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Low-temperature multistage warm diffusion flames

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ABSTRACT

We report on experimental evidence of the existence of a new self-sustaining low-temperature multistage warm diffusion flame, existing between the cool flame and hot flame, at atmospheric pressure in the counterflow geometry. The structure of multistage warm diffusion flames was examined by using thermometry, laser-induced fluorescence, and chemiluminescence measurements. It was found that the warm diffusion flame has a two-staged double flame structure, with a leading diffusion cool flame stage on the fuel side and a second intermediate stage on the oxidizer side, with strong heat release in the second stage that can be comparable to that of the first stage. The results demonstrate that the spatially-distinct multistage character is due to the low-temperature fuel reactivity that allows for the production of reactive intermediates in a leading cool flame. These intermediates are then oxidized, on the oxidizer side, in a second stage via intermediate-temperature chemistry. In the case of dibutyl ether, the low-temperature peroxy branching pathway supports the first cool flame oxidation stage and produces intermediates such as alkyl and carbonyl radicals. The alkyl and carbonyl radicals then react with the hydroperoxyl radical and molecular oxygen to form the second oxidation stage. A detailed analysis revealed that ozone addition in the oxidizer promotes the second stage oxidation by increasing both the radical pool population and the flame temperature, but does not fundamentally change the multistage flame structure. Furthermore, the analysis revealed that with the increase of fuel concentration, a single-stage cool flame can ignite to a warm flame or a hot flame. Moreover, a warm flame can extinguish into a cool flame or ignite to a hot flame when the fuel concentration is substantially reduced or increased, respectively. Finally, under certain conditions, a hot flame can extinguish directly into either a warm flame or a cool flame. Hence, the results suggest that the multistage warm flame can act as a critical bridge between cool flames and hot flames and that it is a fundamental burning mode characteristic of low-temperature non-premixed combustion. The multistage warm diffusion flame is particularly relevant to combustion in highly turbulent flow fields and in microgravity environments, owing to the possibility of long residence times.

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1. Introduction

The chemical character of conventional high-temperature flames has been thoroughly investigated by analytical, experimental, and computational methods [1]. For finite chemical time scales, the convective, diffusive, and radiative heat losses dictate the domain of existence and burning intensity of such flames. The coupling with highly nonlinear chemical kinetics leads to the classical hysteretic behavior where critical limits (extinction and ignition) are uniquely defined in the case where the characteristic reaction time scales are much longer or shorter compared to the characteristic transport time. In most fuels of practical interest, it is known that the parent fuel can undergo oxidation via a variety

of different reaction classes such as high-temperature small radical chemistry, intermediate-temperature peroxide chemistry, and low-temperature peroxy chemistry [2]. The competition between different reaction classes at different temperatures results in interesting kinetic behavior such as negative temperature coefficient (NTC) reactivity, in which the overall rate of fuel oxidation is non-monotonic with respect to temperature. The result of such kinetics is the existence of multiple flame regimes, such as conventional high-temperature (hot) flames and low-temperature (cool) flames. These different flames can manifest themselves as multiple points of criticality on the classical S-curve [3–5] in which the overall reactivity, demarcated by some progress variable, is determined as a function of Damköhler number. Indeed, it is such chemical complexity, aided by the disparity in characteristic activation energies of the competing reactions, that results in separated reaction chemistry in a single reaction zone [6]. Specifically, recent simulations [7–9] and experiments [10] have shown that, for fuels with

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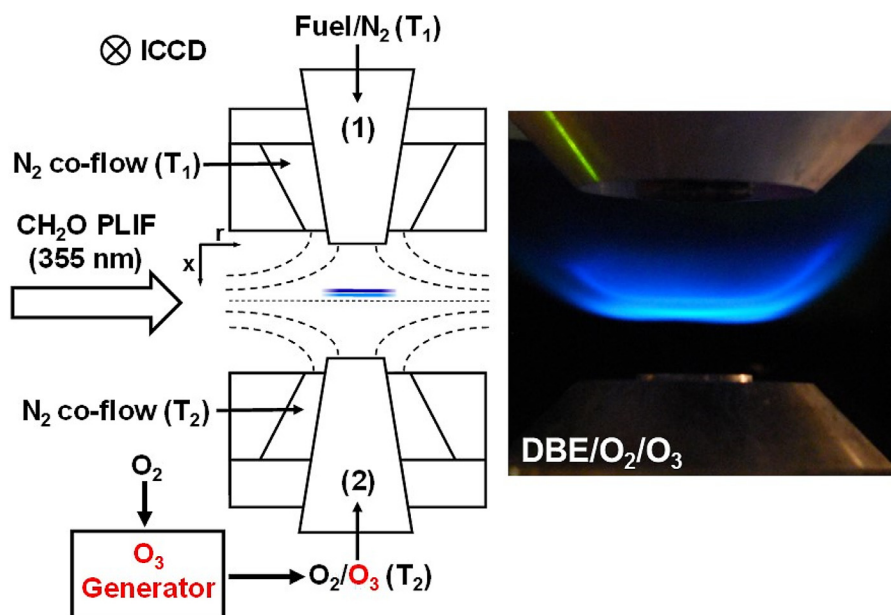


