COMBUSTION OF TWO-COMPONENT MISCIBLE DROPLETS IN REDUCED GRAVITY

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Background

Many efforts to understand sprays have focused upon studying isolated droplets. Information gained from these studies is often not directly transferable to spray situations. However, isolated-droplet studies are useful in that they allow certain phenomena to be studied under well-controlled and simplified conditions. When theory and experiment agree for simplified situations, predictions for more complex cases (where accurate experimental data may not exist) may be made with more confidence. Though small droplets are of practical concern (e.g., < 50 μm as found in some spray systems), it is difficult to perform detailed experiments with them. This is because small droplets are difficult to deploy and ignite without significant perturbations, and they may be difficult to resolve photographically. Hence, most detailed investigations into single-droplet combustion have focused upon larger droplets which are easily observed. In this program, reduced-gravity combustion behaviors of miscible binary droplets initially about 1 mm in diameter are studied. Gravity levels significantly less than 10^{-3} g's are required to render buoyancy negligible for these size droplets.

In practice, liquid fuels are usually multicomponent. It is important to understand the vaporization mechanisms of multicomponent fuels, especially the rates that fuel components with differing properties are vaporized from the liquid phase. This is because gas-phase phenomena depend on the components that have been vaporized, while liquid-phase phenomena depend on the components remaining in the liquid. The simplest multicomponent fuels are (arguably) binary and miscible. When there is a significant volatility difference between droplet components, binary droplets may exhibit a three-stage combustion history where two periods of nearly d^2-law combustion (where the square of the droplet diameter decreases linearly with time) are separated by an intermediate period where vaporization rates are low and flame diameters decrease significantly. These behaviors are thought to be a result of a sudden buildup of the lower-volatility component at the droplet surface[1,2], necessitating a sudden increase in the droplet surface temperature. During the first period of d^2-law vaporization, most of the fuel being vaporized is the higher-volatility component, while during the second d^2-law vaporization period, if liquid-phase species diffusivities are sufficiently small, both liquid components are vaporized at rates nearly proportional to their average liquid mass fractions[3]. As the droplet surface temperature increases, the droplet interior may be heated above temperatures required for bubble nucleation and growth in regions rich in high-volatility species that are present as a result of chemical stratification in the liquid[1-3]. As a result, a bubble may grow and fragment a droplet (the droplet may experience a "disruption" or "microexplosion").

Scientific Objectives

This research focuses on the combustion of binary miscible droplets initially in the mm size range. Experiments are performed using the NASA Lewis 2.2 sec drop tower in Cleveland, Ohio, where mixtures of alkanes and/or alcohols are studied. The fuel components are selected to have significantly different volatilities. Initial oxygen mole fractions from about 0.15 - 0.5 and initial pressures from 0.3 - 2 atm are employed. Different inerts are used (He, CO_2, Ar, N_2) to change burning rates and sooting behaviors. Objectives are to observe the following:

> Transient droplet diameters (including three-staged combustion behaviors and microexplosion).
> Transient flame behaviors (sudden flame contraction, luminosity, extinction).
> Behaviors of observable soot particles.

Theoretical and computational research in support of this program has also been undertaken. This research includes:

> Analytical studies to determine the effects of small but nonzero gravitational levels on droplet gasification.
> Analytical studies of hydrodynamic stability of spherically-symmetrical droplet gasification (to address the question as to whether spherically-symmetrical droplet gasification may be destabilized from capillary, i.e., Marangoni effects).
> Computational modeling of effects of capillary stresses on droplet gasification.

Significance

The reduced gravity experiments should yield insight into the efficiency of liquid-phase species diffusion, since this diffusion may markedly influence combustion behaviors (e.g., three-staged combustion behaviors). By noting times for flame contraction to occur, calculations for effective liquid-phase species diffusivities may be made (e.g., with asymptotic or with numerical models). Finally, unique sooting behaviors may appear since gas-phase compositions may differ significantly initially (e.g., by varying diluents and oxygen concentrations) as well as during an experiment (from variations in the composition of fuel gasifying from a droplet). Differences in sooting may be observable photographically by noting behaviors of apparent flame luminosities and soot particles large enough to be observed.

The theoretical and computational efforts will aid interpretation of the reduced-gravity experiments as well as improve understanding of fundamental phenomena (e.g., capillary stresses and buoyancy effects).

