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I. INTRODUCTION

The use of liquid monopropellants in a regenerative gun involves a complex process of high-speed bulk injection and combustion of the propellant. Unfortunately, little is known about the extent of liquid breakup into drops before combustion takes place. However, droplets are probably important in both the ignition and propagation of flame to any bulk material. In addition, for sub critical studies the surface tension of individual droplets provides a well defined liquid-gas interface for the detailed study of the gas evolution from the liquids and flame zone structure above the liquid surface. Such studies are difficult with bulk liquids.

Earlier studies of droplet heating and ignition have used a variety of techniques. Typical examples include moving electrically heated furnaces to enclose suspended drops^{1,2} at atmospheric pressure and freely falling highly reproducible drops at slightly elevated pressures.³ Others have extended combustion measurements up to 2000 psia (14 MPa) with direct ignition of drops⁴ or drops simulated by liquid on porous spheres.⁵

In our previous studies,⁶ liquid propellant drops have been studied in hot flows at atmospheric pressure, in a manner similar to Law and co-workers.³ This paper reports our efforts to extend these measurements to gun ignition pressures. The goal is to measure parameters such as the delay to ignition and burning rate as well as studying the details of the ignition and combustion processes.

Two techniques have been explored in this effort. The first was pressurization and heating by the combustion of mixtures of gases in a closed vessel. However, this approach proved to be unsatisfactory because the motion of the gases in the chamber was both greater and longer lasting than expected. Although these observations provided preliminary high pressure experience with single drops, they were inadequate because of poor reproducibility; details may be found elsewhere.⁷ The second approach was to use approximately constant pressure with rapid and controlled heat input to the drop. The use of steady pressurization also increased the safe operation of the vessel by a factor of two.

II. EXPERIMENTAL

For these observations, a windowed vessel is pre-pressurized. Then, drop heating is achieved by local resistance heating of a small volume inside the vessel, opening an electromagnetic damper (shutter), and convecting the hot gas over a suspended drop with a small (ca. 30 psi or 0.21 MPa) increase in total gas pressure. For all of the observations reported here the gas was nitrogen. Flow velocities have not been measured but are estimated from the motion of thermal disturbances to be on the order of 10 cm/sec. The Reynolds numbers are on the order of ten or less. Timing was implemented such that the heater and recording devices were turned on for an appropriate time before the damper was opened and the gas pressure was increased. This technique has been used here to study drop behavior over the range from 150 to 1250 psi (1.0 to 8.6 MPa).

The overall schematic diagram of the apparatus is shown in Figure 1. The windowed vessel for these studies had a cylindrical interior with a 4.4 cm

