Initiation and propagation of laminar premixed cool flames

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ABSTRACT

Cool flames, being essential features of chemical kinetics of large hydrocarbon fuels, are closely related to the negative temperature coefficient (NTC) phenomenon and engine knock. In this work, the coupling of cool flame chemistry and convective–diffusive transport is computationally and experimentally investigated. A 1-D planar premixed cool flame induced by a hot pocket is first simulated for $DME/O_2/N_2$ mixtures with detailed chemistry and transport, demonstrating the existence of a residence time window for quasi-steady propagation. Then with residence time limited by aerodynamic straining, a steady-state premixed cool flame is simulated in a counterflow of heated N_2 against a $DME/O_2/N_2$ mixture. It is found that with a high strain rate, corresponding to short residence time, low-temperature heat release is suppressed, resulting in a stretched low-temperature S-curve system response; and that with a sufficiently low strain rate, corresponding to long residence time, ignition induced by low-temperature chemistry would transition to a high-temperature, intensely burning flame. Consequently, a steady-state premixed cool flame exists only for residence time in a strain rate window. A symmetric counterflow configuration is then simulated to determine the cool flame temperature and flame speed at a fixed local strain rate, showing very different controlling chemistry and characteristics as compared to the normal laminar flames governed by high-temperature chemistry. In a companion experimental investigation, premixed cool flames in the counterflow were observed with a high-sensitivity CCD camera in the UV spectrum, with/without a bandpass filter corresponding to the characteristic wavelength of excited HCHO. The chemiluminescence from the cool flame is found to become more intense with increasing equivalence ratio, even for rich mixtures, while the position of the cool flame is insensitive to variation in the equivalence ratio at the same strain rate. These observations qualitatively agree with the numerical simulations, demonstrating the essential features of premixed cool flames.

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1. Introduction

Cool flames, discovered in 1817 [1], have been studied extensively [2]. Its controlling kinetics of the low-temperature chemistry [3] has been shown to be closely related to the phenomena of two-stage ignition and the negative temperature coefficient (NTC) [4], associated with the autoignition [5] and engine knock of large hydrocarbon fuels [6]. They are normally characterized by faint bluish luminescence from excited formaldehyde [7], relatively small heat release and very low CO_2 production. Furthermore, contrary to the normal hot flames in which chemical reactions and transport are strongly coupled, cool flames have been mostly observed in homogeneous systems such as heated closed reaction vessels [8] and jet-stirred reactors [9], exhibiting oscillatory volumetric heat release and chemiluminescence.

Noting that gas-phase nonuniformities invariably exist in practical systems, recently there has been considerable interest in understanding the coupled effects of cool flame chemistry and transport, leading to the postulation and identification of this class of laminar flames whose flame temperature can be substantially lower than the adiabatic flame temperature corresponding to the (near-)complete consumption of the controlling reactant(s). In particular, Law and Zhao [10] and Zhao and Law [11] demonstrated computationally that with low strain rates and/or high pressures, low-temperature chemistry can strongly couple with transport to induce a secondary S-curve on the lower branch of the conventional (primary) S-curve, with its own distinct ignition and extinction states. Subsequently, using infrared detection Deng et al. [12] experimentally observed a diffusion cool flame in the nonpremixed counterflow at atmospheric pressure. Furthermore, diffusion cool flames have been suggested to explain some experimental results on microgravity droplet combustion [13,14], and have also been

directly observed in experiments on plasma-assisted combustion [15].

Recognizing that recent investigations [10–15] on the coupled behavior of low-temperature chemistry and convective–diffusive transport have been mainly on *diffusion* flames, it behooves us to explore if steady, *premixed* cool flames can also be initiated and established, and if so, what are the associated flame structure and propagation rates, including the role of the two-stage ignition delay in establishing such flames. The relevant issue here is that while a diffusion flame is primarily controlled by the counter-diffusion of the fuel and oxidizer reactants, with chemistry playing a secondary role in the global flame characteristics, the structure and propagation of premixed flames inherently depend on the coupled influence of chemical kinetics and transport [16]. Consequently an assessment of the essential role of low-temperature chemistry on flames must necessarily involve studies on premixed flames. In fact, because of the very weak exothermicity of the low-temperature chemistry, and the inevitable heat loss in a flame system, it is not evident *a priori* that a flame embodying this chemistry can be readily initiated and support steady propagation. It is also noted that while premixed cool flames have been studied previously, for example as early as the 1960s [17,18], the increased understanding of flame structures since then is expected to yield further insight and interpretation of this important phenomenon.

The present study consists of three tasks. We first extend, in the next section, the classical NTC phenomenon describing the transient *ignition* of a *homogeneous* mixture to one in which ignition is accomplished by a localized heat source in the 1-D planar domain, which is inherently *nonhomogeneous* in nature and is affected by both chemistry and diffusive transport. Consequently the interest here is the dynamics and chemistry of the ignition process leading to low-temperature flame propagation and its subsequent transition to the high-temperature flame. We shall also show that the transient ignition event depends sensitively on the temperature of the ignition source when low-temperature chemistry is involved.

