THE COMBUSTION OF UNSUPPORTED HEPTANE/HEXADECANE MIXTURE DROPLETS AT LOW GRAVITY

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The results of an experimental study of the effect of fuel composition on burning of unsupported droplets in a reduced gravity environment are reported. The liquids studied were mixtures of heptane and hexadecane. The experiments were conducted in a drop tower which provided an experimental observation time of about 1.2 s. The experimental technique consisted of projecting a test droplet within a closed container in a near vertical trajectory and then releasing a camera positioned at the apex of the droplet's trajectory, the combustion chamber containing the droplet, and associated instrumentation into free fall when the droplet reached the apex. In this way the test droplet was effectively captivated with respect to the moving frame of reference. Some droplets occasionally drifted with respect to the camera during free fall, and this motion corresponded to a droplet Reynolds number of the order of .01 or smaller.

The evolution of droplet diameter was similar to that of the classical "d^2" law, though with some evidence of droplet heating and extinction being revealed for some of the liquids. Microexplosions were not observed for the range of compositions studied.

Introduction

This paper reports the results of an experimental investigation of the combustion of unsupported droplets of miscible binary mixtures of heptane and hexadecane in a low gravity environment (on the order of 10^{-3} that of earth normal gravity). The drop tower method was used to measure the evolution of diameter from which the burning rate and total burning time were obtained as a function of initial droplet composition. A typical experiment was carried out by propelling upward in a near vertical trajectory the test droplet within a small combustion chamber containing air, and then releasing a high speed movie camera, test chamber containing the droplet, and associated instrumentation into free fall when the droplet reached the apex of its trajectory. In this way the droplets were effectively levitated with respect to the falling package for the duration of free fall (which in the present experiments was 1.2 s). Some of the droplets studied exhibited a very small drift velocity which corresponded to a Reynolds number (based on the initial droplet diameter) of the order of .01. The information obtained consisted primarily of a high speed movie record of the droplet burning process.

A unique feature of the experimental method was that of providing unsupported droplets in a low gravity environment and of recording their complete combustion. The creation of stationary unsupported droplets, though, was a major experimental challenge. The importance of low gravity effects resides in understanding how fuels burn in a spacecraft propulsion environment. Mixtures were studied because fuels are seldom encountered which are single component. A binary mixture in particular represents the most elementary configuration of a multicomponent fuel, and the effect of composition is most directly elucidated for such mixtures. The mixture components were chosen to yield nearly ideal solutions in order to facilitate physical property predictions which might be required in future model development.

The drop tower was first used to study the combustion of single component fuel droplets suspended from a fiber in 1957 by Kumagai and Isoda et al.[1] Since that time the method has been used to study the low gravity combustion of coal particles, emulsions, and high pressure effects under low gravity for single component fuels (see, for example, Ref. 2 to 6). These studies used droplets suspended from a fiber. Seven studies are known in which the combustion of *unsupported* fuel droplets at low gravity was reported.[7-13] All involved single component liquids. In six of these studies the method of droplet generation in these studies consisted of first holding captive a test droplet by a support fiber or needle and then freeing the droplet by a sudden jerking motion of the support.

Recently, Avedisian et al.[13] observed the complete burning of unsupported single component fuel droplets (heptane and toluene) in a drop tower. The major improvement in their work involved using a different method of generating the droplets. The droplets were generated by using a variant of the "ink-jet" method,[14] which made it possible to resolve some of the timing and droplet size problems encountered in prior work.

Previous studies of multicomponent fuels have been reported at earth normal gravity. Both suspended droplets[15] and freely falling[16] or rising[17,18] droplets have been employed. A unique feature of a miscible mixture of liquids is the potential for so-called disruptive combustion or "microexplosion." In this process, the droplet literally explodes during burning. The mechanism involves superheating volatile liquid which is trapped within the droplet interior during burning because of the relatively slow rate of liquid phase mass diffusion within the droplet. A bubble forms by homogeneous nucleation, it grows rapidly, and the droplet then shatters if the bubble growth rate is high enough. The liquid state at which microexplosions are initiated depends on liquid composition and ambient pressure. In the present work the pressure was fixed at .101 MPa thus leaving composition and fuel type as the free parametric variables. The method of droplet generation has also recently been suggested as influencing the process.[16] The microexplosion phenomenon was not observed for the mixture compositions studied.