Experimental Approach

The reduced-gravity experiments are performed using the NASA Lewis 2.2 sec Drop Tower. A new drop rig was built for this purpose. The electronics on the apparatus, except for some modifications, were based on the design of an existing NASA apparatus which is described elsewhere[4]. The mechanical design is new, though it is based on knowledge and experience accumulated at NASA. A ceramic fiber (≈ 12 μm) is sometimes used to stabilize droplets; concepts for the fiber were obtained from Dr. D. Dietrich[5]. In an experiment, a droplet is grown (in a pressure chamber) between two opposed hypodermic needles. The needles are retracted simultaneously in opposite directions, leaving the droplet essentially at the location where it was grown. The deployment induces oscillations in the droplet shape at a frequency higher than 370 Hz (the cine camera framing rate). These oscillations damp early in the combustion history. The droplet is ignited using two symmetrically-placed ignition sparks (≈ 100 mJ each). The spark electrodes are retracted after ignition, and droplet and flame behaviors are observed photographically using two ccd video cameras (for real-time observations) and two 16 mm cine cameras. Orthogonal views are taken; one backlit view is a close-up of the droplet while the other shows the flame (no backlight). In most experiments a ceramic fiber was used which held droplets nearly motionless and even if there was a drift along the fiber it was very small (≈ 0.1 mm/s) and insignificant in terms of possible residual convection. Experiments that did not use a fiber typically had a droplet drift velocity (up to several mm/s).

A Sun workstation-based digital image processing system was developed for analysis of cine films and ccd images from drop tower experiments. Basic concepts for this system were obtained from the literature[6]. Software for the Sun-based system was developed with the assistance of Virtual Visions Software[7].

Experimental Results

During the summer of 1992, exploratory experiments with heptane/hexadecane mixture droplets were performed at the NASA Lewis Research Center 2.2 sec drop tower. An existing droplet combustion apparatus[4] available at NASA Lewis was used for these experiments. Experiments were attempted both with and without a ceramic support fiber (≈ 10 μm diameter) being present. The ceramic fiber was utilized to prevent significant droplet drift after deployment. Results from these preliminary experiments have been reported elsewhere[8].

In Fall 1994 and Winter 1995, a large number of experiments were performed at NASA Lewis with the new drop apparatus. Because of time limitations, we have not been able to analyze all of the films in time for this workshop. We are presently in the process of reviewing the experiments, though we do have some detailed results and a number of general observations regarding 35 of the 133 successful experiments (out of 203 total experiments that were performed). The high success rate is due to the fact that the support fiber was used in many of the experiments. Results and observations are listed below.

Fuel Mixtures

The experimental program covered a wide range of parameters. Two main groups of fuels were studied - alkanes and alcohols. Within the alkanes, mixtures of heptane/hexadecane ranging from pure heptane through 95, 80 70, and 60% heptane (by mass) were investigated the most. Other mixtures studied also consisted of pentane and dodecane mixed with mainly heptane at various ratios. The alcohols studied were methanol and ethanol mixed with 1-dodecanol in proportions which were much the same as for the hydrocarbons. A few exploratory experiments were also performed with mixtures of heptane/methanol, heptane/ethanol, and heptane/hexadecane/dodecanol. These tests have not yet been reviewed though one observation regarding a mixture of heptane/methanol at proportions of 80/20% shows two burning regimes with regard to sooting; in the first burning period of time the droplet is sooting and it is non-sooting the second period.

More complex mixtures involving three liquid-phase components initially in a droplet were investigated as well. The components were selected to have significantly different volatilities with the hope that two flame contractions could be observed during the combustion of a single droplet. In at least one experiment it appears that a double contraction does occur. Finally, several mixtures of heptane/hexadecane and pentane/heptane where the low volatility fuel was in excess were investigated and it was observed that flame contractions occurred. This may be due to sooting effects[9].

Gas Mixtures

A large range of diluents (in addition to atmospheric air), consisting of N_2, CO_2, He and Ar were used in different mixtures and at different ratios ranging from 5 to 70% of diluent. The gas mixtures were composed mainly of O_2 and one of the diluents but some experiments were conducted with combinations of diluents. The primary reason that He was originally selected was to increase the burning rate in order to burn droplets to completion for droplets as large as possible. It turned out that He in addition strongly inhibited sooting as well as the likelihood of achieving microexplosions. Adding He also increased the brightness of the blue reaction zone. Some of these observations are discussed in the following paragraphs. CO_2 was used to observe its effect on sooting behavior; it was found that replacing N_2 with CO_2 reduced sooting. This effect has been reported in the literature for droplets burning in normal gravity[10]. It was also found that replacing N_2 with Ar increased sooting, though experiments with Ar have yet to be fully analyzed. An attempt was made to determine a sooting threshold (based on visual observations) for Ar and N_2 when these species are added to a basically non-sooting mixture consisting of O_2 and He. The He or N_2 concentrations were varied systematically in order to observe changes in sooting behavior. These tests have not been yet fully analyzed but it appears that such a threshold does exist.