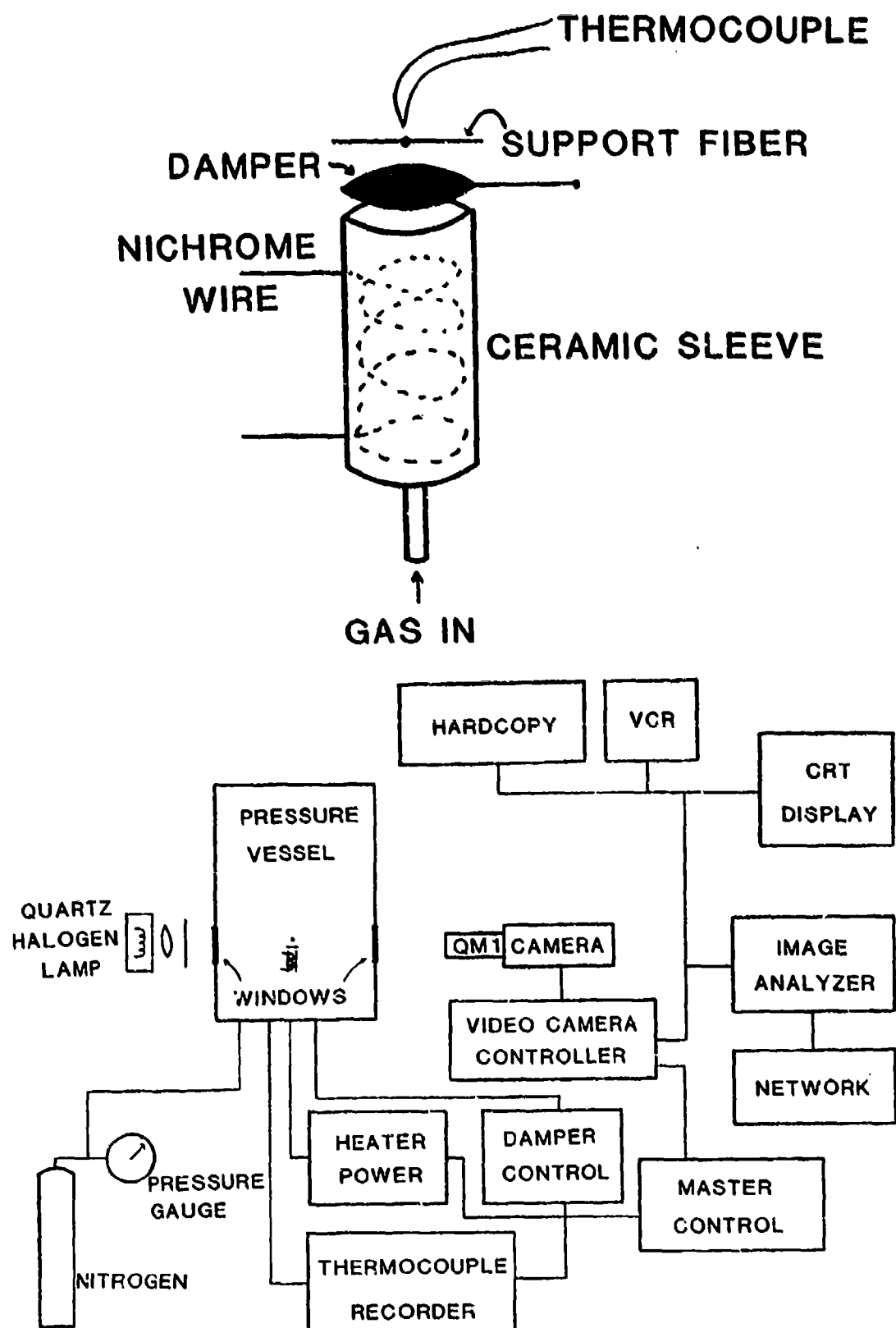


Figure 1. Schematic Diagram of Constant Pressure Apparatus Showing (A) The Details of the Heating Device and (B) The Overall Configuration

diameter; total volume was approximately 230 cm^3 . Events were imaged through a 90 mm aperture 2800 mm focal length lens. Typical image area was a few millimeters square. Strong back lighting was used. Some studies were recorded with a 16 mm high speed camera. However, for the majority of the work reported here, the imaging was done with a high speed video system (Spin Physics Model SP2000). In addition to the obvious advantages of rapid turnaround, this system was coupled to an 16-bit computer based video frame grabber. Image processing was used to delineate the edges of the drops. These data were then fitted to circles or ellipses by a VAX 11/780 to provide the potential for accurate drop diameter measurements.

Temperature was measured with a 50 μm chromel-alumel thermocouple mounted within a few drop diameters downstream from and out of the wake of the drop. The resulting signal was recorded with typically 5 msec resolution by a transient recorder. By recording temperatures in flows without drops, modifications were made to minimize turbulence and other effects as indicated by temperature fluctuations. At near atmospheric pressures the temperature profiles were almost ideal with a rapid rise and smooth plateau. In contrast, a typical record from a 300 psi (2.1 MPa) run is shown in Figure 2. As can be seen, the flow degrades at higher pressures.

