Cool Flames in Propane-Oxygen Premixtures at Low and Intermediate Temperatures at Reduced-Gravity

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INTRODUCTION

The Cool Flame Experiment aims to address the role of diffusive transport on the structure and the stability of gas-phase, non-isothermal, hydrocarbon oxidation reactions, cool flames and auto-ignition fronts in an unstirred, static reactor. These reactions cannot be studied on Earth where natural convection due to self-heating during the course of slow reaction dominates diffusive transport and produces spatio-temporal variations in the thermal and thus species concentration profiles (Griffiths, et al., 1971; Melvin, 1969). On Earth, reactions with associated Rayleigh numbers (Ra) less than the critical Ra for onset of convection (Ra_{cr}~600; Tyler, 1966; Fine, et al., 1970; Barnard and Harwood, 1974) cannot be achieved in laboratory-scale vessels for conditions representative of nearly all low-temperature reactions. In fact, the Ra at 1g ranges from 10^4 - 10^5 (or larger), while at reduced-gravity, these values can be reduced two to six orders of magnitude (below Ra_{cr}), depending on the reduced-gravity test facility.

Currently, laboratory (1g) and NASA's KC-135 reduced-gravity (μ g) aircraft studies are being conducted in parallel with the development of a detailed chemical kinetic model that includes thermal and species diffusion. Select experiments have also been conducted at partial gravity (Martian, 0.3g_{earth}) aboard the KC-135 aircraft. This paper discusses these preliminary results for propane-oxygen premixtures in the low to intermediate temperature range (310-350°C) at reduced-gravity.

GROUND-BASED COOL FLAME AND AUTO-IGNITION STUDIES

Laboratory Studies in a Mallard-LeChatlier Static, Unstirred Reactor: A classic Mallard-LeChatlier apparatus (Mallard, 1880) was developed and used to conduct 1g and reduced-gravity experiments. A schematic of the hardware is shown in figure 1. It consists of a furnace, a fused-silica spherical vessel of given diameter (i.d.=10.2cm) and a gas delivery system. The furnace employs resistive heating elements in the rear and top panels, an internal mixing fan to circulate the hot air, three parallel 7.5cm diameter (0.32cm thick, 1.2cm separation distance) quartz windows on both the top and side walls of the furnace for viewing and a gas feedthrough built into the door. The temperature uniformity within the furnace is $\pm 10^{\circ}$ C throughout its operating range (20-600°C), measured at random spatial locations using 0.51mm diameter wire, unsheathed, type-K thermocouples. The vessel pressure is recorded at 100Hz using a Setra 0-25psia Model 204 transducer (accuracy: ± 0.028 psia= ± 1.4 Torr) situated at the vessel inlet, on the cold side of the oven door. Two intensified, ultraviolet and visible-sensitive cameras operating at maximum gain (~10⁻⁶ fc minimum sensitivity) and full exposure, record the integrated light intensity at 30 frames/s.

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Fig. 1: Schematic of static, unstirred reactor, furnace and gas delivery system

Representative experimental results obtained at 1g for different initial pressures at two different vessel temperatures (a: $T=310^{\circ}C$, b: $T=320^{\circ}C$) are shown in figures 2a and 2b, respectively. At low pressures, steady, dark (non-visible) reaction is observed with induction periods on the order of minutes. As the pressure increases at fixed temperature, multiple cool flames occur. Further pressure increase results in a decrease in the first induction period. At sufficiently high initial pressures, two-stage ignition (a cool flame preceding a hot ignition) occurs. These and similar results at different temperatures (in the range of 300-400°C) are being used to compile detailed ignition diagrams.



Fig. 2: Pressure histories at 1g at (a) T=310°C, (b) T=320°C for five experiments at different initial pressures. Mixture composition is C₃H₈:O₂ (1:1).

