Formation conditions of double cellular detonation in $C_2H_4/O_2/O_3/N_2$ mixtures with two-stage reactions

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1 Introduction

Detonation is characterized by the coupling between shock waves and reactions [1]. The reaction process following the leading shock determines key detonation features, such as cellular stability [1]. A single cellular detonation structure forms in mixtures with a continuous exothermic process. However, in mixtures exhibiting two-stage reactions, a double cellular structure is observed [2, 3].

The double cellular structure was first identified in a gaseous nitromethane/oxygen mixture in the experiments conducted by Presles et al. [4]. Sturtzer et al. [5] subsequently analyzed the ZND structure of the nitromethane/oxygen mixture and proposed that the two successive distinct exothermic steps are responsible for the formation of the double cellular structure. Similar double cellular structures were later observed experimentally by Diakow et al. [6] and Ng et al. [7] in dimethyl ether-oxygen mixtures. Joubert et al. [8] separately added H_2, CH_4, and CH_4 to NO_2/N_2O_4 mixtures to induce two exothermic steps, successfully observing double cellular detonation in experiments. Virot et al. [9] further evaluated the influence of mixture pressure on detonation structures in H_2-NO_2/N_2O_4-Ar mixtures, finding that decreasing pressure can transition the double cellular detonation into a single cellular detonation.

Although double cellular detonation waves are widely observed in experiments, analyzing detailed flow structures through experimental observations is difficult. The multi-scale characteristics of double cellular detonation complicate the numerical assessment of wave structures, particularly when incorporating detailed chemistry. Consequently, most of the existing numerical studies adopted two-step global reaction model [9-12]. Double cellular detonation requires further assessment using more realistic chemistry models. Additionally, both Vasilev et al.'s experiments [2] and Mével et al.'s simulations [13] indicate that two-stage exothermic reactions are not a sufficient condition for the formation of double cellular detonation. This suggests that the mechanism underlying double cellular detonation formation is still not fully understood and requires further investigation. These knowledge gaps serve as the primary motivation for the current study.

In this work, two-dimensional simulations incorporating detailed chemistry are conducted to investigate transient detonation waves in $C_2H_4/O_2/O_3/N_2$ mixtures. Ozone is utilized to artificially create and regulate an additional reaction stage during the ethylene oxidation process [14]. Cellular detonation waves predicted under varying ozone content conditions are evaluated, and the formation conditions for double cellular detonation are analyzed.

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2 Numerical methods and setups

The schematic diagram of the computational domain is shown in Fig. 1. Initially, the domain is filled with a static $C_2H_4/O_2/O_3/N_2$ mixture at T_0 = 300 K and P_0 = 1 atm. The parameter $\kappa = 3X_{O3}/(2X_{O2}+3X_{O3})$ is defined and adjusted to regulate the reaction process behind the leading shock. $X_{C2H4}:X_{O2}:X_{O3}:X_{N2}$ = 1:3(1-κ):2κ:20. The left boundary is modeled as a wall, while the right boundary is set as a supersonic outlet. Cyclic conditions are applied to the top and bottom boundaries. An initiation zone with a temperature of 3000 K and pressure of 30 atm is set to induce the detonation wave.

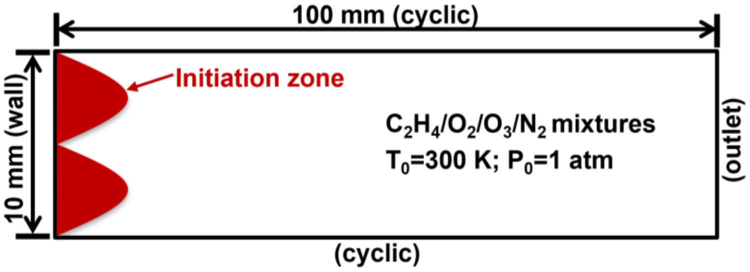

Fig. 1. Schematic of the simulation domain and settings.