Fig. 1. Experimental apparatus (left) and visual image (right) of a multistage warm diffusion flame of dibutyl ether/oxygen/ozone. The right image was taken using a $f/3.5$ lens aperture with a five-second exposure time.

low-temperature chemistry, there can exist multistage premixed double flames at near-limit conditions. The occurrence of this multistage double flame structure is due to the existence of two different sets of chain-branching reactions involving small radical chemistry and peroxy chemistry at high and low temperatures, respectively.

The existence of burner-stabilized premixed double flames, where a premixed hot flame is preceded by a distinct premixed cool flame, has been experimentally demonstrated separately by Foresti [11] and Agnew et al. [12] in 1955, although much of the chemical origin of such curious character was unknown. Only recently have such flames been studied in sufficient kinetic detail to explain the origin of the chemical character and dynamics of such flames [7–10], owing to the advancements in the understanding of the main low-temperature branching pathways [13]. Further, a third intermediate oxidation stage, between the premixed cool flame and hot flame, has also been observed in confined vertical-tube reactor configurations for premixed reactants [14] and was attributed to the complex reaction pathways of the intermediates produced in the first oxidation stage, although the exact chemical mechanism was not clear. Such configurations have been revisited recently in novel micro flow reactors and have provided a more detailed understanding of the chemistry that governs such behavior in premixtures for different fuels [15–21].

Analogous to the premixed flame, it is reasonable to expect that such chemical complexity could also manifest in non-premixed flames. Recently, significant effort has been allocated towards studying non-premixed cool flames in terrestrial counterflow [3,4,22–26] configurations and in droplets in microgravity conditions [27–38]. However, neither experimental nor computational endeavors have revealed as to whether such multistage flame behavior can exist in diffusion flames of any kind. Specifically, all references to multiple stages of oxidation have been that of consecutive, but not coexistent, modes of non-premixed burning. Such consecutive modes include the known two-stage ignition process through which peroxy chemistry undergoes sufficient branching to trigger radical runaway and transition into second stage oxidation governed by small radical chemistry. Therefore, the possibility of the existence of self-sustaining coexistent low-temperature multistage diffusion flames, and whether their exist-

ence can only be ephemeral, is currently unknown. Indeed, here we demonstrate the existence of such a flame at low temperatures, which we term a low-temperature multistage warm diffusion flame. To the authors' knowledge, such multistage character has not been experimentally observed for diffusion flames, likely owing to the difficulty in establishing such flames during the oxidation of initially unmixed reactants, as will be discussed in this work.

In this work, we report on the experimental evidence of the existence of low-temperature multistage (double) diffusion flames at terrestrial conditions. Such flames were interrogated by means of thermometry, laser-induced fluorescence, and chemiluminescence measurements. We describe the chemical character of such flames and demonstrate that the existence of spatially-distinct oxidation zones is a result of primary partial oxidation of the fuel and secondary consumption of the intermediates produced during the first oxidation stage. We further demonstrate that, for complex fuels of practical interest, multistage oxidation behavior is the dominant mode of low-temperature burning in many cases, as opposed to single-stage oxidation. The results are of relevance to combustion of petroleum fuels and biofuels, particularly in turbulent environments where relatively long residence times can occur.

2. Experimental and computational methods

2.1. Experimental methods

The opposed flow configuration of two impinging gaseous jets was used in order to establish and examine diffusion flames in an atmospheric counterflow burner, as shown in Fig. 1. The details of the experimental setup are described elsewhere [39,40] and will only be briefly reviewed here. The fuel was injected through a vaporization chamber using a steel syringe pump (Harvard PHD 22/2000). Heated nitrogen was concurrently issued to assist vaporization. The vaporized mixture was then ejected from a nozzle (13-mm diameter) to meet an oxidizer jet. A nozzle separation distance of 22.5 mm was maintained throughout this study. The fuel used in this work was dibutyl ether (Sigma Aldrich, anhydrous, 99.3%), for reasons that will be elucidated shortly (Section 3.1). The oxidizer mixtures used were pure oxygen and an ozone-doped oxygen stream. Mixtures of oxygen/ozone were generated using

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