Experimental study of the dynamics and structure of self-sustaining premixed cool flames using a counterflow burner

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ABSTRACT

Self-sustaining premixed cool flames are successfully stabilized in a dimethyl ether/oxygen counterflow burner through ozone addition, creating a new platform for the quantitative measurement of cool flame extinction limits, ignition limits, and structure as well as the validation of low-temperature chemical kinetic models. First, results show that stable premixed cool flames can exist over a broad region of equivalence ratios and strain rates, which allows for the ignition and extinction limits of both cool flames and hot flames to be measured at a variety of conditions. It is seen that at low fuel concentrations the cool flame extinction limit surpasses the hot flame extinction limit, providing experimental validation to previous numerical predictions. Furthermore, the experiments demonstrate that the cool flame speed's dependence on equivalence ratio is far weaker than that of near-limit hot flames. It is also found that a hysteresis exists between cool flames and hot flames near the hot flame extinction limit at low fuel concentrations. The examination of cool flame structure through planar laser-induced fluorescence reveals that the CH_2O profile of the premixed cool flame is much thicker than its hot flame counterpart at the same fuel concentration, confirming the importance of CH_2O as an important cool flame product. Numerical calculations based on a detailed chemical kinetic model are able to capture these various trends and show good qualitative agreement with the experimental results. The present experiments provide a new method to study premixed cool flames in a laboratory setting and to advance the fundamental understanding of low-temperature chemistry and near-limit flame dynamics.

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1. Introduction

As part of the continued push for new internal combustion engines with higher efficiency and lower emissions, technologies such as homogeneous charge compression ignition (HCCI) and reactivity controlled compression ignition (RCCI) have been increasingly studied in recent years [1–5]. For these kinetically-controlled systems, combustion takes place at lean conditions and moderate temperatures, which in many cases are reached through a two-stage ignition process governed by both high-temperature and low-temperature chemistry. While the scientific understanding of conventional hot flames is well established, fundamental knowledge of cool flame dynamics and chemistry is still quite limited.

The first discovery of a cool flame took place centuries ago [6], and since then many studies have provided insight into the characteristics of the mysterious "pale blue flame" [7]. Of particular importance for internal combustion engines were experiments showing the connection between cool flames and engine knocking [8,9]. Recent investigations have even applied the cool flame to reduced gravity [10,11] and microgravity settings [12–15]. Yet despite these studies and others involving cool flames using heated surfaces [16,17], heated reactors [18,19], and stirred reactors [20,21], it has been notably difficult to establish self-sustaining cool flames in order to examine their detailed structure, extinction limits, flammability limits, and other dynamics in a laboratory setting without flame-wall thermal coupling.

Recent numerical simulations [22] demonstrated that a well-defined premixed cool flame can propagate for a broad range of equivalence ratios below the flammability limit of a hot flame. However, there is no direct experimental data to support this finding due to the difficulty in experimentally establishing a self-sustaining cool flame. Although Foster and Pearlman [10,11] measured cool flame speeds in a spherical chamber, the initial mixture temperature was so high that the mixture was partially oxidized in front of the flame, and as a result the flame was not propagating in a well-defined non-reacting mixture.

The main difficulty in sustaining cool flames is intrinsically tied to their slow chemistry at low temperatures and atmospheric pressure. The primary chain-branching pathway in low-temperature oxidation involves the alkylperoxy radical isomerization reaction

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($RO_2 = QOOH$) and subsequent breakdown into an oxygenated radical and two OH radicals [23]. In order to initiate this RO_2 chemistry, the fuel radical (R) must first be obtained from the fuel molecule (RH) by H-abstraction. However, at low temperatures the active radical concentration is quite low, resulting in slow fuel induction chemistry. Moreover, increasing the temperature to promote induction chemistry also tends to increase the rates of reactions such as $H_2O_2 \, (+ \, M) = OH + OH \, (+ \, M)$, which leads to the establishment of a hot flame governed by high-temperature chemistry. Therefore, in order to create a stable cool flame, one must find a way to create a sizable initial radical pool without favoring the reactions associated with high-temperature chemistry.

One solution to the difficulty of cool flame induction is the use of plasma-assisted combustion. While direct plasma discharge [24] will indeed produce a large initial radical pool and a stable cool flame, the complicated plasma-flame kinetic interactions make this setup difficult for detailed study. Upstream plasma discharge into pure oxygen, however, will create stable ozone molecules which can carry the O radicals necessary to ignite a cool flame. This avoids initiating additional reactions in the unburned mixture and thus creates a well-defined boundary condition for cross-validation between experiments and modeling. This method has recently been successful in creating self-sustaining cool diffusion flames [25] for which the extinction limits and flame structures were measured in a counterflow burner.

The objective of this study, therefore, is to create an experimental platform for producing self-sustaining premixed cool flames through ozone addition. An ozone-assisted counterflow burner is utilized to establish cool flames and to allow for the quantitative investigation of cool flame dynamics and structure through the measurement of extinction limits, ignition limits, and species profiles. Numerical simulations are also conducted to understand the experimental results and to assess the predictability of kinetic models for cool flames.