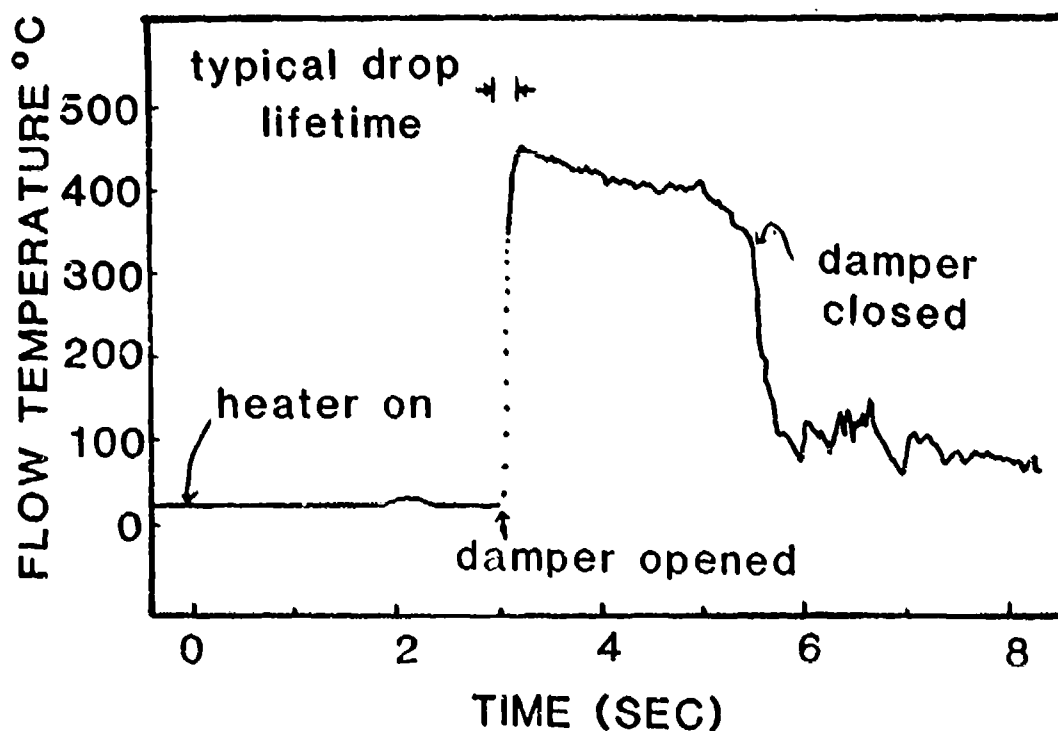


Figure 2. Typical Temperature Record Showing Rapid Rise and Fall With Damper Action

In order to get reproducible drop sizes, a piezoelectric drop generator was used to produce drops. One drop was caught on a 50 μm fiber and transferred to the holder in the vessel. The drop holders were fused silica fibers with diameters from 50 to about 15 μm . Diameters below 25 μm allowed even the smaller drops to be quite spherical.

The liquid used in all of these observations was the liquid monopropellant designated LCP 1846. This material is a stoichiometric mixture of 61% (by weight) hydroxylammonium nitrate (HAN), 19% triethanolammonium nitrate (TEAN), and 20% water. Much recent work has been done to fully characterize the physical properties of this class of materials.⁸ Briefly, the density is 1.43 g/ml at 25°C. The boiling point is estimated at 123.7°C; its value cannot be easily measured because of the onset of exothermic reactions of the HAN near 120°C. The surface tension is not substantially different from that of water. The vapor pressure has been measured up to 65°C to be less than half that of water, probably due to a well organized and extensive network of hydrogen bonding in the liquid. The viscosity behavior is typical of a molten salt. Viscosity values of 5.47 cst (kinematic) and 7.85 cp (dynamic) have been measured at 25°C. The critical point of this liquid is unknown; it is expected to be outside the range of the present observations. This material does not have a well defined ignition point. Rather, slow heating under laboratory conditions results in vaporization of the water (if in a dry environment), reaction of the HAN near 120°C, and finally reaction of the TEAN at higher temperatures. The nature of changes in this process when heated rapidly and in a combustion environment filled with water vapor and other combustion products is a goal of the present study.

III. OBSERVATIONS

With this configuration, the experimental variables were much more controllable and reproducible than in our combustion-heated studies, but a strong element of randomness remained. The response of the drops to heating can be placed in three classes. The first mode was to change opacity after perhaps 100 msec, and then decrease in diameter while undergoing some shape distortions probably related to internal chemistry. The circular drop images from a typical record of this type were computer fit and the results plotted in Figure 3. Typical uncertainty from the fitting routine is less than the plotting symbols used. The zero of time in Figure 3 is the point at which heat flow can be seen in the video record. These data should not be compared to a fuel drop in an oxidizing flow. In this case there is clearly in-depth internal chemistry rather than simply heating and vaporization/pyrolysis followed by gas phase reactions.

