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Numerical investigation of the instability for one-dimensional Chapman-Jouguet detonations with chain-branching kinetics

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Numerical investigation of the instability for one-dimensional Chapman–Jouguet detonations with chain-branching kinetics

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The dynamics of one-dimensional Chapman–Jouguet detonations driven by chain-branching kinetics is studied using numerical simulations. The chemical kinetic model is based on a two-step reaction mechanism, consisting of a thermally neutral induction step followed by a main reaction layer, both governed by Arrhenius kinetics. Results are in agreement with previous studies that detonations become unstable when the induction zone dominates over the main reaction layer. To study the nonlinear dynamics, a bifurcation diagram is constructed from the computational results. Similar to previous results obtained with a single-step Arrhenius rate law, it is shown that the route to higher instability follows the Feigenbaum route of a period-doubling cascade. The corresponding Feigenbaum number, defined as the ratio of intervals between successive bifurcations, appears to be close to the universal value of 4.669. The present parametric analysis determines quantitatively the relevant non-dimensional parameter χ, defined as the activation energy for the induction process ε_I multiplied by the ratio of the induction length Δ_I to the reaction length Δ_R. The reaction length Δ_R is estimated by the inverse of the maximum thermicity $(1/\dot{\sigma}_{\max})$ multiplied by the Chapman–Jouguet particle velocity u_{CJ}. An attempt is made to provide a physical explanation of this stability parameter from the coherence concept. A series of computations is carried out to obtain the neutral stability curve for one-dimensional detonation waves over a wide range of chemical parameters for the model. These results are compared with those obtained from numerical simulations using detailed chemistry for some common gaseous combustible mixtures.

Keywords: Detonations; Numerical simulations; Instability; Limit; Chain-branching kinetics; Nonlinear dynamics

1. Introduction

Gaseous detonations are self-sustained combustion-driven waves traveling at supersonic velocities on the order of 2 km/s. The classical theory of Zel'dovich, von Neumann and Döring from the early 1940s, which has come to be known as the ZND model, postulates that detonation waves have a steady one-dimensional structure consisting of a leading shock sustained by the chemical reactions in the adiabatically compressed gases [1]. However, this classical steady ZND structure for detonation waves is seldom observed. It has been well demonstrated both experimentally and theoretically that detonations are instead a complex spatial–temporal unstable phenomenon. The inherent chemical–gas dynamics instabilities can manifest themselves by a variety of unstable patterns ranging from a multidimensional complex of transverse shock waves to regular oscillating fronts under appropriate circumstances.

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Over the past four decades, many investigations have been carried out to study theoretically and numerically the one-dimensional pulsating detonation as a first stage in improving understanding of the detonation instability problem. The majority of these studies on pulsating detonation dynamics, however, have adopted a one-step chemistry approximation where an Arrhenius form of the reaction rate accounts for global features of a more complex chemistry [2–7]. Application of such a simplified one-step model, although often convenient for analytical or numerical purposes due to its simplicity and computational efficiency, has several drawbacks when attempting to study the complex interaction between the gas dynamics and chemical kinetic processes involved in a detonation wave in real systems.

In practice, the chemical kinetics for typical hydrocarbon mixtures are known to be of the chain-branching type, which proceed by a sequence of different reaction stages. The reaction zone structure generally has a thermally neutral region of radical accumulation that exists at the start of the chemical reactions corresponding to chain-initiation and chain-branching reactions, and a main reaction layer of chain-recombination or chain-termination reactions where radicals recombine to form the products and release chemical energy. While the detonation instability has been widely studied both theoretically and numerically using one-step Arrhenius kinetics, such a simplistic reaction model cannot reproduce the features of real detonations governed by chain-branching kinetics described above. It is difficult or even impossible to choose the one-step chemical kinetic parameters in such a way that the computed reaction zone structure gives close quantitative agreement with that calculated using realistic chemistry for typical combustible mixtures.

Recently, there has been renewed interest in clarifying the pulsating detonation instability with multi-step kinetic models [8–12], which mimic the essential dynamics of real chain-branching systems having separate induction and main heat release layers. For the model three-step chain-branching reaction mechanism studied by Short and Quirk [8], the chain-branching cross-over temperature T_B is used as a bifurcation parameter to control the ratio of the chain-branching induction length to the length of the recombination zone. It was found that the detonations are stable when the structure is dominated by the chain-termination zone. As the ratio increases by increasing T_B, the detonation undergoes a Hopf bifurcation leading to a nonlinear periodic oscillation. Further increase causes the pulsation to change from a regular oscillation towards a chaotically irregular, aperiodic oscillation, and eventually to failure.

The nonlinear dynamics of one-dimensional pulsating detonations have also been studied both asymptotically and numerically using a simpler two-step reaction model [10, 12]. The model consists of a thermally neutral, temperature sensitive induction stage, at the end of which the fuel is converted instantaneously into chain-radicals, followed by a temperature-independent exothermic reaction stage. An important finding from these studies further supports the fact that a bifurcation boundary between stable and unstable detonations may be found when the ratio of the length of the heat release layer to that of the induction zone layer is of the order $\vartheta(\varepsilon_1)$, where ε_I is the activation energy in the induction zone.

The purpose of this work is two-fold. The first is an extension of the earlier work by Short and Sharpe [12] to carry out a series of numerical computations and examine the long-term nonlinear dynamics of one-dimension pulsating detonation with a generic two-step reaction model. Simulations are performed for a wide range of initial conditions and reaction parameters of the model. The aim of the parametric study is to identify numerically the neutral stability curve for one-dimensional detonations with chain-branched kinetics and introduce a well-defined quantitative parameter which can be used to characterize the one-dimensional detonation stability. The second part of this study is to carry out simulations using detailed chemistry and to compare directly with the results from the two-step kinetic model.

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2. Problem formulations and numerical methodology

2.1 *Basic governing equations*

The governing equations used to simulate the inviscid, one-dimensional propagation of a detonation wave are the one-dimensional reactive Euler equations. They have the following non-dimensional form:

$$\frac{\partial \rho}{\partial t} + \frac{\partial(\rho u)}{\partial x} = 0 \tag{1}$$

$$\frac{\partial(\rho u)}{\partial t} + \frac{\partial(\rho u^2 + p)}{\partial x} = 0 \tag{2}$$

$$\frac{\partial \rho e}{\partial t} + \frac{\partial[u(\rho e + p)]}{\partial x} = 0 \tag{3}$$

assuming a polytropic equation of state and an ideal thermal equation of state:

$$e = \frac{p}{\rho(\gamma - 1)} + \frac{u^2}{2} - q \tag{4}$$

$$T = \frac{p}{\rho} \tag{5}$$

The variables ρ, u, p, e and q are the density, velocity, pressure and specific total energy and the local chemical energy release, respectively. All the flow variables have been made dimensionless by reference to the uniform unburned state as follows (the symbol ˜ denotes dimensional quantities and subscript (o) indicates quantities ahead of the detonation front):

$$\rho = \frac{\tilde{\rho}}{\tilde{\rho}_o}, \quad p = \frac{\tilde{p}}{\tilde{p}_o}, \quad T = \frac{\tilde{T}}{\tilde{T}_o}, \quad u = \frac{\tilde{u}}{\sqrt{\tilde{R}\tilde{T}_o}} \tag{6}$$

