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Multistage oscillatory "*Cool Flame*" behavior for isolated alkane droplet combustion in elevated pressure microgravity condition

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Abstract

Recently, large diameter, isolated *n*-heptane droplet experiments under microgravity conditions (aboard the International Space Station) exhibited "*Cool Flame*" burning behavior, resulting from a heat loss mechanism that extinguishes hot combustion and a transition into a sustained, low temperature second stage combustion. In atmospheric pressure air, a single combustion mode transition to "*Cool Flame*" burning is followed by diffusive extinction. But with increasing pressure, *multiple cycles* of hot initiation followed by transition to "*Cool Flame*" burning are observed. This paper reports experimental observations that characterize the transition time histories of this multi-cycle, multi-stage behavior. Transient spherosymmetric droplet combustion modeling that considers multi-stage detailed kinetics, multi-component diffusion, and spectral radiation is applied to analyze the experimental observations. The simulations indicate that as parameters change the chemical time scales dictating low temperature degenerate chain branching, multiple hot/cool flame burning transitions are induced by increasing the cool flame burning heat generation rate compared to the diffusive loss rate. The balance of these terms in the negative temperature coefficient kinetic regime defines whether reactions accelerate into re-ignition of a hot flame event, burn quasi-steadily in the cool flame mode, or diffusively extinguish. The rate of reactions controlling ketohydroperoxide formation and destruction are shown to be key re-ignition of hot combustion from the cool flame mode. Predictions are found to be in good agreement with the experimental measurements. Modeling is further applied to determine how these observations are dependent on initial experimental conditions, including pressure, and diluent species.

Keywords: Cool Flame; Droplet combustion; Microgravity; Radiation; FLEX

1. Introduction

Since their discovery, "*Cool Flames*" have intrigued researchers over the years. Classically, the term "*Cool Flame*" is associated with premixed systems in which a transient

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propagating wave proceeds through a premixed fuel/air mixture with one or more repetitive occurrences leading to high temperature burning of the remaining reactants. Quasi-steady, cool flame phenomena have also been produced in flowing premixed reactants, typically followed by a second stage hot flame. Bradley and coworkers [1] studied cool flame and second-stage ignition phenomena in a quasi-steady configuration by using a vertical flow reactor arrangement and the oxidations of acetaldehyde or propionaldehyde. Fairlie et al. [2,3] studied transient "*Cool Flames*" and the associated repetitive phenomena using premixed propane–oxygen static reactor experiments in microgravity. Their study involved both experimental and numerical aspects. The onset, spatial growth and stability of the "*Cool Flame*" was attributed to the interactions among the elementary reactions and rates governing negative temperature coefficient (NTC) behavior and the temperature field controlled by thermal diffusion to the reactor wall. In their pioneering work, Tanabe et al. [4] reported the two-stage ignition behavior of *n*-heptane droplets. Time dependent temperature field around the igniting droplets were observed by interferometry to determine the two step temperature rise. Moriue et al. [5] utilized the tethered droplet configuration to experimentally measure the ignition delays of "*Cool Flame*" and hot flame appearance for single *n*-decane, *n*-dodecane, *n*-tetradecane and *n*-hexadecane droplets in a multiphase system. The flames were produced by inserting the droplet in a temperature controlled furnace and observing flame evolution using Michelson interferometry. Cuoci et al. [6] simulated Moriue's *n*-decane experiments with a detailed hydrocarbon oxidation mechanism. Their model was able to predict the multi-stage ignition behavior.

Recently, long duration, "*Cool Flame*" burning for large diameter, isolated droplet combustion was first observed for atmospheric pressure, pure *n*-heptane droplets experiments in air aboard the International Space Station (ISS) [7,8]. In these non premixed experiments, large *n*-heptane droplet burning exhibited dual modes of combustion and extinction. In the first stage following ignition, the droplet underwent classical high temperature burning with a visible flame surrounding the droplet. The transient, high temperature burning was observed to extinguish radiatively, followed by a continuing lengthy period of quasi-steady surface regression of the droplet without any visible flame. This lengthy "*second stage*" phenomenon was observed to extinguish, leaving behind a smaller droplet, which experienced a short period of time-dependent evaporation.

Detailed numerical analysis of these experiments [8] indicates that the "*second stage*" behavior results from the existence of multistage kinetic regimes that are well known in terms of two stage autoignition phenomena [9–11] and the premixed "*Cool Flame*" phenomena with large carbon number normal alkanes [12]. In the "second stage", heat generation as a result of negative temperature coefficient, degeneratively branched, oxidation chemistry was shown to be dynamically balanced by diffusive heat loss, leading to the lengthy period of "flameless" quasi-steady droplet regression, concluded by diffusive extinction. Though the phenomena are different in principle from those governing classical premixed cool flames, the low temperature droplet burning behavior has been generally conotated as "*cool flame*": droplet burning. It is not surprising that similar observations would occur with even larger carbon number normal alkanes such as *n*-octane, *n*-decane, etc., all of which have more rapid negative temperature coefficient kinetic rates than *n*-heptane [13]. More recent ISS *n*-heptane isolated droplet combustion experiments at three atmospheres pressure and the same oxygen index but with diluents inhibiting heat loss shows "multiple cycles" of dual stage combustion i.e. *hot–cool* flame transitions. This paper presents experimental examples of this multistage, multiple cycle mode of isolated *n*-heptane droplets under high pressure microgravity conditions. The experimental observations are then compared against *apriori* detailed numerical predictions using a novel spherosymmetric droplet combustion model that includes full multi-stage detailed chemical kinetics, multi-component transport, and spectral radiative interactions. Predictions from the model are found to compare favorably with the experimentally measured droplet and flame diameter evolution histories, and diffusive extinction results. Further modeling calculations are analyzed to elucidate the effects of different ambient conditions; i.e. pressure and diluent concentration.

2. Experimental setup and procedure

The experiments are conducted under the **FL**ame **EX**tinguishment (FLEX) program onboard the International Space Station (ISS). The Multi-User Droplet Combustion Apparatus (MDCA) installed in the Combustion Integrated Rack (CIR) facility was employed. The MDCA uses an opposed needle deployment technique [14] to deploy droplets of chosen diameter on an 80 μm single filament silicon carbide (SiC) fiber in these microgravity experiments (freely floating droplet experiments can also be performed). The tethering fiber was necessary for these tests in elevated pressure in order to eliminate the relative velocity between the gas phase and the droplet upon deployment. Ignition of the tethered droplet was accomplished using two symmetrically positioned Kanthal hot wire igniters. The experimental diagnostic system consisted of a black-white cam-