<u>KC-135 Aircraft Reduced-Gravity and Martian-Gravity Results</u>: Representative cool flame results obtained during KC-135 parabolic maneuvers at reduced-gravity using propane-oxygen mixtures at T=320°C are shown in figures 3a and 3b for pressures less than (and equal) to 7.7psia. At 7.7psia, one cool flame is observed with an induction period of 15s, followed by a steady, non-visible reaction that continues throughout the remainder of the available test time. Beyond 20s, the g-level is time-dependent and varies from 10^{-2} g to 1.8g (see lower curve in fig. 3b). This transitional period induces buoyant mixing in the vessel and a complicated pressure history. At lower initial pressures, experiments conducted at 3.9psia and 6.2psia show little evidence of reaction during the available reduced-gravity test time, while reaction in the subsequent "pull-up"/transitional g period following the reduced-gravity period is observed. Figure 3b shows a plot of the pressure history associated with the experiment conducted at 3.9psia along with the vertical-component of the acceleration. These results clearly demonstrate the need for additional reduced-gravity test time.



Fig. 3: Cool flames in C₃H₈:O₂ (1:1) mixture at different initial pressure at T=320°C and μg; Reaction continues beyond the available reduced-gravity test time.

As mentioned, select experiments were also conducted at $0.3g_{earth}$ (Martian-g) to study the effect of "weak" convection/mixing without changing any physical parameters or the mixture composition (i.e., the Ra can be changed by varying g). The pressure histories obtained at 1g and µg and those obtained at 0.3g and 1g are plotted in figures 4a and 4b for nearly identical initial pressures (ranged from 7.5 to 7.8pia), fixed temperature (320°C) and fixed mixture composition.

At 1g, five sequential cool flames are observed. The first cool flame follows an induction period of approximately 27s. At 0.3g, two sequential cool flames are visually observed and the first induction period shortens to 15s. In contrast, only a single cool flame is observed at μ g followed by a monotonic pressure increase until the end of the available 20s test time at which time the pressure gradient remains positive. The vertical-components of acceleration associated with these three tests are also shown in the lower half of figures 4a and 4b.



Fig. 4: (a) Comparison of pressure histories obtained at reduced-gravity (KC-135 aircraft) and 1g and (b) Martian gravity versus 1g for nearly identical initial pressures at T=320°C. The 1g data is the same in both (a) and (b). Mixture composition is C₃H₈:O₂ (1:1). The vertical component of acceleration is shown in the lower portion of the plot (negative value indicates downward acceleration).

<u>Imaging Cool Flames in Propane-Oxygen and Propane-Air Premixtures:</u> For the propaneoxygen and propane-air experiments reported herein, the overall intensity of the integrated ultraviolet and visible emission (integrated over the spectral sensitivity of the intensified camera) is significantly lower than that observed in earlier experiments with butane-oxygen reported by Pearlman (1999). At maximum camera gain (10^{-6} fc), very few propane-oxygen and only one propane-air cool flame(s) have been imaged at reduced-gravity. Some of the clearest images obtained from the KC-135 testing at a: μ g, b: 0.3g, and c: 1g in the propane-oxygen system are shown in figure 5. In all sequences shown, the temperature is fixed at 320°C.



Fig. 5: Representative cool flame images in C₃H₈:O₂ at 320°C, 10.2cm i.d. quartz vessel at (a) μg ~10⁻²g (12.0 psia), (b) 0.3g (7.6psia), and (c) 1g (7.8psia). Time lapse between sequential images in 1/3s. Images have been enhanced for clarity.

CONCLUSIONS

Preliminary experimental results have been obtained for a propane:oxygen system at 1g, μ g, and 0.3g. Multiple cool flames have been observed at 1g and 0.3g, yet only a single cool flame has been observed at μ g within the available test time. Reaction is observed to continue beyond the 20s available on the KC-135 aircraft. Qualitatively, the frequency of oscillation is observed to decrease as g-level decreases (second and subsequent induction periods decrease with a decrease in g-level). Cool flames at 0.3g appear qualitatively similar to those at 1g, yet those at μ g are radially (presumably spherically) symmetric. Ignition diagrams will be developed using this and subsequent reduced-gravity data. Additional work is needed to quantify the role of g-jitter (gravitational fluctuations) on the spatio-temporal structure and evolution of reduced-gravity cool flames. Numerical models will be tuned based on these results and used to predict the behavior of cool flames beyond the available 20s in ground-based facilities. These results will guide the planned space-flight experiment.

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