Postprint: Study on Flame Instability under Low-Temperature Chemistry Effects

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Date: 2017-10-17T00:00:00+00:00

Abstract

Considering detailed chemical reactions and transport processes, direct numerical simulation of the incompressible Navier-Stokes equations was conducted to study flame hydrodynamics and cellular instability under low-temperature chemical influences. The results indicate that the disturbance growth rate of flames in preheated mixtures that have undergone low-temperature chemical reactions is significantly higher than that of flames in mixtures without low-temperature reactions. Analysis of key parameters of the preheated mixture flame reveals that, compared with the non-preheated mixture flame, the dimensionless heat release, Lewis number, and Zeldovich number all decrease substantially. According to existing theory, under conditions where the Lewis number exceeds unity, the reduction of these parameters attenuates the inhibitory effect of cellular instability on disturbance growth. Consequently, the growth rate of the preheated flame increases markedly. Simultaneously, the formation of numerous free radicals and other small molecules in low-temperature reactions reduces the Lewis number of the preheated mixture flame, thereby decreasing both the depth and height of its cellular flame structure.

Full Text

Preamble

Studies of Low-Temperature Chemistry Effects on Flame Instabilities

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Abstract

Direct numerical simulations of the incompressible Navier-Stokes equations were performed with detailed chemical kinetics and transport properties to investigate hydrodynamic and cellular instabilities in flames influenced by low-temperature chemistry. The results demonstrate that the disturbance growth rate of flames with preheated mixtures undergoing low-temperature chemical reactions is significantly greater than that of flames without low-temperature reactions. Analysis of key flame parameters reveals that compared with non-preheated flames, the dimensionless heat release, Lewis number, and Zeldovich number all decrease substantially for preheated mixtures. According to existing theory, when the Lewis number exceeds unity, these parameter reductions weaken the inhibitory effect of cellular instability on disturbance growth. Consequently, the growth rate increases markedly for preheated flames. Additionally, the formation of numerous free radicals and small molecules during low-temperature reactions reduces the Lewis number of the preheated mixture, resulting in smaller depth and height of the cellular flame structures.

Keywords: low-temperature chemistry; DME/air; hydrodynamic instabilities; cellular instabilities

Funding: National Natural Science Foundation of China (No. 91441119, No. 91441131)

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0 Introduction

Premixed flame hydrodynamic and cellular instabilities typically appear in a coupled manner. When the Lewis number is less than unity, cellular instability amplifies hydrodynamic instability, whereas for Lewis numbers greater than unity, cellular instability exerts a suppressive effect. Hydrodynamic instability occurs across all wavenumbers, though it is suppressed at very low wavenumbers when flame thickness and stretch effects are considered. These instabilities induce flame surface wrinkling, which increases flame propagation speed—an important mechanism for flame self-turbulization and deflagration-to-detonation transition. In modern advanced engines, pressurization and lean premixed combustion are widely employed. Increased pressure significantly reduces the ignition delay time of premixed gases, particularly for large-molecular-weight fuels such as aviation kerosene, gasoline, and diesel, which exhibit pronounced low-temperature chemical reactions. Consequently, high pressure and elevated preheat temperatures intensify low-temperature reactions while continuously decreasing low-temperature ignition delay times. When the low-temperature reaction timescale becomes comparable to the large-scale flow timescale in the combustion chamber, the flame dynamics are influenced by low-temperature chemistry [1]. In this scenario, the unburned mixture consists not of the original oxidizer and fuel, but rather of intermediate products from low-temperature reaction pathways along with residual fuel and oxidizer. Previous studies on hydrodynamic and cellular instabilities have focused exclusively on high-temperature combustion, with no consideration of instability mechanisms during flame propagation under low-temperature reaction conditions. However, in practical combustion chambers, when low-temperature reactions occur, the chemical and thermal properties of the mixture change significantly, which must alter the mechanisms of flame propagation instability. This paper investigates flame propagation instabilities in lean premixed dimethyl ether (DME)/air mixtures under the influence of low-temperature chemistry.