The above equations are then coupled with the chemical kinetic model to describe the detonation structure. As in [12], a two-step reaction scheme is used in this study to mimic the essential dynamics of a chain-branching reaction. The first step represents a thermally neutral induction zone or ignition process, with a temperature-sensitive Arrhenius form of the reaction rate given by:

$$\frac{\partial(\rho\xi)}{\partial t} + \frac{\partial(\rho u \xi)}{\partial x} = H(1-\xi) \cdot \rho k_I \exp\left[E_I\left(\frac{1}{T_s} - \frac{1}{T}\right)\right] \tag{7}$$

where ξ is the reaction progress variable in the induction period and $H(1-\xi)$ is a step function, i.e.:

$$H(1-\xi)\begin{cases} = 1 & \text{if} \quad \xi < 1 \\ = 0 & \text{if} \quad \xi \geq 1 \end{cases} \tag{8}$$

Here, the reference length scale x_{ref} is chosen such that the one-dimensional ZND induction length is unity, i.e. $k_I = -u_{vn}$ where u_{vn} is the particle velocity behind the shock front in the shock-fixed frame for the CJ detonation. The reference time scale is therefore set to the reference length scale divided by the initial sound speed of the reactant (i.e. $t_{ref} = x_{ref}/c_o$). At the end of induction period (i.e. $x = 1$), the second step begins to describe the rapid energy release after the branched-chain thermal explosion and the slow heat release in the radical

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recombination stage. The reaction rate equation for this step is given by:

$$\frac{\partial(\rho\lambda)}{\partial t}+\frac{\partial(\rho u\lambda)}{\partial x}=(1-H(1-\xi))\cdot\rho k_R(1-\lambda)\exp(-E_R/T) \tag{9}$$

where λ and k_R denote the chain-recombination reaction progress variable and rate constant for the heat release process. Similar to previous studies, the reaction rate constant k_R is used as a parameter to control the ratio of the reaction time to the induction time. The present model differs slightly from that of Short and Sharpe [12] only in the rate of the second step, which is chosen in Arrhenius form for generality. The local chemical energy that has been released at any instant during the reaction is equal to $q=\lambda Q$, where Q is the total chemical energy available in the mixture. The Mach number of the Chapman–Jouguet detonation (CJ) corresponds to a chosen value of Q can be determined by:

$$M_{CJ}=\frac{D}{c_o}=\left[\left(1+\frac{\gamma^2-1}{\gamma}Q\right)+\sqrt{\left[\left(1+\frac{\gamma^2-1}{\gamma}Q\right)^2-1\right]}\right]^{1/2} \tag{10}$$

Consistent with previous non-dimensionalization, the energy release Q and activation energies E have been scaled with RT_o. For ease of comparison in the following discussion, we also introduce alternative scaling for the activation energies:

$$\varepsilon_I=\frac{E_I}{\delta},\quad \varepsilon_R=\frac{E_R}{\delta}\quad\text{with}\quad \delta=\frac{\left[2\gamma M_{CJ}^2-(\gamma-1)\right]\left[2+(\gamma-1)M_{CJ}^2\right]}{(\gamma+1)^2M_{CJ}^2} \tag{11}$$

where δ is the temperature jump across the leading shock. Therefore, ε is simply the reduced activation energy normalized by the temperature behind the leading shock of the CJ detonation. For typical hydrocarbon mixtures, the reduced activation energy of the induction stage ε_I is large because in the induction zone, energy is required to break the strong chemical bonds of the fuel and convert it into radicals. Typical values for ε_I usually ranges from 4 (for H_2–O_2 mixture) to 12 (for heavy hydrocarbon mixtures). In contrast, the second step involves only reactions between energetic free radicals. For typical chain-branching reactions, therefore, the induction stage generally has a larger activation energy compared to the second step. Hence, we set for the present study:

$$E_I\gg E_R\quad or\quad \varepsilon_I\gg\varepsilon_R \tag{12}$$

2.2 *Stability parameter*

One of the motivations for this study is to identify the quantitative parameter that controls one-dimensional detonation stability driven by chain-branching kinetics. From the earlier work [10, 12], it was found that the bifurcation boundary between stable and unstable detonations occurs when the ratio of the length of the heat release layer to that of the induction zone layer is $\vartheta(\varepsilon_I)$. Based on these previous analyses, we have introduced mathematically the following stability parameter to characterize the detonation stability:

$$\chi\equiv\varepsilon_I\frac{\Delta_I}{\Delta_R}=\varepsilon_I\Delta_I\frac{\dot{\sigma}_{\max}}{u_{CJ}} \tag{13}$$

where Δ_I and Δ_R denote the characteristic induction length and reaction length, respectively. ε_I is activation energy governing the sensitivity of the induction period as defined previously. It should be pointed out that a similar stability parameter can be derived from Short's stability analysis of the amplitude of oscillations in self-supported ZND detonations [10]. Generally, the induction length is simply defined as the length of the thermally neutral period. However, there

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is no standard definition for reaction length. In this study, it is suggested that the characteristic reaction length can be defined using the thermicity parameter. For the present chemical model of ideal gases with constant specific heat ratio, the thermicity is expressed as [1]:

$$\dot{\sigma} = (\gamma - 1)\frac{Q}{c^2}\frac{d\lambda}{dt} \tag{14}$$

The thermicity $\dot{\sigma}$ basically denotes the normalized chemical energy release rate and has a dimension of 1/time. Hence, the inverse of maximum thermicity $\dot{\sigma}_{\max}$ can be taken as a characteristic time scale for the heat release, which may provide an appropriate choice to define a characteristic reaction time. Using this characteristic reaction time, we estimate the reaction length as:

$$\Delta_R = \frac{u_{CJ}}{\dot{\sigma}_{\max}} \tag{15}$$

where u_{CJ} is simply the particle velocity at the CJ plane in shock-fixed coordinates. Note that the above definition of different chemical kinetic parameters should be general enough such that it can be determined easily from any detailed chemical kinetics model.

2.3 *Numerical methodology*

The unsteady one-dimensional reactive Euler equations with the present chemical kinetic model are solved numerically using a second-order centered scheme, namely the SLIC (Slope Limiter Centered) scheme; its detailed description can be found in [13] and [14]. In the present work, adaptive mesh refinement (AMR) is also adopted as a technique for manipulating the local resolution of numerical simulations based on a hierarchical series of Cartesian grids [15]. For all the simulations, an effective numerical resolution of 128 points per induction zone length of the steady ZND detonation is used to ensure the detailed features of the pulsating front are properly resolved. The computations are initialized by the steady solution of the ZND detonation.

