Droplet Combustion Experiment (DCE)

Principal Investigator:

Dr. Forman A. Williams University of California, San Diego La Jolla, California

DROPLET COMBUSTION EXPERIMENT

John B. Haggard, Jr. and Vedha Nayagan NASA Lewis Research Center 21000 Brookpark Road Cleveland, OH 44135

Frederick L. Dryer Department of Mechanical and Aerospace Engineering Princeton University Princeton, NJ 08544

Forman A. Williams Center for Energy and Combustion Research University of California, San Diego La Jolla, CA 92093

Abstract

The first space-based experiments were performed on the combustion of free, individual liquid fuel droplets in oxidizing atmospheres. The fuel was heptane, with initial droplet diameters ranging about from 1 mm to 4 mm. The atmospheres were mixtures of helium and oxygen, at pressures of 1.00, 0.50 and 0.25 bar, with oxygen mole fractions between 20% and 40%, as well as normal Spacelab cabin air. The temperatures of the atmospheres and of the initial liquid fuel were nominally 300 K. A total of 44 droplets were burned successfully on the two flights, 8 on the shortened STS-83 mission and 36 on STS-94. The results spanned the full range of heptane droplet combustion behavior, from radiative flame extinction at larger droplet diameters in the more dilute atmospheres to diffusive extinction in the less dilute atmospheres, with the droplet disappearing prior to flame extinction at the highest oxygen concentrations. Quasisteady histories of droplet diameters were observed along with unsteady histories of flame diameters. New and detailed information was obtained on burning rates, flame characteristics and soot behavior. The results have motivated new computational and theoretical investigations of droplet combustion, improving knowledge of the chemical kinetics, fluid mechanics and heat and mass transfer processes involved in burning liquid fuels.

Introduction

The general objectives of the droplet combustion experiment (DCE) are to improve understanding of the mechanisms of burning of liquid fuel droplets. These objectives are important for both scientific and practical reasons. Scientifically, the conservation equations of combustion, especially in the presence of phase changes, are too complex to be solved in arbitrary configurations and contain many detailed chemical-kinetic, transport and radiative parameters having uncertain values. These equations, however, now can be solved in the one-dimensional, spherically symmetrical, time-dependent situations established in DCE, so that comparisons of predictions with experimental results can be made to improve knowledge of uncertain parameters that control combustion mechanisms. Practically, fossil fuels provide 85% of our energy needs and liquid fuels more than 95% of energy usage in the transportation sector. Improvements in methods of combustion gained from knowledge generated in DCE studies can help to conserve these natural resources, reduce rates of emission of greenhouse gases and decrease air pollution associated with combustion processes. An additional practical motivation for DCE lies in the realm of fire and explosion safety of liquid fuels, which can be quite different in microgravity compared with normal gravity and which could be improved by knowledge obtained in such experiments.

The DCE fuel, n-heptane, is a representative liquid hydrocarbon constituent of transportation fuels, a higher normal alkane that has been studied most extensively in earlier combustion experiments, including ground-based experiments on droplet burning. It was selected for DCE because this extensive earlier data base facilitates the comparisons with ground results and the development of new, firmly based, fundamental knowledge; the aforementioned uncertainties in chemical-kinetic, transport and radiative parameters all apply to heptane, so that there are things to be learned.

The first microgravity droplet combustion experiment, performed by Kumagai¹, in fact employed heptane. His apparatus provided about 1 second of microgravity, enabling him to burn to completion in air droplets of initial diameters slightly less than 1 mm. Since the burning time increases as the square of the initial diameter in these diffusion-controlled processes, the largest ground facility currently available, the 10-second drop shaft in Hokkaido, can be used to study the entire history of heptane droplets burning in air only up to initial droplet diameters of about 3 mm. Maintaining good spherical symmetry in droplet combustion requires the gravity levels, below 10⁻⁴ to 10⁻⁶ earth gravity, obtainable in drop towers and in space; experiments in aircraft flying parabolic trajectories, which in principle could provide longer test times, generate degraded data as a consequence of their higher gravity levels. The only other alternative to space-based experiments, namely the use of sounding rockets, could provide the longer test times at excellent gravity levels but has not been implemented for droplet combustion because of the associated time and expense. Experiments on droplets larger than 3 mm in initial diameter are desirable because of the additional physical phenomena, such as strong influences of radiative energy loss, that occur at these sizes.