Description of the Apparatus

Figure 1 illustrates a schematic diagram of the Cornell drop tower facility. The major components are the following: 1) the drop shaft which spans two floors, 2) drop package, 3) a droplet generation unit, 4) a high speed movie camera with associated lighting, 5) timing control unit, 6) electromagnet, 7) deceleration tank, and 8) associated instrumentation such as combustion chamber, support stand, and retrieval system. Only a brief description of the procedures and instrumentation will be provided here. More details are described in another publication.[13]

The heart of the experiment is the droplet generator and timing control unit. The droplet generator was based on the ink jet method for generating droplets. The design is similar to that used by Wang.[14] A continuous droplet stream was created by the generator in a near vertical trajectory and then shut off. The last droplet emerging from the nozzle was the one studied in the experiment. When this droplet reached the apex of its trajectory, it was ignited by a spark discharge from two electrodes placed at fixed positions on either side of it.

The intent was to create stationary droplets. However, some of the droplets studied exhibited a small drift velocity. This velocity was indicative of both inaccuracies in releasing the platform precisely at the instant the droplet reached the apex of its trajectory and the fact that the trajectories of the droplets were not vertical (to prevent falling droplets from impacting on the nozzle exit; the trajectory could not be disturbed once set without changing the timing parameters). The droplet velocity for the present experiments corresponded to Reynolds numbers, based on the initial droplet diameter and ambient gas kinematic viscosity, on the order of a maximum of .01. For these Reynolds numbers, the effect of convective motion on the possible internal circulation and droplet burning rate is conjectured to be small, though comparison with a full numerical solution of the type carried out by Sanders and Dwyer[19] would be necessary to more precisely ascertain the effect of motion on the measured burning rate.

A measure of the success of solving the above described timing difficulties is demonstrated in the typical motion picture sequence illustrated in Fig. 2. The framing rate was 250 frames/s. The electrodes appear on either side of the droplet in the first frame only for illustration (in subsequent frames the photographic sequence was narrowed to allow fitting it on a single page) and the black blotches at the top and bottom of each frame are frame markers of the movie camera. The position of the droplet with respect to these markers (the moving frame of reference) provided a measure of the relative motion of the droplets during their burning. As is evident, the motion of the droplet with respect to the camera (i.e., the markers) is minimal. The comparatively focused droplet image illustrated in Fig. 2 demonstrates that the droplet motion was essentially in a plane perpendicular to the line of sight of the camera.