[Figure 1: see original paper]

1 Physical and Mathematical Model

This study considers two-dimensional, unsteady reactive flow. The incompressible Navier-Stokes equations are solved directly using a fractional-step projection method with fourth-order spatial discretization and a Crank-Nicolson scheme for the unsteady term. The chemical reaction mechanism employs a simplified, stiffness-free mechanism that has been successfully used in previous DNS studies [2], comprising 30 species and 175 reactions. Throughout the computations, the Chemkin software package [3] is coupled to account for detailed chemical kinetics, while the Transport package [4] is coupled to accurately compute species transport. Unlike previous DNS studies using this mechanism, the present work considers detailed species transport processes. The physical model is a two-dimensional planar flame with unburned mixture inlet on the left side and burned gas outlet on the right side, while periodic boundary conditions are applied at the top and bottom boundaries.

2 Results and Discussion

For large-molecular-weight fuels, significant low-temperature chemical reaction processes occur in the negative temperature coefficient (NTC) region. This study selects dimethyl ether (DME), a simple surrogate fuel that exhibits these low-temperature reaction characteristics. The reduced chemical mechanism with 30 species and 175 reactions has been validated against detailed mechanisms for flame propagation speed, ignition delay time, and S-curves including ignition and extinction turning points under various pressures and equivalence ratios. The comparison confirms that this mechanism accurately simulates flame behavior under both low- and high-temperature reaction conditions. Figure 2 [Figure 2: see original paper] presents ignition delay curves for DME/air mixtures at an equivalence ratio of 0.5 and pressures of 0.1 MPa and 1.5 MPa, where solid lines represent high-temperature ignition and dotted lines represent low-temperature ignition. In the NTC region, low-temperature reactions become prominent, resulting in low-temperature ignition with shorter delay times. In modern advanced engines, as pressure and preheat temperature increase, the high-temperature ignition delay time shortens, while the low-temperature ignition delay time at high pressure becomes even shorter. Therefore, before the mixture enters the high-temperature reaction zone, low-temperature chemical reactions have already occurred, and low-temperature ignition may have even taken place. This study investigates a two-dimensional planar DME/air flame at an initial temperature of 600 K and equivalence ratio of 0.5 using direct numerical simulation with detailed chemistry and transport to examine the effects of low-temperature reactions on flame hydrodynamic and cellular instabilities. Figure 3 [Figure 3: see original paper] shows the temporal evolution of heat release rate and important intermediate species.

Figure 3 illustrates the homogeneous (0D) DME/air autoignition process in the NTC region. Since the selected condition lies within the NTC region, low-temperature ignition is clearly observed at 0.1 s, accompanied by substantial HO formation. During low-temperature reactions, DME first reacts with O to form CH OCH (R) and HO through RH + O → R + HO , while OH also abstracts H from RH to produce R via RH + OH → R + H O. The resulting R reacts with O in the crucial reversible reaction R + O RO . Under low-temperature conditions, the forward reaction dominates, and subsequent chain-branching reactions initiated by RO lead to low-temperature ignition. The large HO production thus indicates simultaneous generation of R and RO , triggering low-temperature ignition. Concurrently, significant H O formation occurs during low-temperature ignition due to the reactions RH + HO → R + H O and HO + HO → H O + O . Accumulation of H O subsequently leads to the reaction H O → 2OH at elevated temperatures, initiating high-temperature ignition. Consequently, rapid H O consumption is observed immediately before high-temperature ignition, while OH rises sharply at the high-temperature ignition point.

Figure 4 [Figure 4: see original paper] shows the propagation process of a DME/air premixed flame at an initial temperature of 600 K and equivalence ratio of 0.5 without considering low-temperature reactions. At the initial moment, a planar flame with a sinusoidal perturbation is imposed. The computational domain length is 60 flame thicknesses, and the height is one wavelength corresponding to the most unstable wavenumber determined by the flame Lewis number. The flame wrinkles under hydrodynamic instability, reaching a nearly stable configuration after 50 time units. With a flame Lewis number of 1.68 based on the mixture' s thermal diffusivity and fuel mass diffusivity, cellular instability stabilizes the flame by reducing flame speed at protrusions and increasing it at indentations. As the flame deformation increases (i.e., stretch rate increases), the local flame speed increases under diffusion-thermal effects, eventually forming stable cellular structures. Convex flame sections quickly stabilize as their deceleration at low stretch rates matches the upstream flow field. To further investigate the temporal growth of the initial perturbation amplitude A , solutions based on theories by Matalon et al. [5] and Landau [6] are compared with computational results. Figure 5 [Figure 5: see original paper] shows that the computed growth rates agree well with the dispersion relation from Matalon et al., while Landau' s theory, which neglects cellular instability effects, significantly overpredicts the growth rate. This confirms that cellular instability substantially suppresses disturbance growth induced by hydrodynamic instability at the present Lewis number.