Most previous droplet-combustion experiments had air at 1 bar as the atmosphere. Some DCE data points were obtained in air at 1 bar, facilitating comparison with earlier results. Most DCE data, however, employed helium-oxygen atmospheres, designed to facilitate measurements by increasing burning rates and diffusive extinction diameters while reducing soot formation. The use of helium rather than nitrogen as the inert adds a new dimension, enabling effects of different Lewis numbers to be explored more thoroughly.

The DCE experiment was developed over a period of many years. The first space-based droplet-combustion experiment, flown on STS-73, was fiber-supported droplet combustion (FSDC), a glovebox experiment that employed fibers to tether the droplet in spacelab cabin air within the field of the measurement system in the glovebox. That experiment provided data for a number of different fuels, not including heptane, in the same general initial droplet size range as the DCE experiments. On STS-94, FSDC-2 was tested, this time including heptane among the fuels for which data were acquired. To help in comparison with FSDC results, two DCE tests were performed with tethered droplets in cabin air. The intent was to ascertain in detail the influences of the fiber on the combustion by comparing the tethered results with those for free droplets. Since tethered experiments are easier to perform, sufficient fidelity of results with fiber support can reduce the amount of testing needed with free droplets, saving time and expense.

<u>Measurements</u>

Besides ordinary measurements of combustion-chamber pressures and temperatures, camcorder video images of the overall view of each droplet combustion sequence were downlinked. The main data, however, were obtained from magnified backlit images of the droplet recorded on 35 mm motion-picture film and from ultraviolet (UV) images of the flame recorded by a CCD camera equipped with a narrow-band interference filter centered at 310 mm, one of the bands of OH emission characteristic of the flame. The droplet and flame views were selected to be orthogonal to provide three-dimensional information. The UV images were downlinked, but the main data reduction awaited return of the motion-picture film. The combustion chamber contained a crew-view port through which 35 mm still color photographs were taken of some of the tests.

Figure 1 shows a typical backlit sequence of the experiment. In this figure, a droplet of 4 mm initial diameter is burned in an oxygen-helium atmosphere of 35% oxygen at 1 bar. The droplet is formed by injecting the liquid fuel through a pair of opposed and slightly retracting needles to form a bridge. The first frame shows the droplet centered between the needle tips and two opposed hot-wire igniters in place on opposite sides of the droplet. In the second frame, the droplet has been stretched just prior to deployment; the stretching results in more symmetrical deployment with smaller droplet drift velocities. The drift velocities in the present experiments typically were on the order of 1mm/s, about one order of magnitude lower than those achieved in earlier experiments. This small level of motion is attained by rapid extraction of the needles after stretching, between the second and third frames. Needle extraction induces droplet oscillation in the first spherical harmonic mode which, however, viscously damps to immeasurable amplitude, always in less than 1s. The third frame shows the droplet oscillation at maximum amplitude.

After droplet deployment, the hot-wire igniters are activated to initiate combustion. The fourth frame shows the beginning of combustion and demonstrates that there is buildup of soot, the dark speckled elliptical region around the sphere, during ignition. After ignition, the igniters are withdrawn slowly to minimize associated gas flow currents. The last two frames are illustrative of the free-droplet combustion process that continues after igniter withdrawal. Initially the sooting intensifies, and some agglomerated soot escapes as chains in the directions of needle withdrawal, seen in the penultimate frame. As combustion proceeds, the region of high soot concentration becomes more spherical, and agglomeration leads to larger soot particles, the largest of which escape in random directions through the flame, as seen in the final frame. The field of view in these photographs is smaller than the flame diameter so that the flame, a sphere concentric with the droplet, cannot be seen. The condition of the test shown here, one of the largest droplets at the highest pressure and highest oxygen concentration at that pressure, was selected to maximize the visible details of sooting behavior; sooting is less intense for smaller droplets, lower pressures or lower oxygen concentrations.