The other two modes were related but probably distinct phenomena. In both cases the drops remained essentially constant in diameter with an occasional wisp of vapor trailing off, changed their light transmission characteristics at some later point, and then in times on the order of one millisecond would vaporize. The distinction in the two modes was that the gas phase was translucent in some cases, while in others it was opaque. There was no attempt made to study the final event in any more detail because the earlier changes are a clear indication of internal chemistry which is probably dominant. In all three of these modes of pyrolysis, internal gas bubbles of varying size were sometimes observed.

Under these circumstances, the only drop parameter that was clearly unambiguous was the lifetime of the drop, defined as the time from heat flow at the drop position to drop disappearance. Although there is still considerable scatter in even this very basic parameter, there are at least correct trends in most cases. Figure 4 shows the lifetimes of a series of 240 to 280 μm drops as a function of final flow temperature at 500 psi (3.5 MPa).

Considering the variability of physical appearance of the drops, the trend is surprisingly smooth. The effect of the drop holder on this measurement, as the liquid nearer the holder was consumed, was generally not considered important because of the accelerating rate of drop disappearance with time.

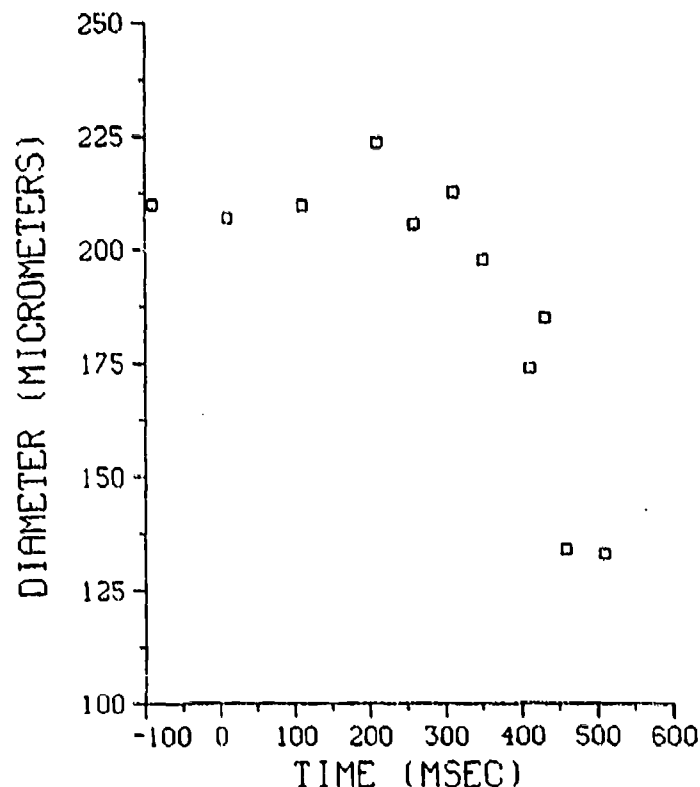


Figure 3. Diameter History of 210 Micrometer Diameter Drop of LGP 1846 in 460°C Flow at 150 psi (1.0 MPa)

Figure 5 shows the observed lifetimes from several observations as a function of pressure at final flow temperatures around 450°C. The amount of apparent scatter is exacerbated by the multiple drop sizes on one plot. However, the 240-280 μm data, acquired as a systematic set, show a good trend with reasonable scatter except for the point at 505 psi (3.5 MPa). Overall, these data suggest that there is an upper limit for the lifetime under given conditions, but that uncontrolled factors are decreasing the values some of the time.

Although microexplosions which destroyed the drops were still observed under conditions where heating was less than maximum and longer times occurred before drop reaction, they most frequently were not present for drops in the 40 to 280 μm diameter range in this experiment. However, major gas bubble formation was not uncommon. When microexplosions were observed, they differed in appearance from those observed at ambient pressure in that the entire drop would appear to be involved in liquid phase reactions. Rather than breaking the drop into a shower of smaller droplets, at these pressures the drop would balloon up and become a cloud of gases. These observations are well below the predicted critical pressure of the original material. Thus it is likely that

the observed behavior is due to liquid phase reactions, either from rapid decomposition and gasification of the drop or from reactions which result in a liquid which is above its critical point. Representative behavior is shown in Figure 6, with the final frames from heating a 185 μm drop in a flow at approximately 300°C. The drop has already swelled slightly in the first image shown; in the last one only a dark cloud remains. These data were recorded at 1000 frames/sec.