O_2 concentrations were varied from 15 to 50% (molar) to study the effect on burning rates and sooting. At atmospheric pressure, it was found that even with He as a diluent (which strongly inhibited sooting), raising the oxygen concentration above 35-40% produced considerable sooting. Behaviors in subatmospheric pressures were different and will be briefly discussed later.

Initial Diameter Effects

In the experiments conducted, efforts were made to provide data for droplets with different initial diameters. The largest droplets investigated were initially about 1.8 mm in diameter while the smallest ones were initially about 0.5 mm in diameter. A full analysis of the effects of initial droplet diameter on combustion histories has not yet been completed, though some initial observations can be presented. Generally, it was found that increasing the initial droplet diameter increased the amount of visible soot. In addition, the largest droplet investigated appears to have an abnormally-low burning rate (see Fig. (1)) compared to initially-smaller droplets with the same initial composition burning in the same atmospheres. Because of free-fall time limitations, the largest droplet shown in Fig. (1) could not burn to completion, so it cannot be ascertained whether the burning rate for this droplet would increase at later times in its combustion history.

Pressure Effects

Most experiments to date have been conducted at atmospheric pressure. However, some exploratory experiments were performed with pressures as low as 0.3 atm (abs) and as high as 3 atm (abs). We have not analyzed this data yet, and only general observations can be offered at this time. For example, mixtures of heptane/hexadecane which were heavily sooting at 1 atm did not visibly soot at subatmospheric pressure for the same gas composition. Vacuum also appreciably reduced burning rates and as such most of the experiments were conducted in an atmosphere consisting of O_2/He at a ratio of 50/50%; however, some experiments used air. Reducing the pressure also has the effect of making it possible to observe the blue flame zone in air for the entire burning history. Because of the brightness of soot radiation, this was not possible at 1 atm.

The tests in high pressure were performed in O_2/He environments. One definite observation that can be offered at this time is that increasing the pressure significantly increases sooting for hydrocarbon droplets (relative to 1 atm, with the same initial droplet diameters and gas-phase compositions).

Visible Soot Dynamics

Heptane/hexadecane mixture droplets burning in air were highly sooting except during the flame contraction. In O_2/He atmospheres, droplets typically were sooting only for an initial period of time after which the soot appeared to disappear, possibly because of decreased fuel residence times.

In He/O_2 environments, the blue flame zone could be observed to lie outside a yellow zone corresponding to radiant soot emissions for the entire combustion history (Fig. (2)). Interestingly, the radius of the yellow zone did not always correspond closely to the radius of the blue zone. In many of the O_2/He experiments, the yellow zone contracted slowly and typically disappeared, while the blue zone remained at about the same diameter until it also contracted, but only for a short time, after which it grew again. The contraction of the blue zone is most likely related to droplet heating effects caused by a sudden increase in the surface hexadecane mass fraction of the droplet. Data showing these behaviors are shown in Fig. (3), while data for the droplet diameter for this experiment are shown in Fig. (4). The disappearance of the yellow zone may be related to decreased fuel residence times inside the flame, since the droplet is continuously decreasing in size. This behavior needs to be explored, further, however.

Soot "shells" (i.e., high concentrations of soot that form a dark ring between the droplet and the flame that is visible in photographs, see Fig. (5)) were observed in numerous tests with heptane/hexadecane fuel mixtures, though never when O_2/He mixtures were used, even though soot was present as evidenced by the yellow colors in the flame photographs. Sometimes more than one soot shell co-existed (concentrically) for a certain period of time, as shown in Fig. (5). These soot shells would slowly merge into a single shell. Soot shells were not present when He alone was used as a diluent, though they were almost always present when N_2 or CO_2 were used as diluents above 0.4 atm (even in combination with He). When (yellow) flame contractions were observed, the soot shells also were observed to contract at the same times (Fig. (6). The soot shell contraction in many instances ends by what appears to be absorption of soot into the liquid phase, though a soot shell may sometimes reappear (Fig. (6)); see Fig. (7) for the droplet diameter history for this experiment.