Figure 2 shows two typical UV flame-image sequences as downlinked. In this figure, the droplets burn in an oxygen-helium atmosphere of 30% oxygen at 1 bar. The frames shown are at 2s intervals, beginning at ignition. The energized hot-wire loops are visible in each of the initial frames (a). In each sequence, the droplet burns for about 10s, exhibiting remarkably spherical flames. In Fig. 2A, the initial droplet diameter d, was 3 mm and the maximum flame diameter, frame (c), about 21 mm. The droplets are not visible at all in these UV images. Figure 2A illustrates diffusive extinction in that the flame initially expands then contracts increasingly rapidly. extinguishing at a small diameter, just after the last frame (f), at which time the droplet has just reached zero diameter and disappeared. In Fig. 2B, the droplet initially was 4 mm in diameter, and the maximum flame diameter, frame (d), was about 25 mm; the magnification is slightly different in the two sequences, as can be seen by comparing the sizes of the igniter wires. Figure 2B illustrates radiative extinction in that the flame can be seen to be extinguishing in the last frame (f), while the flame diameter is still near its maximum value, and at this time the droplet diameter is found from the backlit view to still be roughly 40% of its initial value. In this atmosphere, therefore, initially small droplets extinguish diffusively and large ones radiatively, as discussed more thoroughly in the following section.

The backlit and UV images are digitized and analyzed by a computer-based image-analysis system that identifies edge locations on the basis of gradations in intensity. Diameters are obtained from the area of a circle having the area measured by the image-analysis system. Results for droplet diameters were checked by visual measurements of some film frames and by direct measurement of two orthogonal diameters with the image-analysis system, indicating accuracies better than 5%. Flame images are quite round with well-defined edges marking the outer boundary of the very thin, hottest reaction zone, as seen in Fig. 2. Time histories of droplet and flame diameters are obtained from these results.

Results and Comparisons

Figure 3 shows a representative set of results from STS-83. This figure pertains to droplets burning in oxygen-helium mixtures having 35% oxygen at 1 bar. Many details of the results shown in this figure are given in a publication,² which also makes extensive reference to earlier ground-based results. These earlier results all pertain to initial droplet diameters d_o less than 2 mm. The space-based experiments thus extend the ground-based results to diameters larger by more than a factor of 2. The first observation to be made from Fig. 2 is the nearly linear decrease in droplet diameter squared (d^2) with time (t) after the initial stage. This decrease extends essentially to zero droplet diameter, that is, the droplet essentially disappears prior to extinction in this atmosphere. The same is inferred to be true² for $d_0 = 4$ mm, although that is not seen in the figure because the droplet passes out of the backlit view before completion of combustion; the flame remained in view for the entire combustion history in this test and exhibited an excursion at time t=6s, apparently caused by ignition of a large soot agglomerate passing through the flame. The 15s duration of the flame in this test is well beyond measurement capabilities of any groundbased experiments. The flame-diameter histories in Fig. 3 clearly do not obey the d² law, that is, the curves are not linear. This indicates that the flames are in the outer transient region rather than the inner quasisteady region.² The flames exhibit measurable nonzero (but small) diameters at extinction. Relevant chemical-kinetic information about combustion may be obtained from these flame diameters at extinction. Diffusive extinction occurs in these high-oxygen atmospheres; it is diffusive energy loss, rather than radiative energy loss, that is responsible for extinction.²

Figure 4, also from STS-83, pertains to the same conditions as Fig. 3, except that the oxygen mole fraction was 25%; again the 4 mm droplet passes out of the backlit view prior to extinction, although this did not occur for the 4 mm droplet in 30% oxygen² (not shown here). The droplets in Fig. 4 again exhibit a regime of d²-law burning, while the flames do not, as expected.² The main difference between Figs. 3 and 4 is that in Fig. 4 the flames extinguish at their maximum diameters or slightly thereafter. The extinction in these cases, at the end of the flame-diameter traces, is caused by radiative rather than diffusive energy loss.² After flame extinction, the droplets are seen in Fig. 4 to vaporize more slowly in the residual hot gas; the d² slope decreases. Both flame and droplet diameters at extinction seen in these tests can be employed to extract chemical-kinetic information about the combustion process. Radiative extinctions of this kind have not previously been observed for heptane droplets in ground-based (or any other) experiments.