2. Experiment

An ozone-assisted counterflow burner has been chosen as the experimental platform for this study due to its quasi-one-dimensional geometry, lack of wall-flame interactions (essential due to the weak heat release associated with cool flames), well-defined flame strain rates and boundary conditions, and ease of optical access. The burner itself is the same one used in several previous counterflow studies [25–29], with nozzle diameters of 13 mm. A simplified schematic of the setup for this experiment is shown in Fig. 1. The upper burner nozzle issues pure nitrogen, which is heated to 600 K to decompose the ozone in the premixture (supplied by the bottom burner) in order to trigger ignition of the cool flame. The oxidizer stream initially consists of pure (>99.9%) oxygen at 300 K, which passes through a non-equilibrium plasma ozone generator (Ozone Solutions, TG-20). The ozone generator employs a dielectric barrier discharge at 170 kPa, which produces a variety of species including O, O_3, O_2(v), $O(^1D)$, and $O_2(a^1\Delta_g)$ [30]. Of these, however, only O_3 is stable at 300 K, and none of the other species produced has a lifetime longer than 20 ms. As a result, by the time the oxidizer stream encounters the fuel stream, only O_2 and O_3 are present. After the fuel stream is mixed in, the $O_2/O_3/DME$ premixture is finally directed to the bottom burner exit. In this experiment, DME is chosen over other fuels for several reasons. It is gaseous at room temperature (heating the premixture would prematurely decompose the ozone), possesses fairly strong low-temperature chemistry, already has several detailed kinetic models of its oxidation, and its flame speeds at 1 atm have been well studied [31,32].

The ozone concentration in the oxidizer stream was measured by an absorption spectroscopy method similar to the ones used
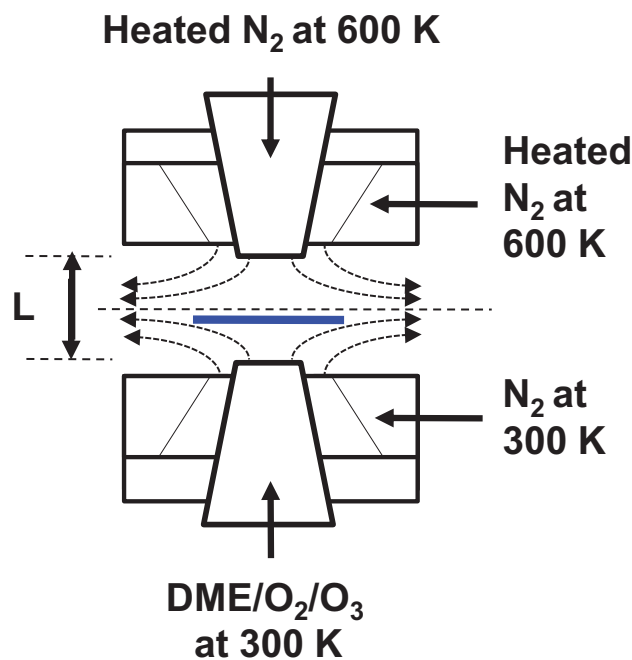

Fig. 1. A simplified schematic of the experiment's counterflow burner in the premixed configuration. L represents the distance between the upper and lower burners (2.25 cm).

in [25] and [30] and showed a dependence on the initial oxygen flow rate. A spectrometer (Ocean Optics, USB2000+) was placed at the exit of the ozone generator to capture a range of wavelengths (280–310 nm) emitted by a Deuterium lamp (Oriel). The absorption spectra with the generator on and off were then compared to quantify the ozone concentration. Measurements were also performed at the burner exit to confirm that no ozone was lost as the premixture moved through the burner. For the range of oxygen flow rates in this study, the ozone concentration in the oxidizer remained between 3.0% and 3.4%.

For all experiments in this study, the burner separation distance is set to 2.25 cm. The strain rate (a) is defined as the density-weighted gradient of the axial flow velocities [26–28,33], specifically,

$$a = 2\left(\frac{U_{lower}}{L}\right)\left(1 + \frac{U_{upper}\sqrt{\rho_{upper}}}{U_{lower}\sqrt{\rho_{lower}}}\right),$$

where U_{upper} and U_{lower} are the velocities at the upper and lower burner exits, respectively, ρ_{upper} and ρ_{lower} are the mixture densities at the same exits, and L is the burner separation distance. Additionally, the equivalence ratio (ϕ) is defined as the ratio of the total moles of fuel (DME) to total moles of oxidizer ($O_2 + O_3$) divided by the stoichiometric ratio. Mass flow rates of N_2, O_2, and DME are controlled through sonic nozzles calibrated to pressure gauges (Cecomp Electronics). All measurements are taken at atmospheric pressure (1 atm).

In order to visualize the premixed cool flame structure, planar laser-induced fluorescence (PLIF) of CH_2O molecules is performed [34]. The third harmonic (355 nm) of an Nd:YAG laser (Quantel, Q-smart 850) excites the CH_2O at an energy of approximately 200 mJ/pulse. Two band filters, passing frequencies between 400 and 450 nm, are positioned in front of an ICCD camera (Princeton Instruments, PI-MAX 4) to isolate the CH_2O fluorescence. The laser beam is expanded into a 200-μm-thick vertical sheet and positioned at the centerline of the burner.

For comparison with the experimental results, the numerical calculations in this paper are carried out using a modified PREMIX