Blue Flame Zones

In air with various mixtures of heptane/hexadecane ranging from 95/5 to 60/40% (by mass) respectively one can observe the following type of behavior from the flame-view images: for approximately 0.04 - 0.06 seconds after ignition, a blue circular flame is visible after which soot starts to form inside this blue region and eventually the blue flame is no longer visible due to high light intensity soot radiation (though at 0.4 atm or lower, sooting is significantly reduced which allows observation of the blue flame zone throughout the combustion history, as noted above). In an atmosphere consisting of O_2-He 30/70% (molar) respectively with the same type of liquid fuel mixtures the blue flame sheet is visible throughout the entire test even when soot is formed inside the region between the droplet and the flame. The flame "sheet" actually has a finite visible thickness, and is at least as thick the droplet diameter (at least in O_2-He environments). A minor change in the O_2-He proportion to 35/65% causes the blue flame sheet to not to be visible for most of the test even though the brightness of the luminous zone appears to be very similar. With mixtures of alcohols (methanol/1-dodecanol or ethanol/1-dodecanol) in atmospheric air the blue flame is not visible at all in the color photographs except for a short time after ignition.

Droplet Microexplosion

Microexplosions occurred quite frequently in the various tests and even though a detailed analysis of this aspect has not yet been performed the following general observations can be made: (1) in most cases where the environment was composed of O_2-He the likelihood of microexplosive events for the same liquid fuel, pressure and droplet diameter was less than when compared to air which suggests that He might inhibit this phenomenon; (2) in a large number of tests (among those that were viewed) the microexplosions did not shatter the droplets, and in most cases the droplets regained spherical shapes, though some fuel was lost during the process and the droplet diameter sometimes decreased sharply; (3) in a number of tests, there were several microexplosions after the first one, and droplets sometimes were observed to trap gas bubbles during the microexplosion event; (4) microexplosions frequently occurred after flame contractions (this behavior has also been observed in previous experiments[11,12,13]).

Flame Contraction Analysis

The onset of flame contraction (i.e., where the flame rapidly decreases in size) is experimentally a rather abrupt event (Figs. (3), (7), (8) and (9)) that can be compared with theoretical predictions. Shown in Fig. (9) are comparisons of flame-contraction predictions of an asymptotic theory[2] with the experimental results that have been reduced to date. The ratio V_c/V_0 is the ratio of the droplet volume at the onset of flame contraction to the initial droplet volume. The solid lines are plots of the equation $V_c/V_0 = Y/[Y + 3\epsilon(1 - Y)]$ where $\epsilon = 8D/K$ is a small parameter of expansion (D is the liquid species diffusivity and K is the burning-rate constant based on droplet diameter) and Y is the initial hexadecane mass fraction. The open and closed triangles and the open circles are data points. The experimental values of V_0 used in this graph were increased by a value of 10% to approximately account for swelling which will occur when the droplets are heated after ignition. Typical ϵ values expected are in the range 0.01 - 0.1, and these values evidently are applicable in Fig. (9). It is noted that similar comparisons of experiment and theory have been published for heptane/hexadecane droplets that were burned in reduced-gravity at supercritical pressures though for a limited range of gas-phase compositions[13]; we will compare our results with these data shortly.

Extinction

Detailed analysis of data from two experiments showed that extinction appears to have occurred; other experiments, which have not yet been fully analyzed, show indications of extinction as well. The extinction diameter measured in one experiment was found to be about 125 μm for a droplet which was initially about 1.1 mm in diameter. This droplet was composed of heptane/hexadecane (10% hexadecane initially by mass) and burned in an O_2/He environment (70% He molar) at 1 atm. The extinction diameter measured in the other experiment was about 75 μm for a droplet which was initially about 0.91 mm in diameter. This droplet was composed of heptane/hexadecane (20% hexadecane initially by mass) and burned in an O_2/He environment (50% He molar) at 1 atm. The 12 μm fiber was used to support both droplets. It is noted that extinction has previously been observed for binary mixtures of alcohols burning in air under reduced-gravity conditions[12].

Red Flames

The side-view photos occasionally showed reddish coloring for the flames. An example of this behavior is shown in Fig. (10), which actually shows flames associated with two small droplets that were ejected during the microexplosion of a larger parent droplet. In the photo, the flame of one of the small droplets appears to be red (initially it was blue, though this is not shown). The film record shows that the other blue flame will eventually turn red as well. An explanation for this behavior is not available at present.