Tests from STS-83 in 30% oxygen exhibited radiative extinction for $d_0=4mm$ but diffusive extinction for $d_0=3mm$, as was shown in Fig. 2. This indicates that this intermediate dilution is close to the boundary between diffusive and radiative extinction and that larger droplets have a greater tendency towards radiative extinction. Modeling calculations, still continuing, verify this general behavior. Figure 5 shows comparisons of numerical modeling results with experimental results for tests at 1 bar in 30% oxygen, for the time dependence of the square of the droplet diameter.³ The computations were performed for 4 different initial diameters. It is seen that the modeling predicts diffusive extinction with zero droplet diameter at extinction for $d_0>4mm$. The two experimental data runs from STS-83 lie between these computational conditions and exhibit intermediate behavior consistent with the calculations. The slopes of the curves, that is, the burning rates, also are in reasonable agreement with the predictions.

Figure 6 provides a more detailed comparison between predicted and measured burning rates under various conditions at 1 bar, for both space-based and ground-based experiments.³ It is seen from this figure that the burning-rate constant K in the formula

$$d^2 = d_o^2 - Kt \tag{1}$$



Figure 1 Representative sequence of events in the backlit view of the DCE experiment (35% O_2 , 65% He, 1 bar, d_o=4mm).



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Figure 2 Two representative UV flame-image sequences $(30\% O_2, 70\% He, 1 bar)$ at 2s intervals, starting from ignition, for (A) d₀=3mm, illustrating diffusive extinction, and (B) d₀=4mm, illustrating radiative extinction; both droplets burned for about 10s.



Figure 3 Squares of droplet and flame diameters as functions of time (STS-83, 35% O₂, 65% He, 1 bar).



Figure 4 Squares of droplet and flame diameters as functions of time (STS-83, 25% O₂, 75% He, 1 bar).



Figure 5 Calculated (curves) and measured (points) normalized d^2 plots for droplet diameters (30% O_2 , 70% He, 1 bar).



Figure 6 Calculated and measured burning-rate constants K as functions of initial diameter d_o at 1 bar, for both ground-based and space-based experiments.

is larger in helium-oxygen mixtures than in air. This is due largely to the higher diffusivity of helium. The computational results for air are in excellent agreement with experiment for both ground-based measurements and the measurements of larger droplets in STS-94. In helium-oxygen mixtures, the results agree well in 35% oxygen, but at lower oxygen concentrations the predicted values of K exceed the measured values. This may be caused by inaccuracies in available diffusivities of helium under these conditions.³ Such inaccuracies are known to exist for both helium and hydrogen. Computations with improved transport properties are needed to test this explanation.

Figure 7 compares computed and measured histories of flame diameters for droplets burning in helium-oxygen mixtures having 30% oxygen at 1 bar.³ As with the droplet diameters, the flame diameters are seen to lie between the computed diameters for $d_0=3 \text{ mm}$ and $d_0=5 \text{ mm}$. In general, then, our knowledge is sufficient to provide good qualitative agreement. More work is needed to provide better tests of our abilities to describe these droplet-combustion processes, especially with respect to flame diameters at extinction.

Much of the extensive data obtained by DCE in STS-94 has not yet been fully analyzed and compared with predictions. The data in atmospheres at reduced pressures in particular need further attention. As is typical in detailed scientific investigations, these analyses are time-consuming, and definitive results are to be expected in five-year rather than one-year periods. It is fortunate that STS-94 has provided so much reliable DCE data that study of it will be able proceed for an extended time in the future. Pursuit of this future research may be expected to further improve our knowledge of droplet combustion.

<u>Conclusions</u>

The DCE experiments have provided a great deal of information on the combustion of heptane droplets, especially in helium-oxygen mixtures. In particular, both radiative and diffusive extinctions were obtained, and the boundary between them was delineated approximately.