3. Numerical results

3.1 *Development of instability*

The development of instability is first investigated near the stability boundary. For each set of chemical parameters (i.e. the heat release Q, specific heat ratio γ and activation energies ε_I and ε_R), we perform simulations to identify the stability limit below which the detonation propagation is stable by varying the value of the reaction rate constant k_R. Leading shock pressure history is recorded and used to determine stable or unstable detonations based on its decay or growth at the long time-evolution. For example, the pressure histories of the detonation front for $Q = 50$, $\gamma = 1.2$, $\varepsilon_I = 8$ and $\varepsilon_R = 1$ with different reaction rate constants k_R are displayed in figure 1. For $k_R = 0.887$ and $k_R = 0.967$, the initial perturbation due to the numerical startup errors damps with time and the propagating detonation eventually reaches a stable configuration after the long-term evolution. However, for a case with $k_R = 1.021$, results show a slow amplification of a weakly pulsating instability, exhibiting a single mode low-frequency oscillation. Results indicate that the stability limit is found to be approximately around $k_R \approx 0.975$. As k_R is increased further, the instability grows quickly from the initial perturbation and the propagating detonation becomes unstable. These results agree well with

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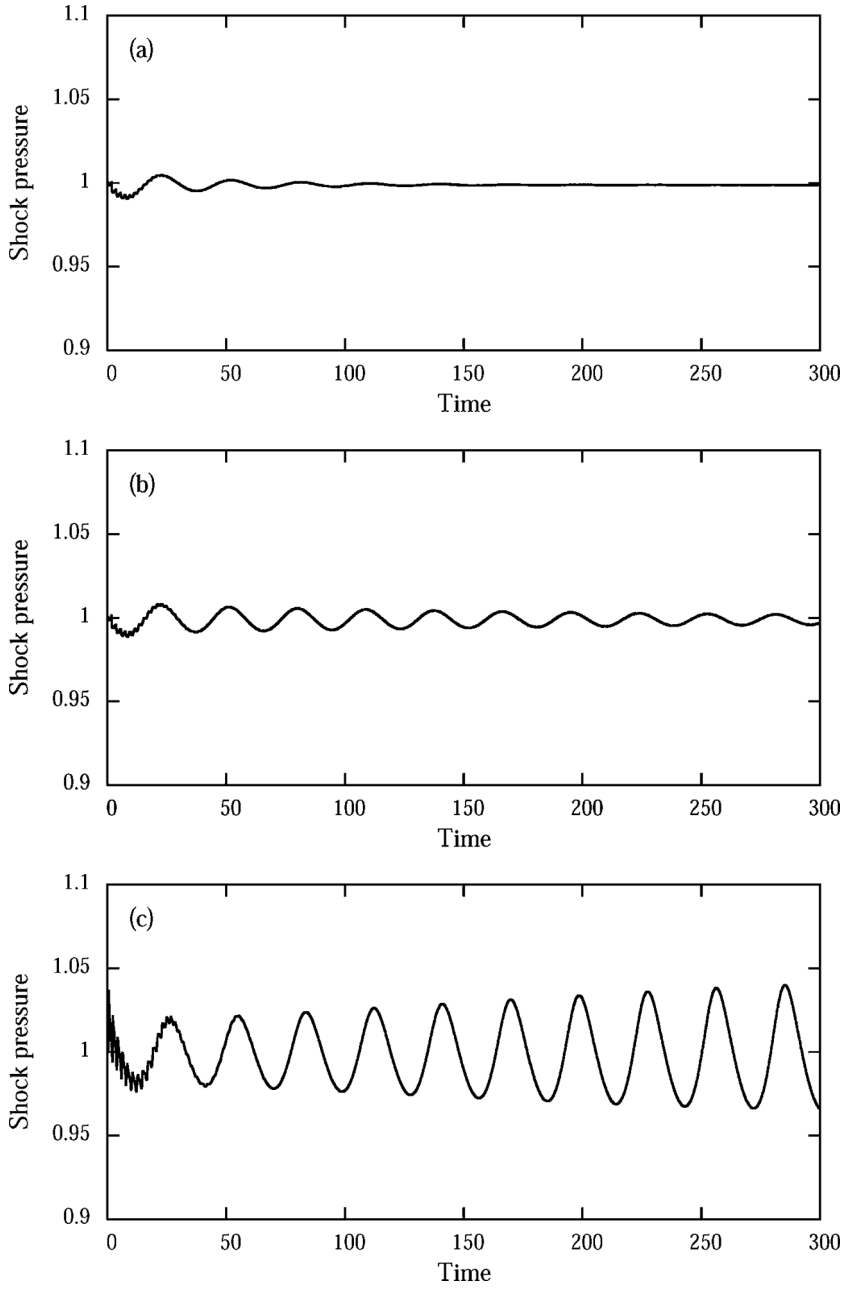

Figure 1. Leading shock pressure history (i.e. a plot of $\tilde{P}_s/P_o$ versus $\tilde{t}/t_{ref}$) for reaction rate constants close to the stability limit: (a) $k_R = 0.887$; (b) $k_R = 0.967$; and (c) $k_R = 1.021$.

the earlier work by Short and Sharpe [12], showing that the increase in k_R resulting in a reduction in reaction length relative to the induction length can cause instability in the detonation propagation. To illustrate the grid convergence, table 1 shows the numerically determined value of the stability boundary for various numerical resolutions. The resolution study indicates that a resolution of 128 grids per induction length of the steady ZND detonation provides sufficient refinement to obtain a converged value of the stability boundary.

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Table 1. Numerically determined stability boundary for different resolutions.

Resolution	k_R
8	1.001
16	0.990
32	0.978
64	0.974
128	0.974
256	0.974

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If the value of k_R moves further away from the neutral stability boundary, the propagating detonation undergoes a period-doubling bifurcation to a higher mode oscillation (figure 2). Similar to those results obtained using a single-step Arrhenius rate law, development of the higher instability mode is done via a series of bifurcation processes. This period-doubling cascade eventually leads to chaos [16]. Similar to our previous study using a single-step Arrhenius rate law, a bifurcation diagram is constructed in this study with chain-branching kinetics to illustrate the sequences of transitions, as given in figure 3. The bifurcation diagram is generated by measuring the peak pressure amplitude of the oscillations for each value of reaction constant k_R. In this way, a bifurcation diagram would display the magnitude of one mode of oscillation as a single point. A detailed explanation of this diagram can be found in Ng *et al*. [16].