Analytical Approaches And Results

To aid interpretation of experimental efforts, analytical studies of the behaviors of gasifying droplets in reduced-gravity environments have been performed. These efforts focused on studying: (1) effects of small gravitational levels on droplet vaporization/combustion; and (2)

hydrodynamic stability of spherically-symmetric vaporization/combustion of two-component droplets. Each effort to date has considered evaporating droplets; combustion will be considered later.

In the gravity-level research, a mathematical model taking into account small (and constant) gravitational levels is developed for vaporization of an isolated liquid droplet suspended in a stagnant atmosphere. A goal of the analysis was to determine how small gravitational levels affect droplet gasification characteristics. Attention is focused upon determining the effects on gas-phase phenomena. The conservation equations (including viscous effects) were normalized and nondimensionalized, and a small parameter that accounts for the effects of gravity identified. This parameter is a Richardson number based on the gravitational acceleration, the droplet radius, and a characteristic gas-phase velocity at the droplet surface. Asymptotic analyses were developed in terms of this parameter, and different spatial regions were identified. Near a droplet, gravitational effects are negligible in the first approximation, and the flowfield is spherically symmetric to the leading order. Analysis shows, however, that outer zones exist where gravitational effects cannot be neglected; it is expected that a stagnation point will be present in an outer zone that is not present when gravity is totally absent. The leading-order and higher-order differential equations for each zone are derived and solved[14]. Sketches of predicted flowfields are shown in Figs. 11 and 12.

The stability analyses have addressed the problem of hydrodynamic stability of a two-component droplet undergoing spherically-symmetrical gasification. The problem is formulated as a linear stability analysis, with a focus of predicting when spherically-symmetric droplet gasification can be expected to be hydrodynamically unstable from surface-tension gradients acting along the surface of a droplet. The conservation equations (including viscous effects) for the gas and liquid phases were perturbed, and the perturbation equations were simplified and solved[15]. Droplet components were assumed to have characteristic liquid species diffusion times that are large relative to characteristic droplet surface regression times. It was initially found that for the conditions assumed (quasisteady gas phase, no initial droplet temperature gradients, diffusion-dominated gasification, appreciable initial concentrations of both components in the droplet, negligible gravity), surface tension gradients should not influence stability characteristics in the limit of vanishingly-small liquid species diffusivities. Further analyses (which have not yet been published) have shown, however, that spherically-symmetrical gasification may be hydrodynamically unstable under conditions that are representative of reduced-gravity droplet gasification experiments when liquid-phase species diffusivities are small but nonzero. A neutral stability criterion has subsequently been derived for droplet evaporation. Analysis of this result is underway.

Computational Approach And Results

Detailed computational modeling (in collaboration with H. A. Dwyer) has focused upon predicting the effects of capillary flows (from droplet surface temperature and/or composition gradients) on droplet vaporization. Briefly, the governing equations are cast in a finite-volume form, and solved numerically for transient liquid- and gas-phase variables. Variable properties in both the liquid and gas phases are allowed. Calculations are performed using the Sun workstation also used for image processing. Modeling of single-component droplet vaporization in a hot environment has clearly shown that surface-tension gradients dramatically influence droplet vaporization and internal circulation behaviors under conditions that are representative of slowly-drifting droplets in reduced-gravity experiments as well as rapidly-translating droplets in practical high-pressure sprays[16,17,18].

Future Plans

The UC Davis drop rig will be modified to so that pressures substantially below 1 atm may be investigated; we presently have problems forming droplets at low pressures, leading to a low experiment success rate. Once this modification is completed, experiments will be performed with binary mixtures of alkanes and/or alcohols in various gas mixtures. By comparing the data obtained with the previously-obtained higher-pressure data, we hope to obtain information on the effects of sooting on droplet combustion phenomena (e.g., onset times for flame contraction). We also plan to investigate use of Ar as a diluent more extensively, since it will likely change the sooting characteristics of the droplets. The computational model will be extended to combustion of two-component miscible droplets, for which surface-tension gradient effects are expected to be especially significant. In addition, the gravity-level and hydrodynamic-stability analyses will be extended to include gas-phase chemical reactions (combustion).

