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Two-stage heat release in nitromethane/air flame and its impact on laminar flame speed measurement

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ABSTRACT

In premixed flames of most hydrocarbon fuels, there is only one stage heat release. However, two-stage heat release occurs in premixed nitromethane/air flames under certain conditions. In this study, numerical simulations were conducted for one-dimensional planar and spherical nitromethane/air flames at different initial temperatures (423∼800 K), pressures (0.5∼10 atm) and equivalence ratios (0.5∼1.3). Using the planar flame, we investigated the characteristics of the two-stage heat release and identified elementary reactions involved in these two stages. It was found that the occurrence of two-stage heat release strongly depends on the equivalence ratio and that single-stage heat release occurs for very fuel-lean mixture. To demonstrate the key reactions involved in the second stage heat release, we modified the original chemical mechanism and compared the results predicted by different mechanisms. The second stage heat release was found to be mainly caused by the reaction NO+H→N+OH. Using the propagating spherical flame, we assessed the impact of two-stage heat release on the determination of laminar flame speed. The positive burned gas speed induced by the second stage heat release was shown to affect the accuracy of laminar flame speed determined by traditional method neglecting burned gas speed and using the density ratio at equilibrium condition. Alternative methods were proposed and used to correct the experimental data reported in the literature.

1. Introduction

Nitromethane (CH_3NO_2) can be used as a fuel additive in internal combustion engines (ICEs) and as a monopropellant in rocket engines. Blending nitromethane into gasoline helps to increase the octane number and to prevent knocking in direct-injected boosted gasoline engines [1]. Recent experiments showed that nitromethane addition to gasoline reduces soot formation but increases the emission of HCN and NO_x [2]. Besides, since nitromethane is the simplest nitro paraffin fuel, it is popularly used to study the combustion properties of liquid propellants [3].

Unlike the combustion of traditional fuels, nitromethane combustion proceeds via two or three steps. Hall and Wolfhard [4] first observed two luminous reaction zones in a low-pressure nitromethane/oxygen flame. Boyer and Kuo [3] simulated one-dimensional (1D) nitromethane combustion with surface vaporization. They identified three distinct reaction regions based upon the appearance and consumption of certain species. Nauclér et al. [5] found that fuel-rich nitromethane/air flames have two separate reaction zones according to their numerical simulation considering detailed chemistry. In shock tube experiments and modeling of homogenous ignition process, two-stage heat release was also observed by Mathieu et al. [6] and Nauclér et al. [7]. Besides, a double cellular structure was observed in the detonation experiments for gaseous nitromethane/oxygen mixtures [8]. Sturtzer et al. [9] calculated the 1D ZND detonation structure using a detailed mechanism for nitromethane. They found that heat is released in two main successive reaction steps characterized by their own induction length, which justifies the existence of a two-level detonation cellular structure [9].

Though the two or three step heat release in nitromethane combustion was identified in previous studies listed above, the chemical reactions responsible for the multi-stage heat release in nitromethane/air flames are still not well known. This motivates the present work, whose first objective is to investigate the two-stage heat release and the corresponding reactions in premixed, planar nitromethane/air flames. The influence of equivalence ratio, initial pressure and initial temperature on the two-stage heat release in nitromethane/air flames is also investigated.

Laminar flame speed is popularly used to validate and develop the chemical mechanism of different fuels [10,11]. Due to its simple configuration and well-defined stretch rate, the propagating spherical flame method is popularly used to measure the

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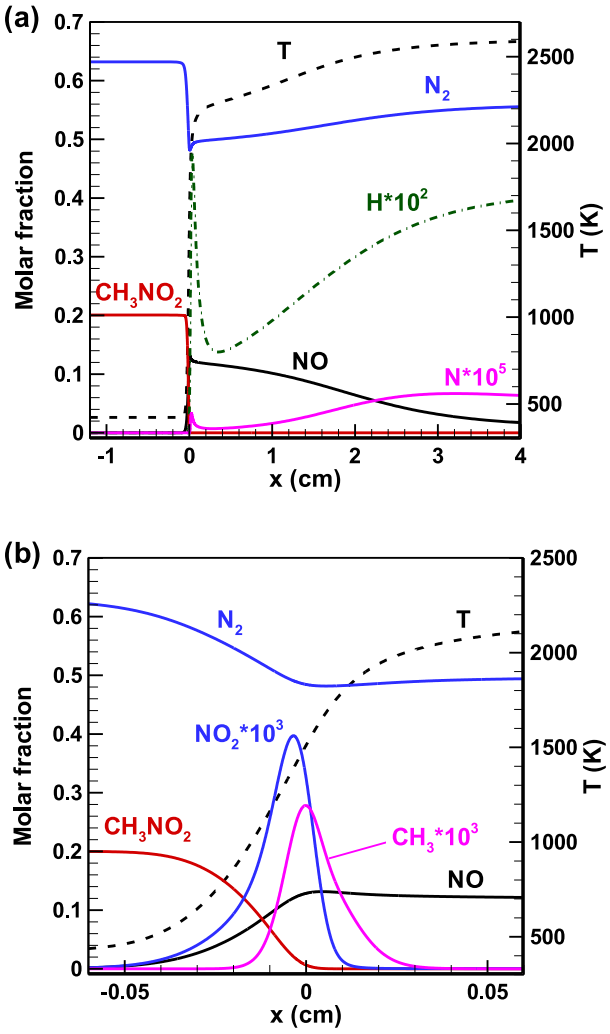