An important characteristic feature of the bifurcation diagram is the ratio between the spacing of successive bifurcations, i.e. the Feigenbaum number:

$$\delta = \frac{\mu_i - \mu_{i-1}}{\mu_{i+1} - \mu_i} \tag{16}$$

where μ_i is the location of the bifurcation point. To compute the Feigenbaum number, the values of reaction rate constant k_R at which the bifurcation occurs are determined directly from the bifurcation diagram. These values of k_R as well as the computed Feigenbaum numbers are displayed in table 2. It is interesting to note that the Feigenbaum number for the present bifurcation diagram also appear to be close to the universal value of 4.669, derived by Feigenbaum for a simple nonlinear model with fewer degrees of freedom. In summary, the bifurcation diagram indicates that the transition process of the pulsating detonation front driven by chain-branching kinetics also follows closely the generic pattern of Feigenbaum's period-doubling scenario [17]. The present result also agrees with that of our prior study in which activation energy of single-step Arrhenius rate law was varied and a similar value of the Feigenbaum number was measured. Hence, the transition pattern for unstable pulsating detonations appears to be independent of the chemical kinetics used in the model. These universality properties may perhaps suggest that some simple model belonging to the Feigenbaum's system and other nonlinear dynamics concepts can be employed as a way to investigate the dynamics of one-dimensional pulsating detonations [16].

3.2 *Neutral stability boundaries*

More computations are carried out in order to obtain a general neutral stability curve for various mixture parameters. In the present study, calculations are performed for three specific heat ratios, $\gamma = 1.2$, 1.3 and 1.4. For each γ, the value of chemical heat release Q is varied to give a range of Mach numbers M_{CJ} between 3.5 and 7.8. We consider a combination of parameters with three possible activation energies $\varepsilon_I = 4, 6, 8$ and two possible activation energies $\varepsilon_R =$

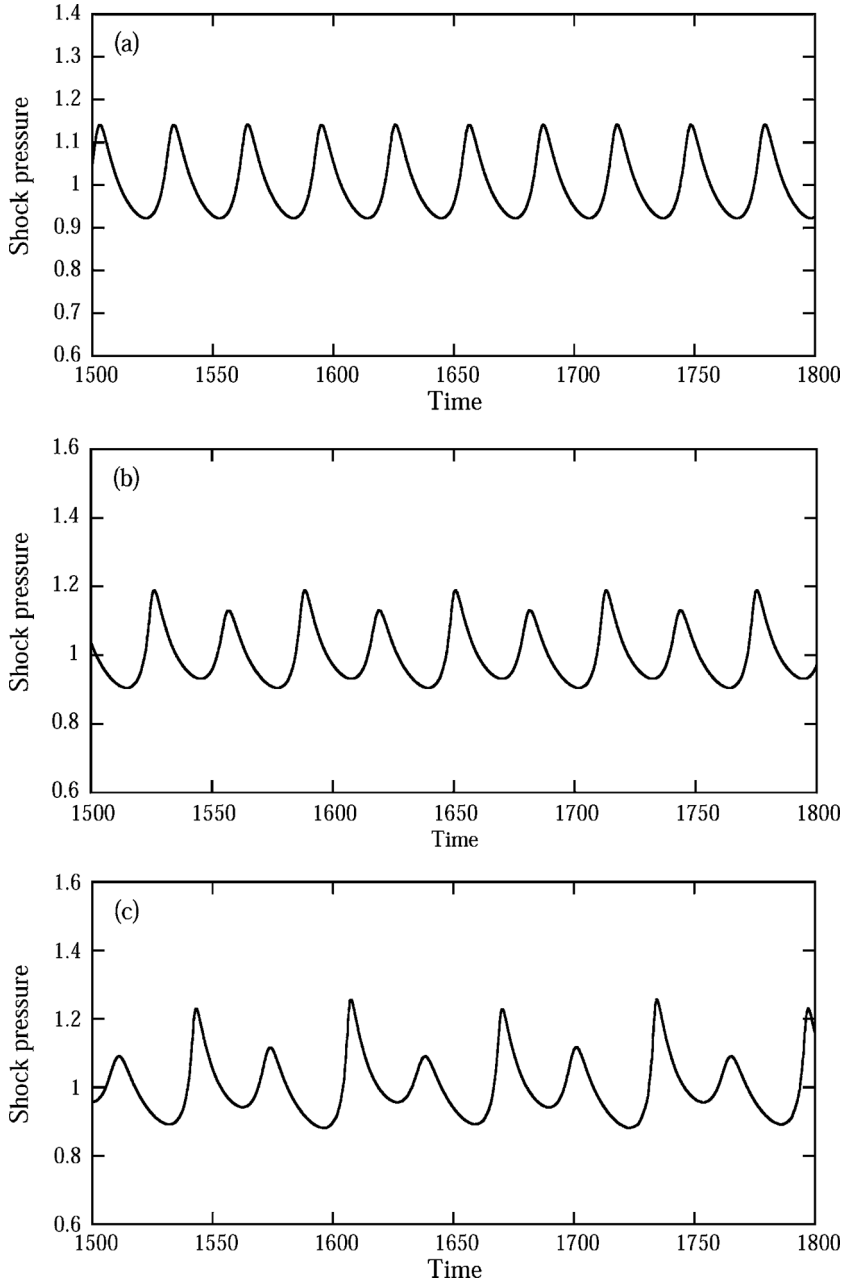

Figure 2. Leading shock pressure history exhibiting a period-doubling bifurcation for unstable detonations: (a) $k_R = 1.126$; (b) $k_R = 1.157$; and (c) $k_R = 1.189$.

0.5, 1. The variation of these parameters should be able to mimic the characteristics of any realistic chemical systems and can be easily obtained for various combustible mixtures. For each set of mixture parameters (γ, Q, ε_I, ε_R), computations were performed in which the value of the reaction rate constant k_R was systematically varied to determine its critical value at which the detonation becomes unstable, as described in section 3.1. The stability parameter χ associated with this critical reaction rate constant k_R is then computed, which corresponds to the value at the stability boundary.

