on the numerical smoke foil. As they propagate into more reactive regions in the mixture, the triple points strengthen as more energy is released in the reaction zone. The situation is different for the Mach stem that forms near the top wall. The mixture in this region is more reactive than that near the bottom wall, which allows ignition to occur when the Mach stem becomes sufficiently wide. The reactive triple point that is formed propagates along the detonation toward the center of the channel. Eventually, a new Mach stem forms, and the process repeats itself. The average detonation velocity is 10% lower than the CJ velocity in the stoichiometric mixture, and the cellular structures that form near the center of the channel are much larger than those computed for the uniform stoichiometric mixture.

The detonation structure formed in the 512 cm channel and shown in Fig. 2h more closely resembles Calhoon and Sinha's stable detonation wave structure than that in the 256 cm channel. The primary



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Figure 2: (a–d) Numerical smoke foils and (e–h) contours of reaction rate (left plot) and temperature (right plot) computed from a detonation propagating through (a, e) a uniform stoichiometric mixture in a 128 cm channel, (b, f) a nonuniform mixture in a 128 cm channel, (c, g) a nonuniform mixture in a 256 cm channel, and (d, h) a nonuniform mixture in a 512 cm channel.

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physical difference for this case is that the distribution of ϕ and, hence, local properties of the detonation vary more slowly along the reaction front. As a result, the mixture between adjacent triple points is closer to being uniform, allowing the size of cellular structures to vary along the detonation front according to the local value of ϕ . Cells that form in the center of the channel (Fig. 2d) are smaller than those formed in the 256 cm channel and similar in size to those computed for the uniform stoichiometric mixture (Fig. 2a). Away from the center of the channel, where the mixture is fuel rich or fuel lean, the detonation cells are larger and more distorted, consistent with experimental observations [6]. The distortion of cells at the edges of the detonable mixture can be explained by considering the propagation of a triple point along a curved reaction front. Triple points moving away from the center of the channel are propagating backwards relative to the propagation direction of the detonation, and their trajectories appear to bend closer to the vertical in the numerical smoke foils. Those moving toward the center of the channel propagate in the same direction as the detonation, and their trajectories appear to be closer to the horizontal in the smoke foils.

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