In this work, the multi-component Navier-Stokes equations are used to describe the flow and reaction process. The solver detonationFoam [15] is employed to solve the governing equations and has been thoroughly validated for gaseous detonation simulations. The equations, numerical methods, and validation details can be found in Ref. [15]. A reduced mechanism for ethylene oxidation and an ozone sub-kinetic model [16, 17] are combined and used, which consists of 45 species and 289 elementary reactions. Uniform grids of 10 μm and 2-level adaptive refinement grids are used in the simulations, with the minimum grid size being $10/2^2 = 2.5$ μm.

3 Results and discussion

Cellular detonation waves for κ = 0.2, 0.4, 0.6, and 0.8 are simulated. The numerical soot foils illustrating the triple-point trajectories are shown in Fig. 2. When κ = 0.2, single detonation cells are observed in Fig. 2(a). Although weak triple-point trajectories are present within the large detonation cells, no stable small detonation cells form. As κ increases to 0.4 and 0.6, the corresponding results in Figs. 2(b-c) reveal obvious double cellular structures, where small detonation cells fill the larger ones. When κ = 0.8, Fig. 2(d) shows that the single cellular detonation wave propagates with only small detonation cells. These results indicate that the influence of κ on the detonation structure (i.e., single/double cellular detonation) is non-monotonic.

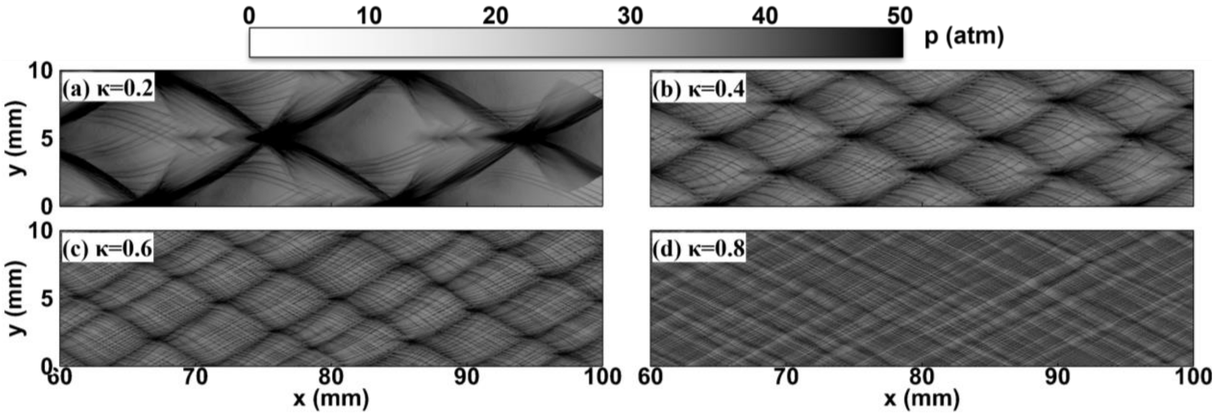

Fig. 2. Numerical soot foils recorded during the detonation wave propagation processes separately for κ = (a) 0.2, (b) 0.4, (c) 0.6 and (d) 0.8.

Taking $\kappa = 0.6$ as an instance, the steady ZND detonation structure is depicted in Fig. 3. It shows that the reaction proceeds with two stages behind the leading shock, which are characterized by the two peaks of the thermicity profile. x_m is defined as the position of the local minimum of thermicity between the two-stage reactions. $x_m = 1.3$ mm for $\kappa = 0.6$. In the first stage (corresponding to the zone $x < x_m$) in Fig. 3), C_2H_4 partially reacts, ozone is totally consumed, and significant heat release is observed. In the second stage (corresponding to the zone $x > x_m$), the remaining C_2H_4 reacts, causing the second-stage heat release in thermicity profile (see Fig. 3). Induction length, Δ_I, is defined as the distance from the leading shock to the thermicity peak location. The induction length of the first-stage reaction, Δ_{I1}, equal 3.6 μm, and the induction length of the second-stage reaction, Δ_{I2}, equal 323.2 μm. Heat release in region $x < x_m$ is caused by the first-stage reaction and is defined as Q_1. Correspondingly, heat release caused by the second-stage reaction in region $x > x_m$ is defined as Q_2. $Q_2/Q_1 = 1.78$ for $\kappa = 0.6$. In Fig. 3(b), pressure and temperature profiles of the ZND structure are also depicted. Pressure at the Von Neumann peak position (i.e., $x = 0$) equals 29.8 atm and the mixture temperature equals 1930 K behind the leading shock. Then the mixture temperature rises as the two-stage reactions proceed. Meanwhile, mixture expands thermally, leading to the pressure decreasing behind the leading shock.