Measurements of droplet diameter were made directly from motion picture sequences typical of those illustrated in Fig. 2. Each motion picture image was projected on a grid and the boundary of each image was traced on the grid. Droplet dimensions in two perpendicular planes were read directly from the traced images. The ratio of the two dimensions was never more than .95 so that the droplets were essentially spherical. Systematic errors were introduced by the ability to discern the boundary of the droplet on the projected image. This problem was alleviated by maximizing the focussing and clarity of the motion picture image (cf, Fig. 2). This error was significantly only near the end of burning when the droplet diameter became very small—on the order of 40 μm or smaller. The uncertainty in measuring diameters of this size can be very high, depending on how focused the droplet image was. For this reason, droplet dimensions
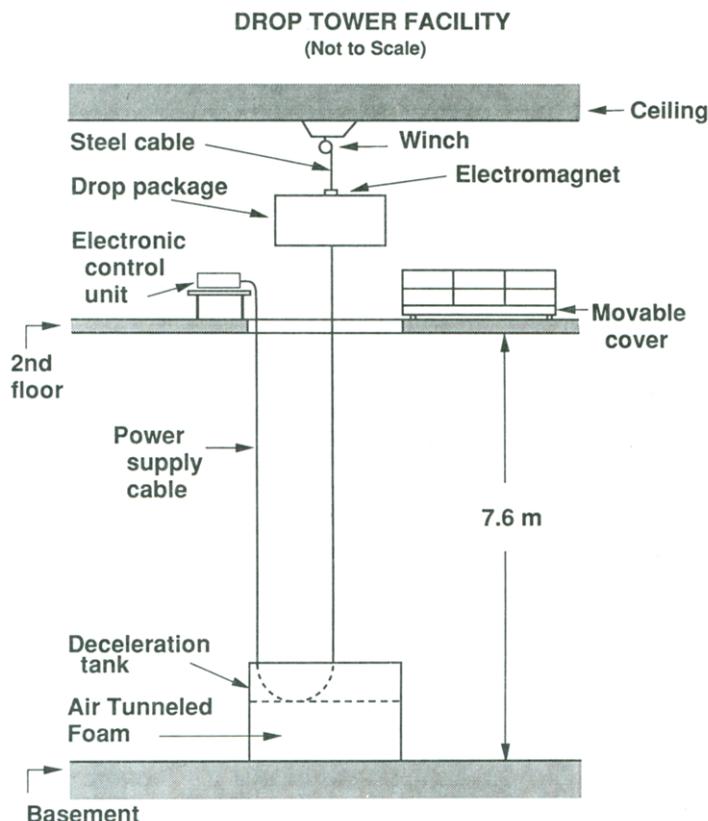
DROP TOWER FACILITY
(Not to Scale)

FIG. 1. Drop tower facility

smaller than about 40 μm are not reported, though images of this size could still be seen on the photographic record.

Direct back lighting was used to provide the type of illumination illustrated in Fig. 2. Because of the low luminosity of the flames ("bluish" in color) which existed around the normal alkane droplets studied, the flame could not be seen clearly with the intense backlighting used. Lower light intensities would have been more favorable in this regard, though at the expense of minimizing errors in the ability to extract the droplet diameter from the photographic record. More luminous flames were required to reveal the flame shape commensurate with the framing rate and light intensity used.[13] The flame shape should not be significantly different from spherical because of the very slow motion experienced by some of the droplets. For larger Reynolds numbers, closer to .1 and higher, the flame shape would be more in the shape of a "tear" dragged along by the droplet.[13]

The existence of a flame around the normal alkane droplets studied here was proven both by the decrease of droplet diameter in the motion picture sequences of droplets in free-fall, and by direct visual observations for burning normal alkane droplets in a stream generated on a stationary table by the droplet generator. In addition, by carefully looking at the combustion chamber during the first few inches of its decent, a flame was always observed around the droplet.

Discussion

The temporal variations of the square of the droplet diameter for several mixtures of heptane and hexadecane are shown in Figs. 3 to 7 in dimensional form. The initial compositions of the mixtures studied were 0, 0.33, 0.50, 0.75, and 1.00 mole fraction hexadecane. Data for pure heptane (Fig. 3) were taken from Ref. 13.