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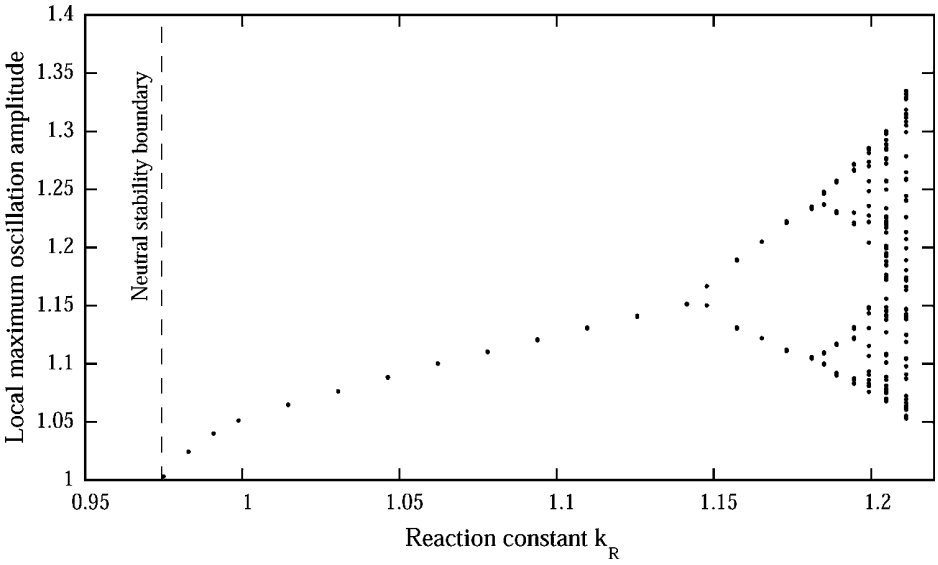

Figure 3. Bifurcation diagram.

Figure 4 summarizes all these computational results by showing a plot in the χ–M_{CJ} plane for different mixture parameters considered as shown in the figure legend. Only the critical value of χ, at which the detonation becomes unstable, is shown for a given set of parameters (γ, Q, ε_I, ε_R). It is interesting to note that all the data points corresponding to different mixture parameters can be essentially collapsed to a single curve. Although there are still some differences among these data points, this can be readily explained by the fact that the value of the exothermic reaction zone length Δ_R, which is necessary to evaluate the stability parameter χ, is obtained using a rather simple mathematical approximation as given by equation (15). Secondly, the difference in the data points can also be attributed to the incremental size of k_R we use and the way we determine the stability boundary. Nevertheless, those discrepancies are not significant and this may suggest a universal curve to define the stability boundary for one-dimensional detonations.

3.3 *Discussion on the stability parameter*

Previous studies by other researchers [10, 12] as well as the present analysis all support that χ is the relevant parameter governing stability. It may be worthwhile to point out that the derivation and physical meaning of this parameter was equivalently discussed by Soloukhin in a comment to Meyer and Oppenheim's work on the shock-induced ignition problem. In discussion of the experiments, Soloukhin and Oppenheim both noted that ignition behind a

Table 2. Values of bifurcation limits and Feigenbaum numbers.

i	Reaction constant k_R	Oscillation mode	Feigenbaum number
1	0.975	1	–
2	1.144	2	–
3	1.183	4	4.33
4	1.191	8	4.88

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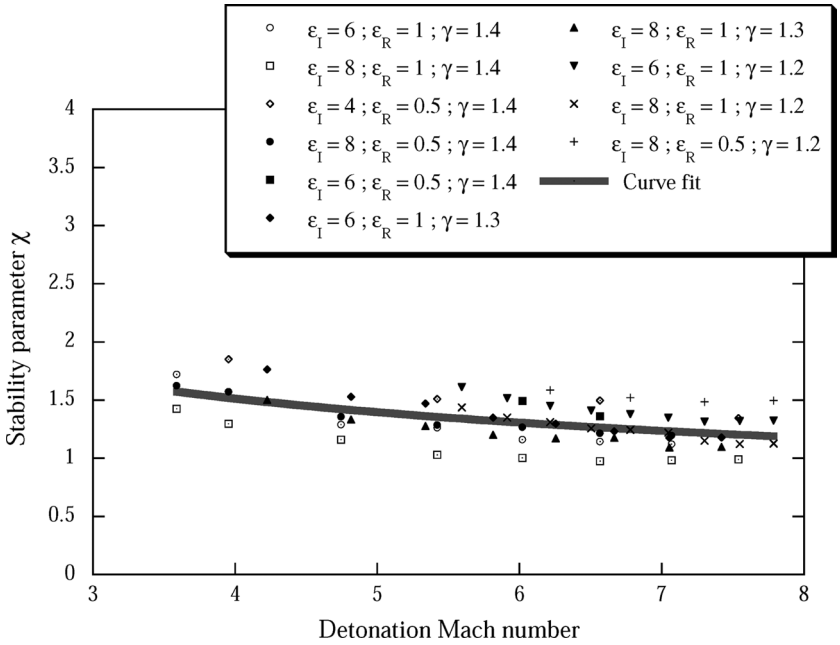

Figure 4. Neutral stability curve in χ–M_{CJ} plane.

shock could either occur uniformly (the strong regime) or originate from several exothermic spots (the mild regime) [18–20]. For the case of detonations, the mild regime can analogously correspond to unstable detonations where instability is caused by pockets of partly burned gas [5, 6], while the strong regime is similar to stable detonations, where chemical reaction is insensitive to perturbations and all the fuel burns uniformly as in the ZND model. Hence the stability in the reaction zone of gaseous detonations may be comparably linked to the regime of ignition behind shock waves, as already noted by Takai *et al.* [21].

We can follow Oppenheim's work to formulate the requirement of stability in the reaction structure of a detonation. It demands that neighboring particles, shocked initially at slightly different shock strengths, would release their chemical energy with similar delays, so that the power pulses can overlap, or be 'coherent' in time and space as to give rise to a single global gasdynamic effect free of instabilities [22–24]. First, to meet this requirement, the sensitivity of the chemical induction length (or equivalently time) to changes in shock temperature needs to be small. Otherwise, particles of gas being shocked at different temperatures due to perturbations in the flow will take significantly different times to burn and thus, can possibly lead to the formation of pockets of partly burnt fuel whose burnout at later time will eventually cause instability in the reaction zone [6]. For gaseous detonation waves, the induction length can be assumed to have the common Arrhenius from:

$$\Delta_I \propto \exp\left(\frac{E_I}{T_s}\right) \tag{17}$$

where T_s is the initial shocked gas temperature and E_I the global activation energy describing the sensitivity of the thermally neutral chemical induction process. The stability thus requires that

$$\left|\left(\frac{T_s}{\Delta_I}\right)\left(\frac{\partial \Delta_I}{\partial T_s}\right)\right| = \frac{E_I}{T_s} \equiv \varepsilon_I \tag{18}$$