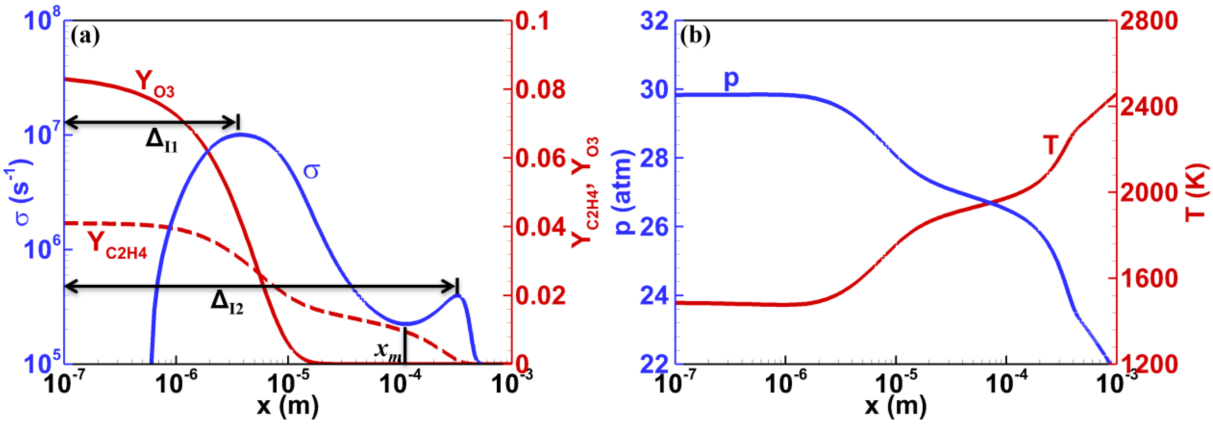

Fig. 3. (a) Thermicity, species mass fractions, (b) pressure and temperature profiles for the ZND structure of $\kappa = 0.6$. The leading shock locates at $x = 0$. Δ_{I1}: induction length of the first-stage reaction; Δ_{I2}: induction length of the second-stage reaction; x_m: position of the local minimum of thermicity between the two-stage reactions.

To investigate how κ affects the detonation structure, thermicity profiles of ZND structures for $\kappa = 0.2$, 0.4, 0.6, and 0.8 are plotted in Fig. 4(a). The results indicate that the reactions proceed in two stages for all cases. The ratio of induction lengths between the second-stage reaction and the first-stage reaction, Δ_{I2}/Δ_{I1}, is defined to examine the correlation between the two-stage reactions. When Δ_{I2}/Δ_{I1} is large, the reactions of the two stages proceed more independently, which promotes the formation of the double cellular detonation wave. Figure 4(b) shows that as κ increases from 0.2 to 0.4, 0.6, and 0.8, Δ_{I2}/Δ_{I1} decreases from 158.7 to 126.4, 86.1, and 53.7, respectively. Hence, for the case of $\kappa = 0.8$ shown in Fig. 4(d), the reactions of the two stages proceed in a highly coupled manner, thereby no obvious double cellular structures are observed (see Fig. 2d).

In Fig. 4(b), the ratios of heat release in the second stage to that in the first stage, Q_2/Q_1, are also calculated. The results show that as κ increases, Q_2/Q_1 decreases and trends toward 1. According to Khasainov et al.'s work [11], similar heat effects from both reaction stages facilitate the formation of double cellular detonation waves. For $\kappa = 0.2$, the heat release from the first-stage reaction accounts for only 24.2% of the total value, which is too low to sustain detonation wave propagation. As a result, the triple-shock structures cannot survive; their trajectories weaken during the detonation wave propagation, and no obvious small detonation cells are observed (see Fig. 2a).