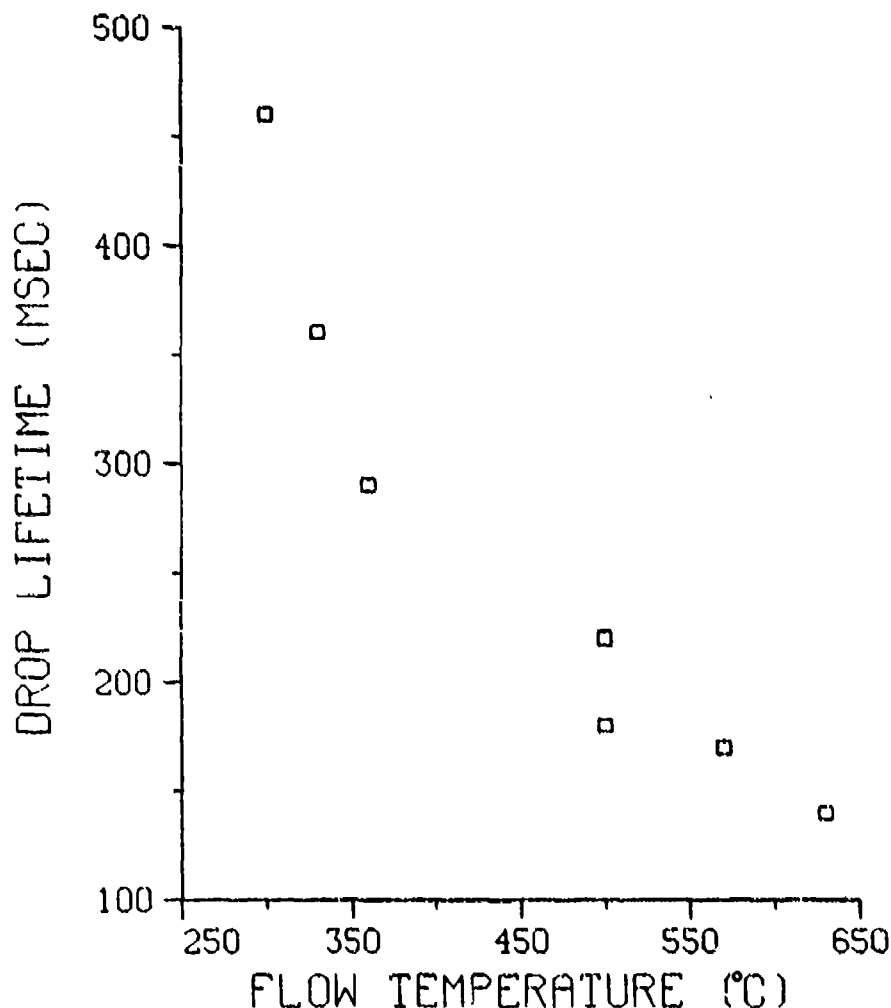


Figure 4. Lifetimes of 240-280 Micrometer Diameter Drops of LGP 1846 as a Function of Flow Temperature at 500 psi (3.4 MPa)

The observed behavior was obviously affected by the presence of the holder; bubbles large or small were generally formed away from the fiber. In order to minimize the fiber influence a 390 μm diameter drop was mounted on a 20 μm fiber and subjected to a flow of 480°C at 500 psi (3.4 MPa). This drop size is near the limit of drop diameter for holding onto the fiber reliably in the flowing gas. The drop images were digitized and fit by computer. The resulting history is shown in Figure 7. In this figure the scatter, especially at early times is much greater than the computer uncertainty in the fit. The diameter uncertainty as indicated by the scatter for about the first

200 msec is probably due to thermal lensing from the non-laminar flow, although the image appears motionless. If it is assumed that the flow is finally well established around 220 msec, then the drop diameter shows a smooth increase, possibly due to drop expansion as it heats up. At 240 msec the drop turns opaque in the time of one frame (0.5 msec), without any change in the diameter. This sudden opacity is typical of drop behavior under the conditions of all of the observations reported here. At later time the drop balloons symmetrically, but on a somewhat slower time scale than at atmospheric pressure. During ballooning the drop also moves somewhat on the fiber. In the next video frame after the last plotted point, only a cloud of vapor remains.