Acknowledgments

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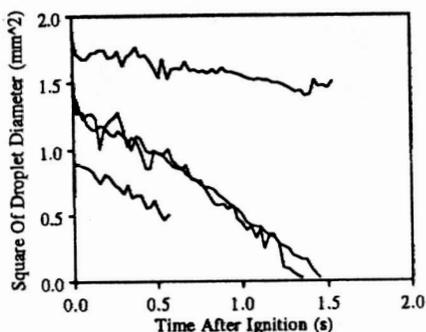
Fig. 1 Droplet diameter histories for heptane/hexadecane mixture droplets (initial hexadecane mass fraction of 0.3) burning in O_2/He environments (35% O_2 molar) at 1 atm.
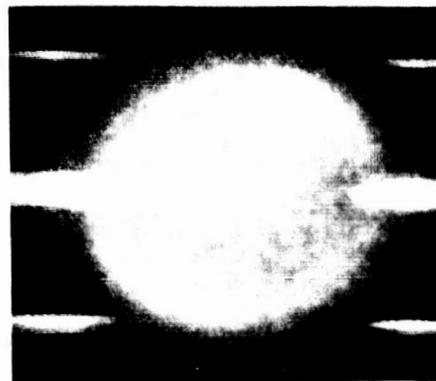

Fig. 2 Flame image of a heptane/hexadecane mixture droplet (initial hexadecane mass fraction of 0.1) burning in an O_2/He environment (30% O_2 molar) at 1 atm.
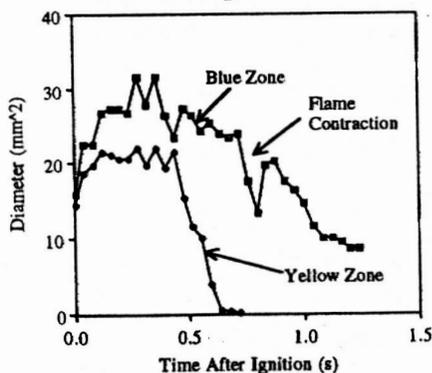

Fig. 3 Blue and yellow flame diameter histories for a heptane/hexadecane mixture droplet (initial hexadecane mass fraction of 0.05) burning in an O_2/He environment (30% O_2 molar) at 1 atm.
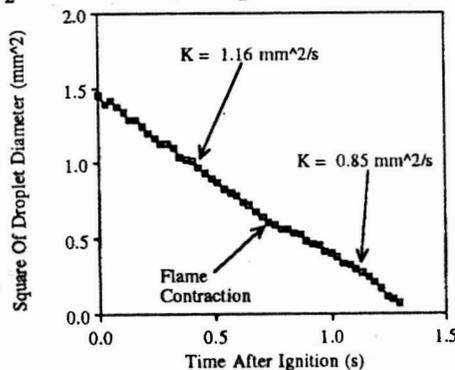

Fig. 4 Droplet diameter history for a heptane/hexadecane mixture droplet (initial hexadecane mass fraction of 0.05) burning in an O_2/He environment (30% O_2 molar) at 1 atm.

Fig. 5 Double soot shell for a heptane/hexadecane mixture droplet (initial hexadecane mass fraction of 0.4) burning in an O_2/He/CO_2 (35% O_2, 55% N_2 molar) environment at 1 atm. The two soot shells slowly merge into one.
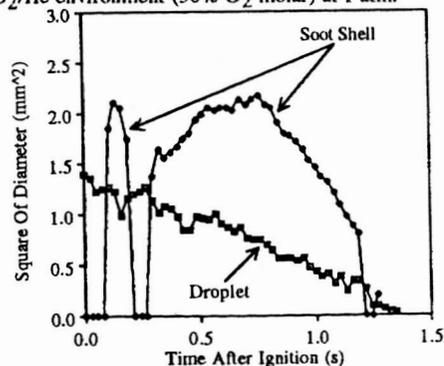