In order to capture and stabilize the low-temperature flame identified in Section 2, in Section 3 we computationally explore the possible existence of a premixed flame of low-temperature chemistry in the counterflow, with its characteristic residence time controlled by the strain rate of the flow. We shall show that such a flame is predicted to exist, with its chemical structure including the states of ignition and extinction depending sensitively on the strain rate of the flow. Our third task, presented in Section 4, provides the first experimental observation that such stretch-stabilized premixed cool flames indeed exist, thereby brings more insights on the existence and characteristics of laminar cool flames, both nonpremixed [10–15] and premixed [17–20].

The fuel selected for the study is dimethyl ether (DME), which is one of the smallest fuel molecules exhibiting the NTC behavior. The reaction mechanism adopted in the computation is a validated reduced model [21] consisting of both low- and high-temperature kinetics; the associated validation and applications are reported in [12,22].

2. Initiation and propagation of the 1-D planar cool flame

We first examine the adiabatic and isobaric autoignition delays of homogeneous stoichiometric DME/O_2/N_2 mixtures, in order to select the proper conditions to study the ignition and propagation of 1-D planar flames. Fig. 1(a) shows the calculated first-stage, second-stage, and total ignition delay times for initial temperature T, denoted as τ_1, τ_2 and τ respectively. It is seen that τ varies nonmonotonically with increasing T, and hence demonstrates the NTC behavior. Consequently, for an appropriate total ignition delay
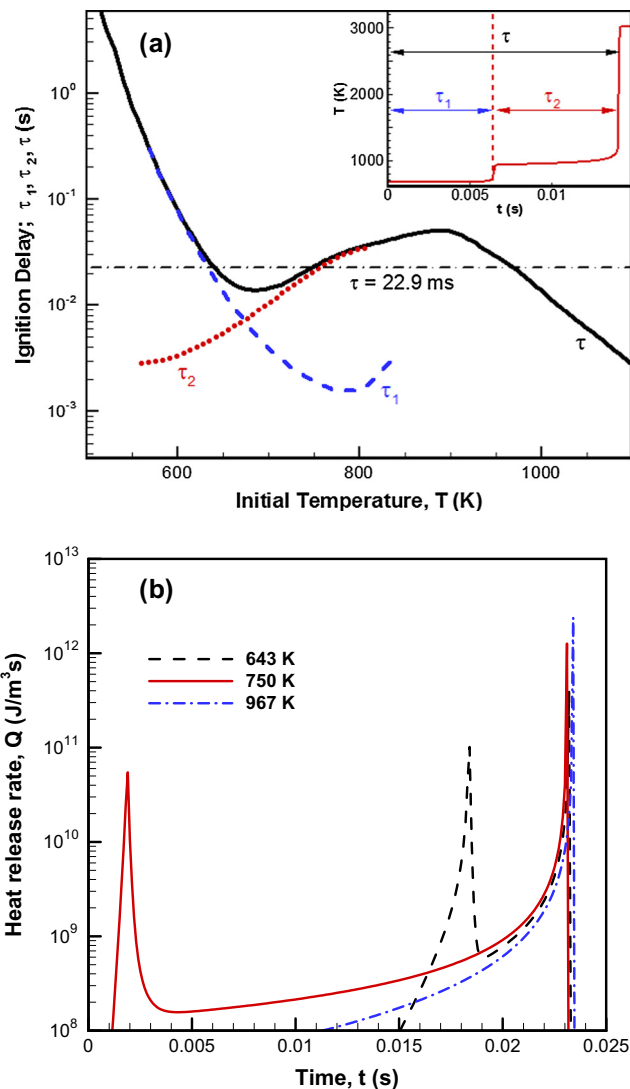

Fig. 1. (a) First-stage, second-stage, and total ignition delays of DME/O_2/N_2 mixture with 15.8% N_2 in volume, under 1 atm. The inset shows the definitions of various ignition delays in a two-stage ignition process. (b) Heat release rate history for ignition processes at 643 K, 750 K and 967 K.

time, say $\tau = 22.9$ ms, there are three distinct ignition situations: at the low temperature state of $T = 643$ K, τ_1 is much longer than τ_2; at the intermediate temperature state of $T = 750$ K, τ_2 is much longer than τ_1; and finally at the high temperature state of $T = 967$ K, the two-stage ignition behavior disappears and combustion is totally taken over by the intermediate-to-high temperature chemistry, exhibiting a single-stage ignition event. These three distinct ignition situations are further characterized by their correspondingly distinct heat release rates, Q, as shown in Fig. 1(b). Specifically, for the low and intermediate temperatures of 643 K and 750 K, heat release occurs in two bursts, corresponding to the first and second stage ignition. Furthermore, the first ignition delay for the 750 K case occurs more readily than that of the 643 K case, as expected. For the high-temperature case of 967 K, there is no first-stage ignition and as such ignition occurs in a single burst, through the second ignition delay chemistry. It is also clear that Q is much lower both before and after the respective ignition bursts.

Based on these observations, we explore how NTC would affect the initiation and propagation of a 1-D planar flame induced by an ignition kernel with these three different temperatures. The