Two stages of burning under low gravity appeared to exist for the mixtures studied: 1) a droplet heating period, and 2) a steady state burning period followed by disappearance of the droplet (i.e., burnout). A third period, when it occurred, was characterized by the droplet diameter remaining constant. This stage was believed to be indicative of extinction. An extinction-like phenomenon was
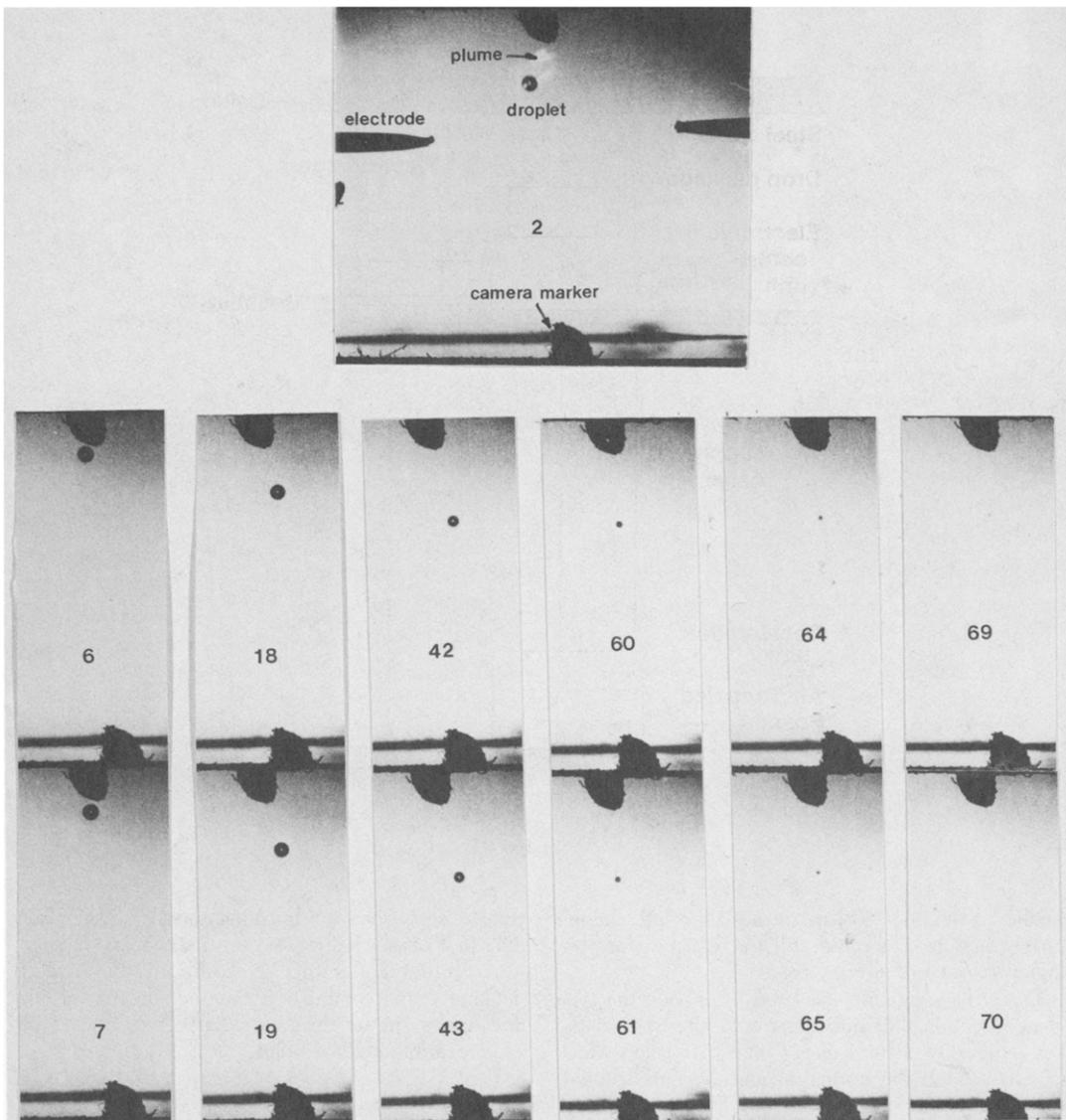
FIG. 2. Motion picture sequence of a droplet containing 0.33 mole fraction hexadecane in heptane at reduced gravity. Time interval between successive frames is 4 ms; droplet diameter in the first frame (top) is 500 μm.

observed only for the mixture components and not for the mixtures themselves (Figs. 3 to 7). The reason for this interesting, if not peculiar, behavior is unknown at present.