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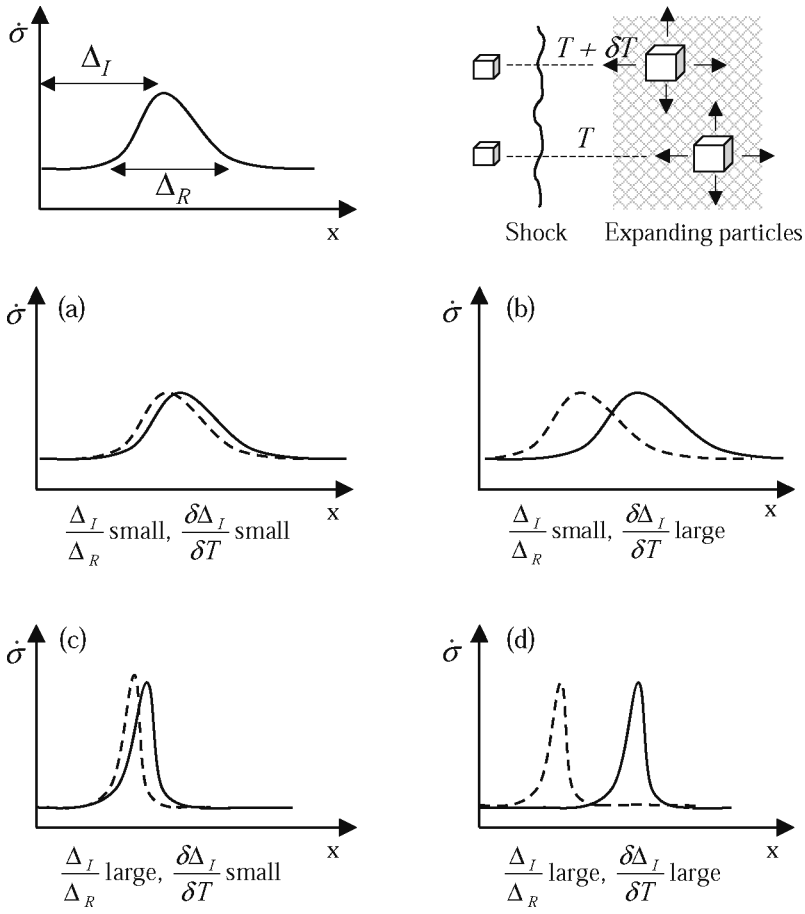

Figure 5. Illustration of the coherent concept between neighboring power pulses, given by the exothermicity profiles for two neighboring gas elements shocked at temperatures differing by δT. (a) small temperature senstivity, long exothermic reaction length; (b) large temperature sensitivity, long exothermic reaction length; (c) small temperature senstivity, short exothermic reaction length; and (d) large temperature senstivity, short exothermic reaction length. Only case (d) results in incoherence of power pulses and the development of instability.

be small, i.e. small activation energy ε_I. However, in addition to the effect of sensitivity in the induction process, the characteristic length scale for energy deposition Δ_R should also play an important role. Note that a relatively long period for energy release (broad power pulses) will still lead to quasi-simultaneous energy deposition and coherence in time and space even if the changes of induction length (or time) are important. Thus, similar to Soloukhin's comment on Meyer and Oppenheim's work [19], stability can be more properly described by the sensitivity to temperature fluctuations of the characteristic induction length relative to the characteristic exothermic reaction length. This concept can be illustrated schematically as shown in figure 5. Mathematically, we can then define based on this physical concept a stability parameter of the form as shown equivalently in the previous section, i.e.

$$\chi \equiv \left| \left(\frac{T_s}{\Delta_R} \right) \left(\frac{\partial \Delta_I}{\partial T_s} \right) \right| = \varepsilon_I \frac{\Delta_I}{\Delta_R} = \varepsilon_I \Delta_I \frac{\dot{\sigma}_{\max}}{u_{CJ}} \tag{19}$$

where the length scale for the exothermic power pulse Δ_R is simply the inverse of the maximum value of thermicity $\dot{\sigma}_{\max}$ multiplied by the CJ particle velocity u_{CJ}, as defined previously. This

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non-dimensional parameter includes the essential terms that influence the characteristics of power pulses or energy release. For small values of χ, we expect that power pulses originating from neighboring particles will overlap, thus leading to a coherent phenomenon in time and space (see figure 5(a)–(c)). In that case, small disturbance in the flow will not cause significant fluctuation of the energy release in the reaction zone structure, giving a stable or weakly unstable system. On the other hand, if this parameter is large, as can occur with conditions given in figure 5(d), the power pulses will not be coherent and this can lead to various gas-dynamic fluctuations in the reaction zone structure. This stability parameter therefore describes the scenario in which the incoherence in the energy release of the gas leads to gasdynamic instabilities in the reaction zone.

4. Simulations using detailed chemistry

Results from the one-dimensional simulations of detonations driven by a generalized two-step reaction model illustrate the importance of chain-branching kinetics and the effect of the two characteristic length scales, namely, the characteristic induction and reaction length. A stability parameter is also examined and based on which a neutral stability boundary is obtained. In order to establish these findings, comparisons with experimental observations and numerical computations taking into account detailed chemistry for realistic mixtures are performed.

Different detonation kinetic parameters are first computed for some real combustible mixtures, which are required to obtain the stabilty parameter χ. The induction and reaction lengths were determined by solving the steady ZND structure of the detonations using the Chemkin package [25, 26]. As an example, the temperature and thermicity profiles of the ZND structure for $H_2 - O_2 - 25\%$Ar detonation are plotted in figure 6 and the two characteristic lengths are also shown on the figure. For the model with detailed chemistry, the induction length Δ_I was defined as the distance elapsed from the shock to the point where the fluid particle reaches its maximum rate of heat release, i.e. maximum thermicity $\dot{\sigma}$. As discussed in section 2.2, the characteristic length for the heat release was taken as the inverse of the maximum rate of heat release multiplied by the CJ particle velocity, i.e. $u_{CJ}/\dot{\sigma}_{\max}$. For the case with detailed

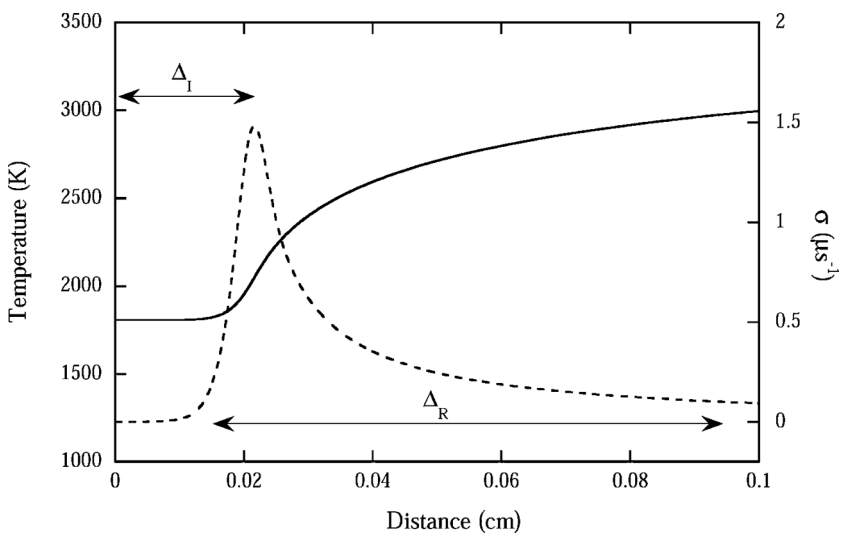

Figure 6. Temperature and thermicity profiles of the ZND structure of $2H_2 + O_2 + 25\%$Ar detonation at $T_o = 298$ K and $P_o = 0.2$ atm.