Fig. 6 Soot shell and droplet diameter behaviors for a heptane/hexadecane mixture droplet (initial hexadecane mass fraction of 0.3) burning in an O_2/He environment (35% O_2 molar) at 1 atm.
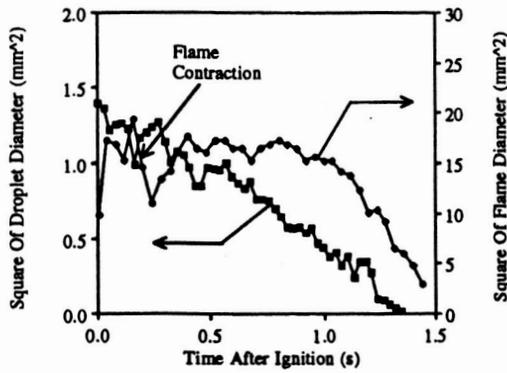
Fig. 7 Droplet and flame diameter behaviors for a heptane/hexadecane mixture droplet (initial hexadecane mass fraction of 0.3) burning in an O_2/He environment (35% O_2 molar) at 1 atm.
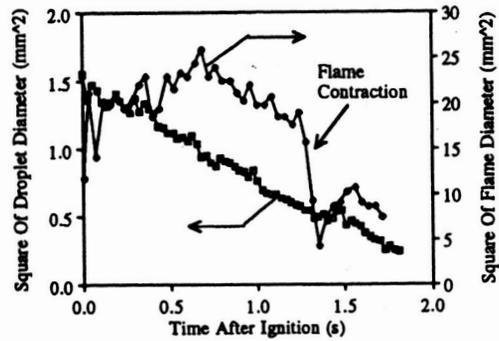

Fig. 8 Droplet and flame diameter histories for a heptane/hexadecane mixture droplet (initial hexadecane mass fraction of 0.05) burning in air at 1 atm.
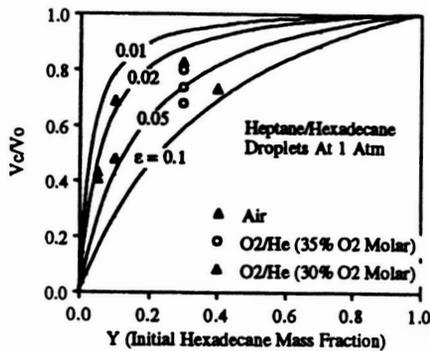

Fig. 9 Comparison of theory and experiment for the droplet diameter ratio at the onset of flame contraction.
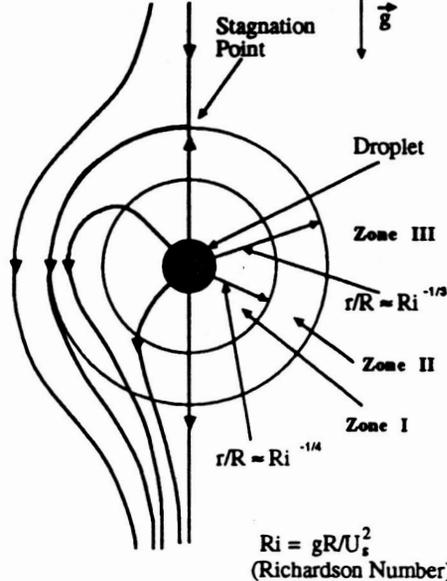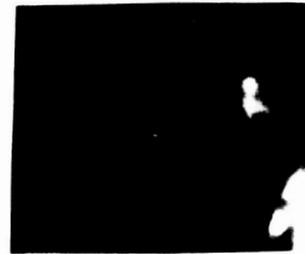

Fig. 10 Photo showing the red and blue flames for small droplets ejected from a larger droplet during a microexplosion. The larger droplet was initially composed of heptane/hexadecane (initially 10% hexadecane by mass). The environment is O_2/N_2 (50% O_2 molar).

$$Ri = gR/U_s^2$$
(Richardson Number)

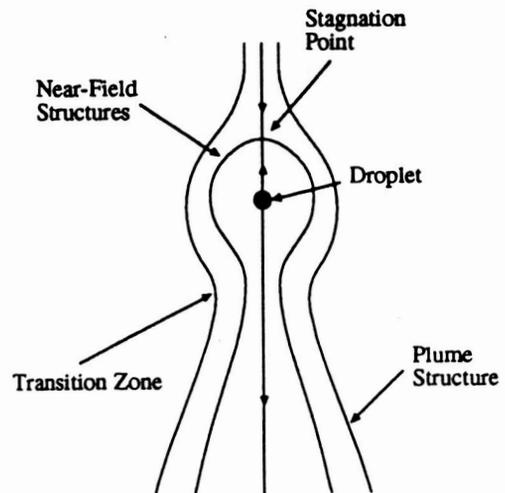Fig. 11 Qualitative sketch of the flowfield near a gasifying droplet.

Fig. 12 Qualitative sketch of the flowfield far from a gasifying droplet.

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