Droplet heating was revealed in Figs. 3 to 7 by an initial period after ignition during which the droplet diameter was approximately constant. The droplet evaporation rate is considered to be low during this period because most of the heat transferred to the droplet is used to heat up the droplet to its steady state wet bulb temperature. A phe-

nomenon similar to the droplet heating believed to exist in the present low gravity mixture data has also been previously observed during low gravity burning of single component unsupported droplets.[8,10]

The second stage of combustion of the mixtures studied was characterized by the evolution of the droplet diameter squared exhibiting a nearly linear variation with time. This period may be indicative of steady burning during which both the droplet temperature and the rate of change of diameter
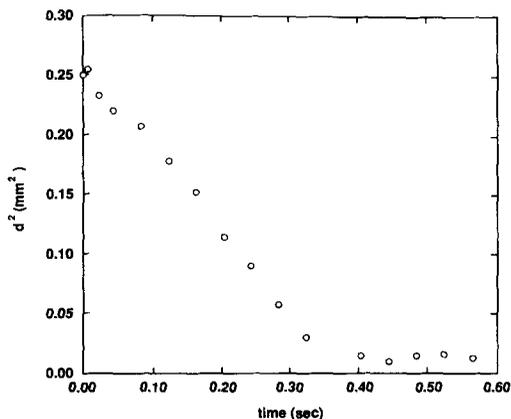
FIG. 3. Variation of diameter squared with time for heptane.[13]

FIG. 5. Variation of diameter squared with time for 0.50 mole fraction hexadecane in heptane.

squared—d^2—are nearly constant. This constant is defined as the "burning rate" constant. A measure of it in Figs 3 to 7 would be provided by the slope of the variation of diameter in this linear region. If such steady burning persisted throughout the entire period of combustion, and the flame shape was spherical and in a constant ratio to the droplet diameter, then a spherically symmetric stationary droplet evolution law for mixtures of the following classical form would result:[21]

$$d^2 = d_0^2 - Kt \qquad (1)$$

where d_0 is the initial droplet diameter and K is the so-called burning rate constant which is determined by measuring the rate of change of diameter:
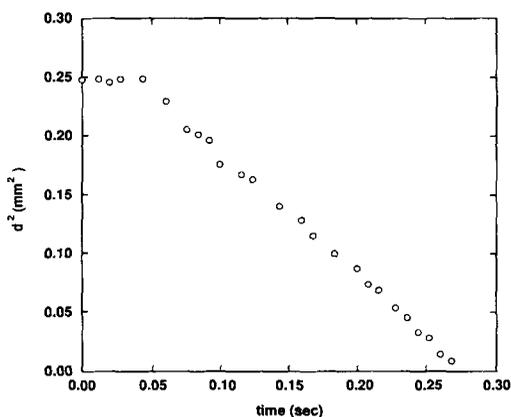
$$K = -d(d^2)/dt \qquad (2)$$

Eq. (2) reveals the principle source of error involved in measuring burning rates from droplet diameter data: the data must essentially be differentiated. Such differentiation magnifies uncertainties in measuring diameter.

For the present experimental conditions Eq. (1) appears not to apply over the entire burning period (cf, Figs. 3 to 7) because of possible transient droplet heating after ignition, of possible extinction, and/or to a lesser extent because of the small drift velocity occasionally exhibited by the droplets, none of these effects of which were accounted for in the development which led to Eq. (1). A burning rate such as is defined by Eq. (2) then becomes an ad-hoc construction which pertains to data that can only span a part of the total burning history—the time domain over which d^2 varies most linearly with time. Various interpretations may exist on how to extract a meaningful burning rate when the evolution of