In summary, qualitatively, two conditions are required for the formation of double cellular detonation: the reactions of the two stages must proceed as independently as possible, and the heat releases from both stages must be high enough to support detonation wave propagation. Consequently, double cellular detonation waves are observed only for the cases of $\kappa = 0.4$ and 0.6 in this subsection, and the influence of κ on the cellular detonation structure is non-monotonic.

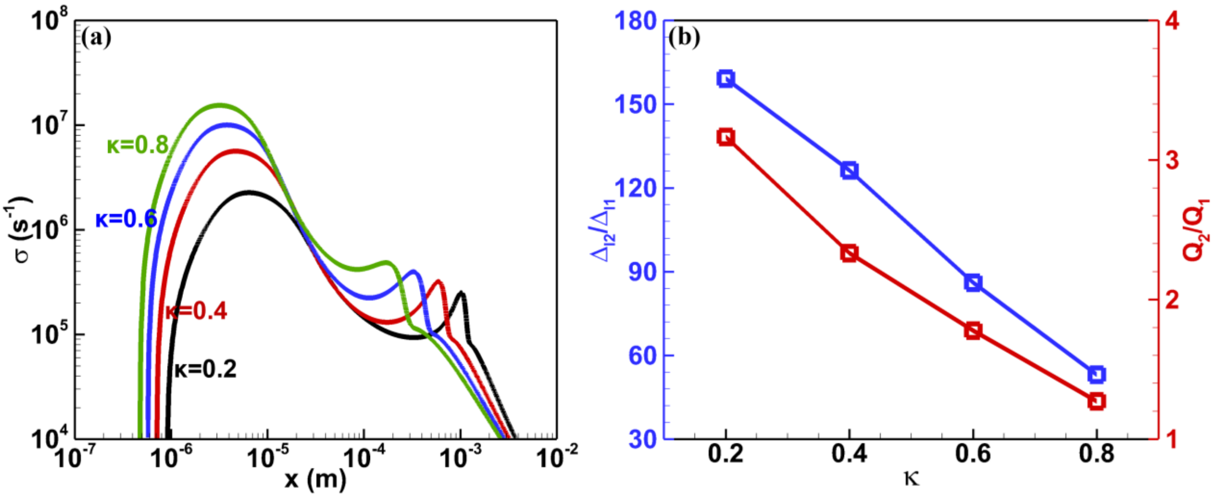

Fig. 4. (a) Thermicity profiles, (b) Δ_{I2}/Δ_{I1} and Q_2/Q_1 for different κ. Δ_{I2}/Δ_{I1}: ratios of induction lengths between the second-stage reaction and the first-stage reaction; Q_2/Q_1: ratios of the heat release in the second stage to that in the first stage.

4 Conclusions

In this work, two-dimensional simulations incorporating detailed chemistry are conducted in $C_2H_4/O_2/O_3/N_2$ mixtures to study cellular detonation waves. The parameter κ, defined as $3X_{O3}/(2X_{O2}+3X_{O3})$, is used to regulate the exothermic process behind the leading shock. Cellular detonation waves for different κ are simulated. The results indicate that the influence of κ on the cellular detonation structure is non-monotonic. Specifically, when $\kappa = 0.2$, the heat released by the first-stage reaction is too small to support an independent cellular detonation structure. As κ increases to 0.4 and 0.6, double cellular detonation waves are observed, with the double detonation cells resulting from the two-stage reactions. When κ further increases to 0.8, the ratio of induction lengths between the second-stage reaction and the first-stage reaction decreases to a small value. The reactions of the two stages proceed in a highly coupled manner behind the leading shock, and the detonation wave propagates with a single cellular structure. Thus, two conditions are required to form double cellular detonation: the reactions of the two stages must proceed as independently as possible, and both heat releases from the two stages must be high enough to sustain detonation wave propagation.

It is important to note that the current work has some limitations. Ozone is used to artificially regulate the two-stage reaction process, and the ozone content used in the simulations is much higher than that used in practical applications. Furthermore, due to the multiscale features of the double cellular detonation wave, both a small grid size and a large simulation domain are required. The grid size used in this work is still insufficient to fully resolve the small wave structures, and the simulation domain is limited, preventing the detonation wave from fully developing. In future work, more realistic mixtures and finer simulation resolutions will be considered.

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