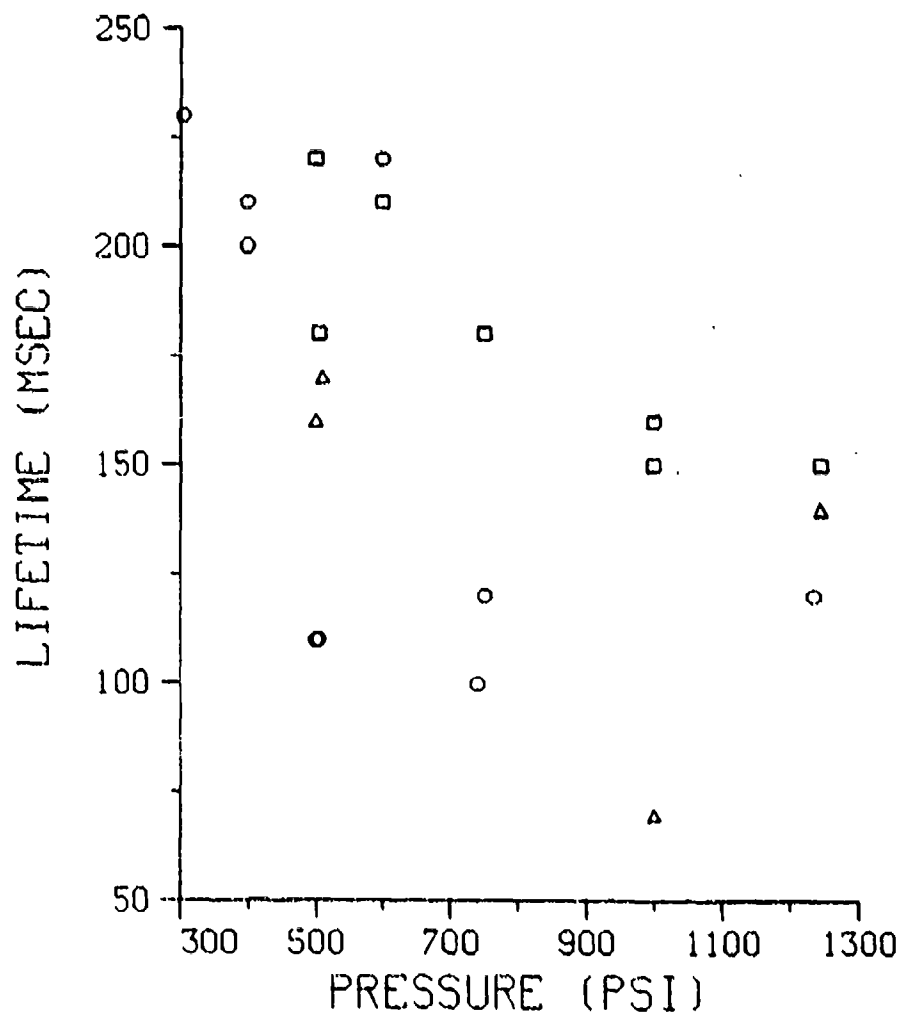


Figure 5. Lifetimes of LCP 1846 Drops as a Function of Pressure in 400-500°C Flows for Drop Diameters 240-280 [□], 160-210 [○], and 80-85 [△] micrometers



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Figure 6. Final Frames from Heating a 185 μm Drop Showing Internal Bubble Formation and Drop Gasification