FIG. 4. Variation of diameter squared with time for 0.33 mole fraction hexadecane in heptane.
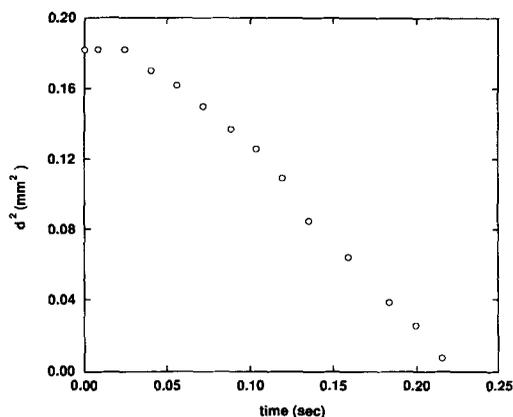

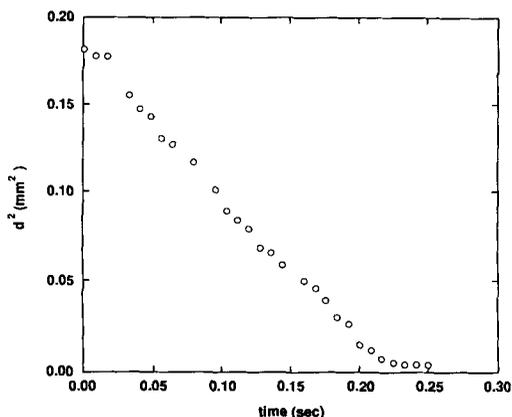FIG. 6. Variation of diameter squared with time for a 0.75 mole fraction hexadecane in heptane mixture.

FIG. 7. Variation of diameter squared with time for hexadecane.

diameter does not follow Eqs. (1) and (2) over the range of complete burning. For example, the burning rate constant for heptane may be calculated as 0.68 mm^2/s if data over the time domain $0 < t < 0.32$ s in Fig. 3 are used. By contrast if only those data are used to define a burning rate constant which are within the time domain over which the variation of d^2 with time is most linear, then we obtain a burning rate constant[13] of 0.79 mm^2/s which agrees with the value of 0.78 mm^2/s obtained by Okajima and Kumagai.[8]

The data in Figs. 4 to 6 show that either the mixtures studied do not exhibit the kind of preferential vaporization characteristic of a distillation controlled mechanism under low gravity conditions, or the resolution of the measurements has not allowed such a mechanism to be revealed. If this mechanism is operative the mixture components will vaporize in the order of their relative volatilities.[16] The burning rate of the component left behind would then be characteristic of that component alone.

For the present data both mixture components apparantly vaporized together throughout the droplet lifetime with no discernible sudden change in the burning rate constant. This fact is suggestive of a diffusion controlled vaporization mechanism, which has also been previously observed for heptane/hexadecane mixtures,[16] though for relatively dilute concentrations of heptane. A third possibility is that the liquid composition somehow remained frozen during burning. In this unlikely event both components would again be evaporating simultaneously, with the burning rate being indicative of some average value of the mixture components.

Microexplosions were not observed for the mixtures studied (cf, Fig. 2). This fact is consistent with prior observations of burning heptane/hexadecane mixture droplets, though in a forced convective environment, which nevertheless used a similar method

of droplet generation.[16] It was conjectured in Ref. 16 that the method of generating the droplets can influence the occurance of disruptive burning. For example, internal liquid motion may be created as the droplet if formed which could influence the initial concentration field within the droplet at the start of burning. During the droplet formation process, deformation of the liquid, whether created by a Raleigh-Taylor breakup mechanism of a liquid ligament, or shearing a jet by mechanically chopping it, could induce internal liquid motion. The nature of the initial concentration field created by this motion could depend on the manner of liquid deformation during the formation process. The concentration field in turn effects the propensity for achieving the necessary internal superheat conditions required to trigger disruptive burning. Certain methods of generating droplets might then be more favorable for inducing microexplosions than others. For example, microexplosions have been observed for droplets which are mixtures of heptane and hexadecane[17] when the droplets were generated by mechanically chopping a liquid jet.