It was established that, at one atmosphere and about 300 K ambient temperature in oxygenhelium atmospheres, heptane droplet combustion experiences diffusive extinction for 35% oxygen and radiative extinction for 25% oxygen for droplets of initial diameters between about 2 and 4 mm. At 30% oxygen diffusive extinction occurs for small droplets (less than about 3 mm initial diameter) and radiative extinction for large droplets (greater than about 4 mm initial diameters). After ignition the square of the droplet diameter decreases linearly with time even through the flame diameter behaves in a more complex manner that is consistent with current understanding of the dynamics of droplet combustion.

Average burning-rate constants increase with increasing oxygen content of the atmosphere and exhibit some variations with initial droplet diameter and with droplet motion. Soot production experiences a rich variety of evolutionary behavior and is much stronger in 35% oxygen than in 25% oxygen. Soot-cloud diameters divided by droplet diameters (not shown above) increase somewhat with time and correlate approximately with time from ignition scaled by the burning time for different initial diameters. Many additional details of the droplet-combustion process were observed and explained.

Much more research remains to be done on the basis of these results. Radiative extinctions need to be analyzed in a quantitative manner theoretically, as do diffusive extinctions for situations in which the flame is influenced by the outer transient zone surrounding the quasisteady burning region. Fuel pyrolysis in the gas and the potential for absorption of pyrolysis products by the liquid require further quantitative study. Many aspects of soot production and soot-particle histories deserve further attention. As with many fundamental scientific investigations, the present work thus has uncovered an appreciable number of additional areas worthy of further study.

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Figure 7 Calculated and measured flame-diameter histories (25% O₂, 70% He, 1 bar).

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Non-Technical Summary

Fire is one of humankind's oldest tools yet one of the least understood. The DCE project helps to improve understanding of droplet burning – fires of liquid fuels – an important topic because such fuels account for more than 95% of energy usage in the transportation sector. In the DCE tests on MSL-1, the liquid fuel studied was normal heptane, a representative hydrocarbon whose combustion characteristics are relevant to those of all petroleum-derived fuels. Improved understanding of heptane droplet combustion ultimately can contribute to reduction of air pollution, better fire safety of liquid fuels and conservation of precious fossil-fuel resources.

In DCE, measurements were made of the combustion histories of individual spherical fuel droplets burning in different atmospheres. Free droplets were formed by injection of the liquid fuel through two opposed hypodermic needles and retraction of the needles to leave the droplet stationary in the combustion chamber. Because of the weightlessness in Space Shuttle the droplet does not fall. The droplet is ignited by two hot wires, which also are retracted to leave the burning spherical droplet alone in the field of view of the measurement system. The principal measurements made were motion-picture views of the history of the droplet diameter and spectroscopic views of the history of the flame diameter. Again because of weightlessness, the flame surrounding the droplet remains spherical instead of rising buoyantly into a familiar teardrop shape.

The combustion history was found to depend on the initial droplet size and the atmosphere in which the droplet burned. In sufficiently oxygen-rich atmospheres, the liquid droplet eventually vaporized completely and the flame around it first grew in size then decreased to a very small diameter, at which point it extinguished. We call this behavior diffusive extinction because the heat loss causing extinction occurs by diffusion. In sufficiently oxygen-poor atmospheres, the flame grows then extinguishes near its maximum diameter, leaving unburnt liquid fuel behind in the droplet. We call this behavior radiative extinction because the heat loss occurs by radiation. In intermediate-oxygen atmospheres, the initially larger droplets exhibited radiative extinction and the smaller ones diffusive extinction. Radiative extinction had not been observed before for heptane droplet combustion because it was not possible to burn large enough droplets in ground-based microgravity facilities. The observed burning times of ten to twenty seconds are too long to be studied in earthbound facilities without influences of buoyancy and therefore needed these Spacelab experiments.

The experiments provided a large amount of data on droplet combustion, including data on the formation of soot during burning, an important process in air pollution. Analysis and interpretation of this data are expected to continue for about another five years. It is through longterm, careful study of experimental results like these that scientific investigation improves our knowledge of the physics and chemistry of combustion.