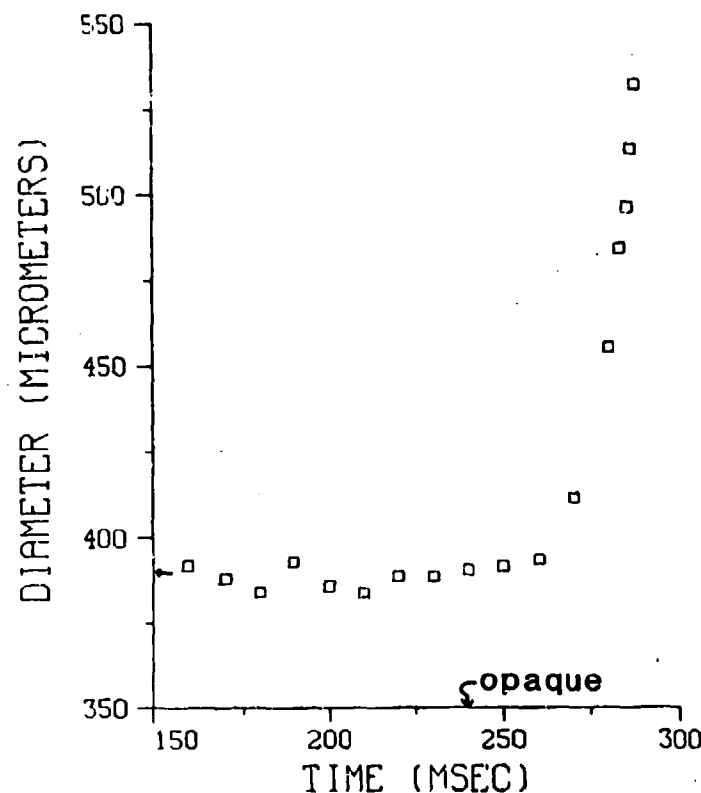


Figure 7. Diameter History of a 390 Micrometer Diameter Drop of LGP 1846 in a 480°C Flow at 500 psi (3.4 MPa)

IV. DISCUSSION

The preliminary studies discussed above show that there is promise in this relatively simple apparatus for understanding the behavior of LGP drops during at least the ignition phase of a regenerative gun cycle. Since internal gas bubble formation is so common with these drops, understanding the effect of the fiber on the drop it holds is important. As gas bubbles generally form away from it, the fiber may be either inert or possibly have some cooling effect. It has been noted here that "drops" with radii near the size of the fiber diameter behave in a manner not dramatically different from larger drops as far as following trends with respect to lifetime of the drop. For larger drops the internal gas bubbles coalesce and expand the drops more symmetrically, in the manner previously observed in free drops.⁷

The cause of the scatter in the lifetimes shown in Figure 5 is undetermined at present. Fluctuations have been observed in the flow temperature measurements, interpreted as resulting from a less than ideal flow and the mixing of cooler air. However, it is be noted that five of the six points plotted in Figure 5 in the 240 to 280 μm range, plotted as squares, show an extremely smooth trend with pressure. These data were taken in a sequence where every effort was made to duplicate all conditions except pressure. The temperature profiles for those drops were similar to all others. Further studies are required to determine what the uncontrolled variables are and to measure their influence on drop behavior.

It is likely that at atmospheric pressure the driving mechanism of microexplosions is the homonucleation of superheated water.⁹ However, the strong visual evidence, including the vapor trails from the surface, the loss of transmission through the drops, and the common conversion of the drop into a cloud of vapor, suggests substantial liquid phase chemistry. It has been established earlier¹⁰ that nitrogen oxides are evolved early in the reactions of these materials. Thus the evidence of liquid phase reactions suggests that the bubble formation in the drops of LGP 1846 at higher pressure may be driven by gas generated from chemical reactions. However, the slightly different behavior at these higher pressures might also be a manifestation of the presence of the fiber. The time scales of the two experiments is not greatly different. The portion of the drop near the fiber may be cooler than the rest of the drop if exothermic reactions are increasing the internal temperature in addition to the conduction. More data are clearly needed at increased temperatures and at higher pressures. Observations with freely falling drops might also provide more reproducibility.

V. FUTURE STUDIES

The work presented here is preliminary but provides the foundation for advancements in several ways. Near term plans include the addition of spectroscopic diagnostics to identify the gas phase products which evolve from the drop, especially during early heating. Spectroscopy of the liquid phase is also being explored. Higher pressures and slightly higher temperatures are attainable without major modification of the apparatus. After the ignition point of these LGPs is reached, studies of the flame structure will be pursued using visualization and spectroscopic techniques. In addition, major parameters causing scatter in the present studies will be identified. An effort is also being made to apply acoustic levitation of drops to this pressurized environment.

VI. CONCLUSION

A high pressure, high temperature experimental facility has been designed and fabricated for the study of liquid gun monopropellants. Preliminary observations show qualitatively correct behavior to variations in pressure, temperature, and drop size. The advantages of steady pressurization are great and should be incorporated in any future studies on the individual droplet level.

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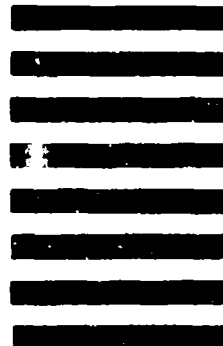


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