One mechanism for extinction which could have been operative in the present experiments is quenching of the flame by the electrodes. Such quenching could occur if the droplet drifted close enough to the electrode for it to act as a heat sink. However, the propensity for extinction did not appear to be due to the proximity of the droplet to either of the electrodes as evidenced by observations made from droplets which were either close to an electrode or positioned nearly at the midplane of the distance between the electrodes throughout their combustion history.

Another mechanism for extinction assumes a finite reaction rate at the flame front (as opposed to the infinite rate typically considered in droplet burning analyses). The attendant "leakage" of fuel through the flame would then lower the flame temperature because of the incomplete combustion at the flame front with the result that the flame is not sustained. This model leads to a relation between the droplet diameter at extinction and burning rate in the following qualitative form:[22] $d_E \sim 1/K$. An absence of extinction for the mixtures studied could have been due to a comparatively higher burning rate, and hence lower extinction diameter, than for the mixture components, but this is just conjecture. If the extinction diameter is smaller than what can be photographically resolved, a droplet would not be visible. The small droplet diameters that apparantly characterized extinction in air for the present experiments approached the limit of our measuring capabilities. This problem could be alleviated by varying the oxygen concentration. In addition, because the burning rate constant may be interpreted in different ways for droplet diameter data which exhibit an initial heating period (or also some scat-

ter) as noted above, quantitative comparisons between measured and predicted extinction diameters would not be unambiguous.

Conclusions

The low gravity combustion of unsupported heptane/hexadecane mixture droplets has shown, within the precision of the measurements made, no discernable change in the gradient of the temporal variation of diameter squared. Such a change would have indicated a preferential vaporization mechanism. The evidence suggests that droplet burning was diffusion controlled. In addition, droplet heating was more pronounced for the mixtures than its' components even with the relatively small droplet sizes studied (<500 μm initial diameter). The absence of microexplosions for the mixtures studied is supportive of an influence of the method of droplet generation on the disruptive burning potential of multicomponent fuel droplets.

An extinction-like phenomenon was thought to be revealed in the present measurements by a period at the end of burning beyond which the droplet diameter did not further decrease. Such a phenomenon was observed for the mixture components, but not for the mixtures themselves.

Acknowledgments

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COMMENTS

M. R. Zachariah, National Bureau of Standards, USA. Could you comment on the relative importance of droplet motion toward or away from the imaging system with regard to the measured droplet size.

Author's Reply. Droplet motion toward or away from the camera was easily detected by blurring of the droplet image in the photographic record (the depth of field was less than 1 mm). We rejected any movie sequences in which blurring of the droplet image had occurred. Only those sequences in which the droplet image remained in focus throughout burning were analyzed. An example of such an essentially two-dimensional motion is shown in Fig. 2 of the paper wherein the droplet is in focus throughout burning.

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B. N. Raghunandan, Indian Institute of Science, Bangalore, India. I notice that you have compared your d^2 data with the theoretical predictions. In fact, many theories of single droplet combustion do show good agreement with the microgravity data. What is more critical is the prediction of flame stand-off ratio (d_f/d_s). Some discrepancies have remained unresolved in this direction. Do you have d_f-data for the complete life-time of the droplet? And how long during the process of combustion is the quasi-steady approximation valid as seen in your experiment.

Author's Reply. The authors agree that the evolution of droplet diameter alone does not determine the extent to which the quasi-steady approximation is valid. It is well-known that the classical quasi-steady approximation is valid only if d^2 varies linear with time *and* (d_f/d_s) is constant. This point is explicitly stated in the paper. We reported our droplet diameter data in the conventional way, d^2 vs. t, but we do not state anywhere in the paper, nor do we imply that quasi-steady conditions are necessarily reached only because d^2 varies linearly with time. The added information needed to determine whether or not quasi-steady conditions are reached is the evolution of d_f/d_s.

Preliminary flame diameter measurements of toluene droplets (not reported in this paper) show that (d_f/d_s) increases with time throughout the entire period of burning.