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Proceedings of the Combustion Institute 35 (2015) 881–888

**Proceedings
of the
Combustion
Institute**

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Self-sustaining *n*-heptane cool diffusion flames activated by ozone

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Available online 21 June 2014

Abstract

A novel method to establish self-sustaining cool diffusion flames with well-defined boundary conditions is experimentally demonstrated by adding ozone to the oxidizer stream in counterflow configuration. It is found that the atomic oxygen produced through the decomposition of ozone dramatically shortens the induction timescale of the low temperature chemistry, extending the flammable region of cool flames. Thus, it enables the establishment of self-sustaining cool flames at the pressure and timescales at which normal cool flames may not be observable. The present method, for the first time, provides an opportunity to study cool flame dynamics, structure, and chemistry simultaneously in well-known flame geometry. Extinction limits of *n*-heptane/oxygen cool diffusion flames are measured and a cool diffusion flame diagram is experimentally determined. Numerical simulations reveal that the extinction limits of cool diffusion flames are strongly governed by species transport and low temperature chemistry activated by ozone decomposition. The structure of cool diffusion flame is further investigated by measuring the temperature and species distributions with a micro-probe sampling technique. The kinetic model over-predicts the rate of *n*-heptane oxidation, the heat release rate, and the flame temperature. Measurements of intermediate species, such as CH₂O, acetaldehyde, C₂H₄, and CH₄, suggest that the model over-predicts the QOOH thermal decomposition reactions to form olefins, resulting in substantial over-estimation of C₂H₄, and CH₄ concentrations. The new method and data of the present study will contribute to promote understandings of cool flame chemistry.

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Keywords: Cool flame; Low temperature chemistry; Extinction; Counterflow diffusion flame; Ozone

1. Introduction

Cool flames were observed accidentally nearly two centuries ago [1,2] and have been studied by the combustion community ever since. The legacy of these initial studies today appears extensively in fuel chemistry literature, in which low temperature

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hydrocarbon chemistry is regarded as the key process for engine knock [3,4]. Lignola and Reverchon [5] summarized three experimental approaches to study cool flames; heated burners, heated flow reactors, and jet-stirred reactors. Despite of the ambiguous boundary conditions in a heated burner, the convenience of optical accessibility facilitated emission spectroscopy measurements, identifying excited formaldehyde as the source of the pale bluish chemiluminescence from cool flames [5]. Flow and jet-stirred reactor experiments provided detailed information on the cool flame relevant chemistry of hydrocarbon (primarily liquid fuels) at temperatures representative of cool flames (500–1000 K) [6–10]. With the development of appropriate fast sampling methods, rapid compression machines have granted the access to the transient cool flame chemistry [11]. Generally, the experiments discussed above rely on external heating and wall heat losses to achieve cool flames, rendering complicated thermal and chemistry coupling with the wall. As a result, detailed and fundamental understandings of cool flame behaviors have not been well established.

Moreover, all of the previous studies were limited to homogeneous fuel/oxidizer pre-mixtures. As a consequence, it is unclear whether cool flame is only limited to premixed systems or not. Interestingly, a recent zero-gravity experiment of droplet combustion [12] has shown that a cool flame might be established even in a diffusive system, hypothesizing the existence of cool diffusion flame after radiation-controlled extinction, later confirmed by numerical simulations [13]. Although the numerical simulation captured the global trend of droplet flame extinction and the subsequent formation of cool diffusion flame, the detailed structure of cool diffusion flame has not been revealed yet. As such, cool flame dynamics remain mysterious and the fidelity of cool flame chemistry in diffusive conditions remains uncertain.

One of the main challenges to stabilize self-sustaining cool flames is that the slow cool flame induction chemistry at low temperatures prevents radical branching, therefore the self-initiation, while on the other hand, the fast high-temperature chain branching reactions populate the radical pool rigorously, thus initiating a hot flame directly [14].

Recent progresses in plasma-assisted combustion in counterflow flames [15,16] provide a hint to accelerate the cool flame induction chemistry at the low temperature. By producing active radicals such as atomic oxygen, an in situ plasma discharge, i.e. generated between the two nozzles of a counterflow burner, stabilizes diffusion flames at flame temperature below 1000 K by modifying the traditional ignition/extinction S-curve. Unfortunately, direct plasma discharge affects and

complicates the combustion chemistry. In order to isolate the plasma-flame coupling, ozone has been frequently used as an atomic oxygen carrier to enhance flame stabilization and ignition [17,18]. Similarly, ozone is also utilized to reduce the time scale of induction chemistry and activate the low temperature chemistry in tubular [19] and jet-stirred reactors [20]. Those studies imply that a self-sustaining cool flame might be observable with ozone addition. As the recent advanced concepts of engine design [21–23] appear to heavily rely on the low temperature combustion, it is pre-requisite to understand the underlying chemical kinetics and flame dynamics at the regime of low temperature chemistry. Consequently, the fundamental challenge dwells in the development of an experimental platform with well-defined chemical and flow boundary conditions, so that both the global properties and the detailed chemical kinetics of cool flames can be simultaneously investigated.

In this regard, the goal of this study is to develop a novel method to establish a self-sustaining cool diffusion flame in a counterflow burner with well-defined boundary conditions for kinetic and dynamic studies of cool flames. In the following, an experimental methodology to establish n-heptane cool diffusion flames by ozone addition in the oxidizer stream is demonstrated at first. Extinction limits of cool diffusion flames are further measured in a function of strain rate. Detailed flame structure (species distributions) of cool diffusion flame is examined by using a micro sampling method with micro gas chromatograph (GC) and compared to numerical modeling results. Finally, the fidelity of chemical kinetic model to describe n-heptane cool diffusion flames is discussed.

2. Experiment

The experimental platform to establish a self-sustaining cool diffusion flame in this study consists of a heated counterflow burner integrated with a liquid fuel vaporization system and an ozone generator with micro plasma dielectric barrier discharge. Schematic of experimental setup is shown in Fig. 1. The details of counterflow burner have been described elsewhere [24–27], thus only briefly described here.

Liquid fuel, n-heptane (Sigma Aldrich, >99% in purity) is delivered by a syringe pump (Harvard Apparatus, PHD 22/2000) through the central capillary tube (200 μm inner diameter) into a pre-vaporization chamber maintained at 550 K. Heated nitrogen is supplied through the annular coaxial tube (2 mm inner diameter) to enhance the atomization and thus the vaporization of the liquid fuel. The fully mixed and pre-vaporized fuel/nitrogen mixture is then directed into the

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