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chemistry, the thermicity is given by the following expression [1]:

$$\dot{\tilde{\sigma}} = \sum_{i=1}^{N_s} \left(\frac{\tilde{W}}{\tilde{W}_i} - \frac{\tilde{h}_i}{\tilde{c}_p \tilde{T}} \right) \frac{\mathrm{d}y_i}{\mathrm{d}\tilde{t}} \tag{20}$$

where $\tilde{W}$ is the mean molar mass of the mixture, $\tilde{c}_p$ is the mixture specific heat at constant pressure, and $\tilde{h}_i$ is the specific enthalpy of specie i.

Another chemical kinetics parameter is the global activation energy in the induction process ε_I. Its value can be obtained by performing constant-volume explosion calculations. Assuming that the induction time τ_i has an Arrhenius form, i.e.

$$\tilde{\tau}_{\mathrm{i}} = A\tilde{\rho}^n \exp\left(\frac{\tilde{E}_I}{\tilde{R}\tilde{T}} \right) \tag{21}$$

The activation temperature $\tilde{E}_I/\tilde{R}\tilde{T}_s$ may be determined by

$$\varepsilon_I = \frac{\tilde{E}_I}{\tilde{R}\tilde{T}_s} = \frac{1}{\tilde{T}_s} \frac{\ln \tilde{\tau}_2 - \ln \tilde{\tau}_1}{\frac{1}{\tilde{T}_2} - \frac{1}{\tilde{T}_1}} \tag{22}$$

where two constant-volume explosion simulations are run with initial conditions $(\tilde{T}_1, \tilde{\tau}_1)$ and $(\tilde{T}_2, \tilde{\tau}_2)$. Conditions for states one and two are obtained by considering the effect of a change in the shock velocity by $\pm 1\% \tilde{D}_{CJ}$ [27].

Values for different detonation kinetic parameters of some common combustible mixtures are shown in table 3. Using these computed parameters from the chemical kinetics calculations, the value of χ is then obtained for each mixture investigated and mapped onto the neutral stability curve obtained in section 3.2 for comparison (figure 7). Also tabulated is the qualitative assessment of the cell regularity as observed experimentally. We first consider the case of stoichiometric H_2—O_2 and C_2H_2—O_2 with high argon dilution. Experimentally it is shown that these mixtures are generally stable in the sense that they have a highly regular cellular structure comprised of weak transverse waves. One-dimensional unsteady simulations with detailed chemistry are also performed for these regular mixtures [14, 31]. For the case of $C_2H_2 + 2.5O_2 + 85\%Ar$ and $2H_2 + O_2 + 25\%Ar$, it is found that their propagation is described by an one-dimensional pulsating mode and the periodic pattern is regular (figures 8 and 9(a)). In figure 7, we see that the stability parameters χ for these two mixtures are slightly above the neutral stability boundary. While for the case of $2H_2 + O_2 + 40\%Ar$ which has a value of χ just below the neutral stability boundary, the oscillation of the detonation front is decaying with time and approaching to a 1D stable configuration (figure 9(b)). These mixtures of stoichiometric H_2—O_2 and C_2H_2—O_2 with high argon dilution are shown

Table 3. Values of different detonation parameters computed for mixture at $T_o = 298$ K and $P_o = 0.2$ atm using Varatharajan and Williams 7-steps reduced mechanism [28] for C_2H_2—O_2; Oran *et al.* mechanism [29] for H_2—O_2; and Konnov mechanism [30] for others.

Mixture	ε_I	Δ_I (cm)	Δ_R (cm)	χ	Cell regularity
$CH_4 + 2O_2$	11.84	0.102	2.30×10^{-2}	52.5	Highly irregular
$C_3H_8 + 5O_2$	10.50	1.66×10^{-2}	1.05×10^{-2}	16.6	Highly irregular
$C_2H_2 + 2.5O_2$	4.82	3.82×10^{-3}	3.57×10^{-3}	5.16	Irregular
$C_2H_2 + 2.5O_2 + 60\%Ar$	4.73	1.19×10^{-2}	1.57×10^{-2}	3.59	Regular
$C_2H_2 + 2.5O_2 + 85\%Ar$	4.83	5.42×10^{-2}	0.114	2.30	Regular
$2H_2 + O_2$	5.28	2.44×10^{-2}	5.72×10^{-2}	2.25	Regular
$2H_2 + O_2 + 25\%Ar$	4.91	2.18×10^{-2}	8.08×10^{-2}	1.33	Highly regular
$2H_2 + O_2 + 40\%Ar$	4.68	2.32×10^{-2}	0.119	0.91	Highly regular

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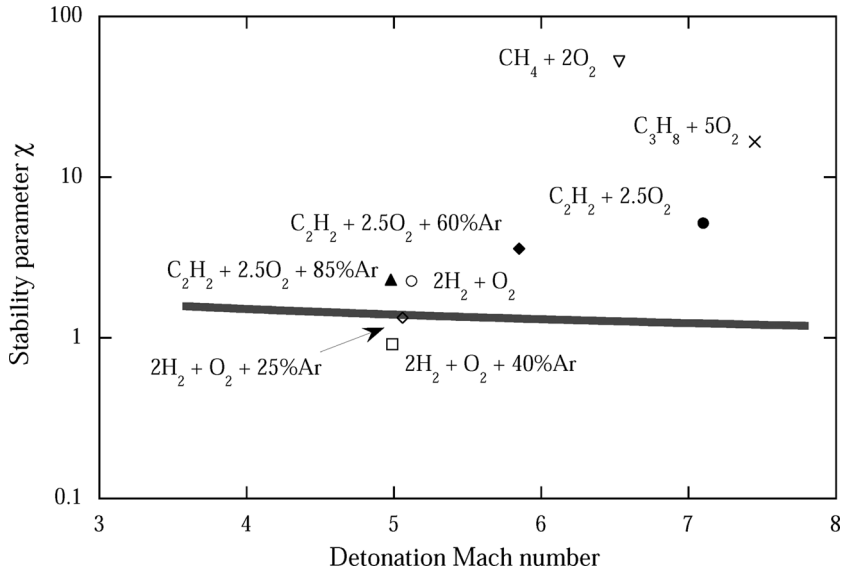

Figure 7. Neutral stability curve.

to be able to maintain their propagation by the one-dimensional shock-induced ignition mechanism.

On the other hand, mixtures without argon dilution or those with hydrocarbon fuels such as methane or propane have a large value of χ and are characterized by a highly irregular cellular structure as observed experimentally. For such mixtures with high value of χ, one-dimensional shock-ignition is no longer possible to maintain the self-propagation [8, 31]. Multi-dimensional effects such as transverse wave structure are essential for their propagation. These effects can thus cause significant irregularities in the detonation structure.