To investigate low-temperature chemistry effects on these flame instabilities, the inlet at the left boundary of the planar flame is supplied with mixture preheated at 600 K for 0.4 s. Figure 6 [Figure 6: see original paper] clearly demonstrates that the evolution of the preheated mixture flame differs significantly from that shown in Figure 4. After 15 time units, the flame perturbation ceases to amplify, and both the depth and height of cellular structures are noticeably smaller than those of the non-preheated flame. A comparison of disturbance growth between preheated and non-preheated flames reveals substantially larger growth rates for the preheated mixture flame, as shown in Figure 7 [Figure 7: see original paper].

To explain this behavior, key parameters before and after preheating are analyzed in Table 1 . The Zeldovich number decreases significantly after preheating, indicating that initiating low-temperature reactions reduces the overall activation energy and relatively thickens the reaction zone. Meanwhile, the reduction in dimensionless heat release Q suggests that low-temperature reactions, particularly low-temperature ignition, convert part of the chemical energy into thermal energy release.

For the Lewis number, this study employs the method from reference [7] based on Markstein length calculations. Table 1 shows that the Lewis number of the preheated mixture flame decreases substantially. This reduction occurs because low-temperature reactions consume considerable fuel while generating small free radicals and smaller molecules such as H , CO, and CH . Lec represents the critical Lewis number below which cellular instability promotes disturbance growth. The preheated mixture flame requires a smaller Lewis number for cellular instability to enhance disturbance growth.

These changes in key flame parameters alter the fundamental instability characteristics of preheated flames. Based on theoretical analyses by Landau and Matalon, Figures 8 [Figure 8: see original paper] and 9 [Figure 9: see original paper] illustrate the relationships between dimensionless heat release, Lewis number, Zeldovich number, and flame instabilities. and represent the contributions of hydrodynamic and cellular instabilities to the total disturbance growth rate, respectively. Increasing heat release enhances hydrodynamic instability; however, for Lewis numbers greater than unity, cellular instability' s suppression of disturbance growth intensifies with increasing heat release, particularly at larger Lewis numbers. Moreover, increasing Zeldovich number strengthens cellular instability' s inhibitory effect on disturbance growth. Consequently, compared with preheated flames, non-preheated mixtures exhibit greater Lewis numbers, heat release Q, and Zeldovich numbers, resulting in stronger suppression of disturbance growth by cellular instability under Lewis $>$ 1 conditions. Therefore, preheated mixture flames display larger growth rates. Simultaneously, the reduced Lewis number leads to smaller cellular structure depth and height, consistent with previous numerical studies on high-temperature flames using one-step global reactions and simple transport relationships for various Lewis numbers.

This study conducted direct numerical simulations of two-dimensional planar lean premixed DME/air flames by solving the Navier-Stokes equations coupled with detailed chemical kinetics and transport processes. The chemical mechanism accurately captures DME's low-temperature reaction characteristics, while detailed transport modeling improves the accuracy of diffusion-thermal-induced cellular instability predictions. The work compares disturbance evolution between flames established with preheated mixtures that have undergone low-temperature ignition and those without low-temperature reactions. The results demonstrate significantly increased disturbance growth rates for preheated mixture flames. Analysis of Lewis numbers (calculated based on stretched and planar flames), Zeldovich numbers, and dimensionless heat release reveals substantial reductions in these parameters for preheated flames. According to existing theoretical analysis, these reductions indicate that cellular instability suppresses disturbance growth driven by hydrodynamic instability under Lewis > 1 conditions, thereby elucidating the interaction mechanisms between low-temperature chemistry and flame instabilities. Additionally, the decreased Lewis number results in noticeably reduced depth and height of cellular structures in preheated mixture flames.

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Note: Figure translations are in progress. See original paper for figures.