Fig. 1. Distributions of temperature and molar fraction of different species in premixed nitromethane/air flame with $\phi=0.9$, $T_u = 423$ K and $P=1$ atm and (b) magnifies the results corresponding to the first stage heat release around $x=0$ cm.

laminar flame speed, especially at high pressures [12–14]. For nitromethane/air mixture, Brequigny et al. [15] measured laminar flame speed from propagating spherical flames at different initial pressures and equivalence ratios. However, Nauclér et al. [5] pointed out that it is difficult to accurately measure the laminar flame speed from propagating spherical nitromethane/air flames due to the two-stage heat release and large flame thickness. Currently, it is not clear how and why the two-stage heat release affects the laminar flame speed measurement of nitromethane/air from propagating spherical flames. Therefore, the second objective of the present study is to answer this question by studying propagating spherical flames in nitromethane/air mixtures.

Both 1D planar and 1D spherical premixed nitromethane/air flames are considered in this study. Using the planar flame, we shall investigate the characteristics of the two-stage heat release and the chemical reactions involved in these two stages. Using the spherical flame, we shall explain how the two-stage heat release affects the laminar flame speed determination. For both planar and spherical flames, the effects of equivalence ratio, initial pressure and initial temperature are examined.

2. Numerical methods

As mentioned before, two 1D premixed flame configurations are considered in this study: one is the unstretched freely-propagating planar flame and the other is the positively-stretched propagating spherical flame. The PREMIX code [16] was used to simulate the planar flame and to get the flame structure and unstretched laminar flame speed. Thermal diffusion and multicomponent molecular transport model were included in all simulations. The number of grid points was always kept to be above 1000 so that the flame structure was well resolved and the results were grid independent. Since nitromethane/air flame with two-stage heat release has very large flame thickness, a large domain size of 50 cm was used in simulation.

The propagating spherical flame was simulated using the in-house code A-SURF [17–19]. A-SURF solves the conservation equations for compressible, multicomponent, reactive flow in a spherical coordinate by the finite volume method. The CHEMKIN package [20] was incorporated into A-SURF to calculate the thermodynamic properties, transport properties and reaction rates. A-SURF was successfully used in previous studies on flame and detonation propagation [13,21–28]. The readers are referred to Refs. [17–19] for details on numerical schemes and code validation of A-SURF. Dynamic adaptive mesh was used to accurately resolve the propagating spherical flame front, which were always fully covered by the finest mesh with the size of 8 μm. To avoid confinement effect [17,29], a large chamber radius of $R_w = 50$ cm was used in all simulations. As summarized in Ref. [13], there are different factors affecting the accurate measurement of laminar flame speed from propagating spherical flames. In the present study, we focused on the influence of two-stage heat release and thereby other factors such as radiation, flame instability and nonlinear stretch behavior were not considered.

In the literature, there are several chemical mechanisms for CH_3NO_2 [6,15,30–32] and NO_2 ([40] and references therein). Since the mechanism of Brequigny et al. [15] containing 88 species and 701 reactions was validated against the laminar flame speed data of CH_3NO_2 at $T_u = 423$ K, $P=0.5\sim3.0$ bar and $\phi=0.5\sim1.3$ and it yields reasonably well prediction, it was used in all simulations in this study.

Two global reactions for nitromethane/air combustion were proposed in the literature [31,33]:

$$CH_3NO_2+0.75(O_2+3.78N_2)= CO_2+1.5H_2O + 3.32N_2 \quad (1)$$

$$CH_3NO_2+1.25(O_2+3.78N_2)= CO_2+1.5H_2O + NO + 4.7N_2 \quad (2)$$

Both global reactions have been used recently [5,6,15]. In Eq. (1), N_2 is the only product containing nitrogen atom [15], while Eq. (2) indicates that NO is a stable product [5]. According to the results to be presented in this study, neither Eq. (1) nor Eq. (2) is unanimously accurate: in fuel-lean nitromethane/air flames, NO is a stable product; while for rich flames NO is converted into N_2. Nevertheless, the equivalence ratios according to Eqs. (1) and (2) have a linear relationship of $\phi_{Eq.\,(1)}=0.6\phi_{Eq.\,(2)}$ and thereby they can be directly converted into each other. Since the only experimental study on propagating spherical flames in nitromethane/air was conducted by Brequigny et al. [15] in which Eq. (1) was used, we also used Eq. (1) in this study. Note that similar to Brequigny et al. [15], we considered the synthetic air with 79.1% N_2 and 20.9% O_2 and thereby the molar ratio of N_2 to O_2 is 3.78 in Eqs. (1) and (2).

3. Two-stage heat release in premixed planar nitromethane/air flame

We first investigated the two-stage heat release in premixed planar nitromethane/air flames.

Figure 1 shows the flame structure of premixed nitromethane/air with $\phi=0.9$, $T_u = 423$ K and $P=1$ atm. The temperature profile indicates that there are two-stage heat release: the first stage