As discussed in section 3.3, for small values of χ the power pulses originating from neighboring particles will overlap, thus leading to a coherent phenomenon in time and space and a stable configuration. On the other hand, if the parameter χ is large, the power pulses will not be coherent, which can lead to the multi-dimensional gasdynamic instabilities in the reaction zone structure. As can be seen from these results, the coherent stability parameter defined in

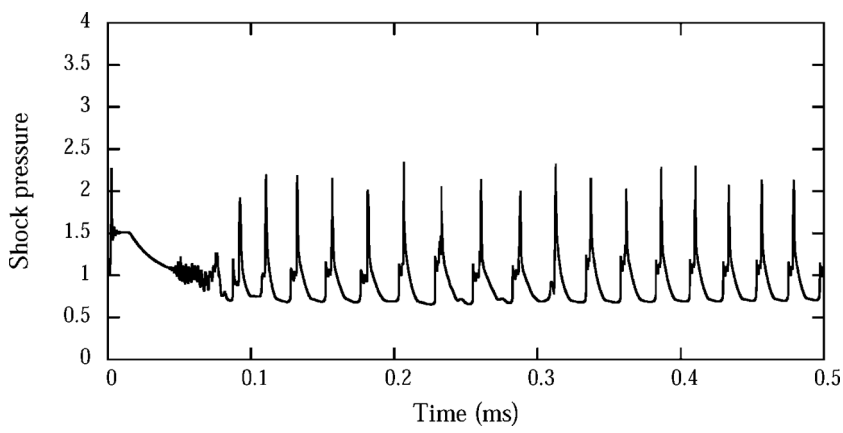

Figure 8. Leading shock pressure history for $C_2H_2 + 2.5O_2 + 85\%Ar$ detonation at $T_o = 298$ K and $P_o = 0.2$ atm.

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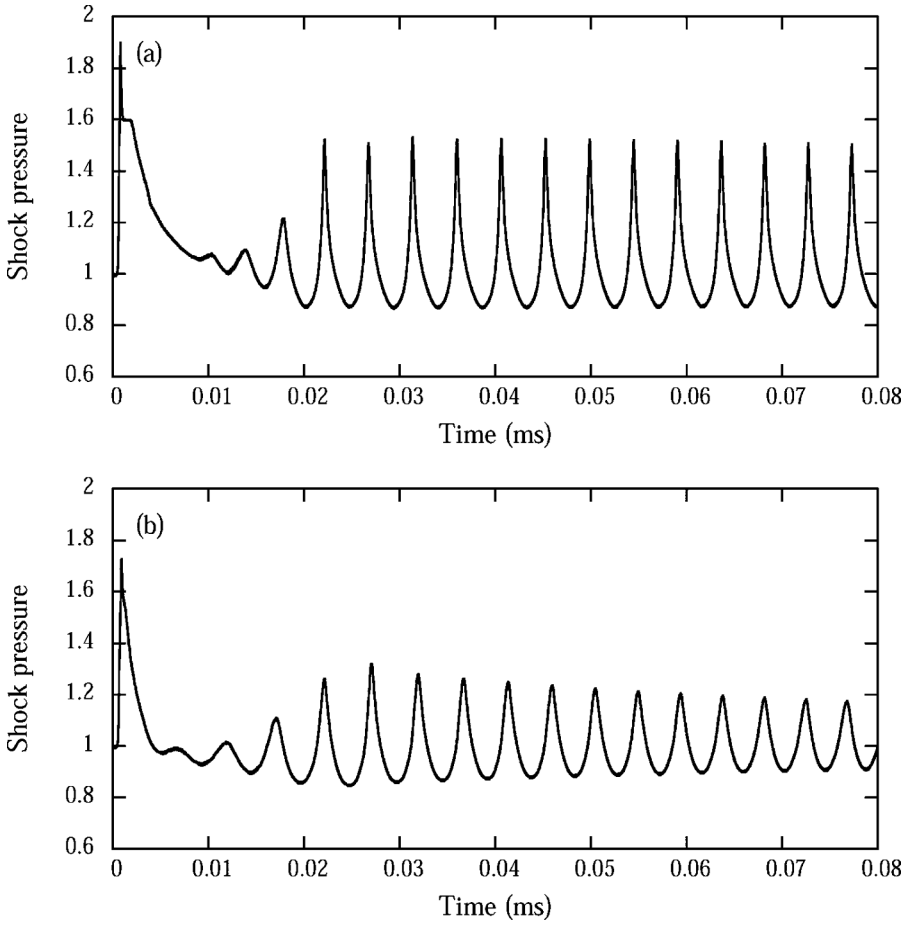

Figure 9. Leading shock pressure history for: (a) $2H_2 + O_2 + 25\%Ar$ detonation; and (b) $2H_2 + O_2 + 40\%Ar$ detonation at $T_o = 298$ K and $P_o = 0.2$ atm.

this study provides a good correlation in classifying the cell regularity, and the neutral stability curve found here also gives a good estimate of the degree of instability of the detonation waves.

5. Conclusion

In this paper we have performed a series of numerical computations of one-dimensional Chapman–Jouguet detonations driven by a two-step chain-branching kinetic mechanism. Use of this mechanism is intended to yield a better understanding of the stability and propagation of real detonations. In the present study, the results concur with previous studies of a similar kind that detonations become unstable when the induction zone length dominates over the main reaction layer, which occurs when increasing the reaction rate constant k_R. The nonlinear dynamics for higher modes of instability is analyzed via the construction of a bifurcation diagram. It is shown that the route to higher instability follows the Feigenbaum's bifurcation scenario.

Unlike prior detonation studies based on one-step Arrhenius kinetics where a single global activation energy E_a is the sole parameter governing stability, the present study suggests a more general non-dimensional parameter, χ. It is defined as the degree of temperature sensitivity in

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the induction zone ε_I multipled by the ratio of induction length Δ_I to the reaction length Δ_R, which is approximated by the inverse of the maximum thermicity $(1/\dot{\sigma}_{\max})$ multiplied by the CJ particle velocity u_{CJ}. The physical meaning of this parameter is pointed out to be equivalent to Meyer and Oppenheim's coherence concept. It states that stability can be achieved if the power pulses originating from neighboring particles will overlap or the reaction has a relatively long period for energy release, leading to a coherent phenomenon in time and space. On the other hand the incoherence in the power pulse or energy release, which is expected for large value of χ, leads to gasdynamics instabilities in the reaction zone.

Using the stability parameter χ, an universal neutral stability curve is obtained for different mixture parameters. Its universality is then examined by some results obtained from computations using realistic detailed chemistry. Although the present study is based on a one-dimensional model, such analysis should provide a qualitative assessment of the complex dynamics controlling the cell regularity as observed experimentally.

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