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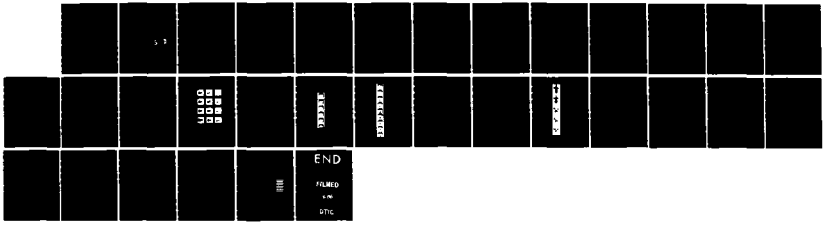

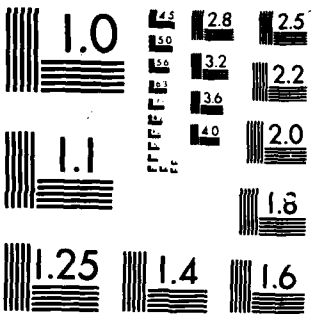

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OPTICAL STUDIES OF ELECTRICAL DISCHARGE-LIQUID PROPELLANT INTERACTIONS

Fred B. Carleton
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Nathan Klein
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January 1986

US ARMY BALLISTIC RESEARCH LABORATORY
ABERDEEN PROVING GROUND, MARYLAND

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

In the course of investigating the ignition of liquid propellants by electrically initiated plasma discharges, it became clear that a number of complex and important processes were taking place within the reaction chamber, commonly called a "plasma plug." This report describes the development of transparent plugs that permit optical access to the reaction chamber, and

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20. ABSTRACT (CON'T)

Optical and electronic systems for high speed cine schlieren, shadow, and direct photography of events taking place within the cavity. A number of observations are presented and briefly discussed that demonstrate the power and versatility of the equipment. Keywords:

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I. INTRODUCTION

The use of an electrically initiated plasma discharge to ignite liquid propellants has been under investigation for several years[1] and has been shown to be both an effective and reproducible ignition technique and a promising method for diagnostic investigation of ignition phenomena. In previous studies, such parameters as geometry of the ignition chamber, size and shape of the venting orifice, location of the electrodes in the chamber relative to the propellant, and the amplitude and shape of the current-voltage pulse that produces the plasma arc, were systematically investigated. Data consisted of voltage and current measurements taken at the ignition chamber and high-speed shadow and schlieren cine photography of the material emerging from the chamber vent. In all of these studies and in the work that is reported herein, the propellants used consist of a mixture of hydroxylammonium nitrate (HAN), a 4-carbon aliphatic ammonium nitrate (C-4AAN), and water in N_2, CO_2, stoichiometric ratio and containing 11 mole/liter nitrate ion.[2]

Since the body of the chamber, commonly called a "plasma plug", was fabricated of opaque materials such as ANSI 316 stainless steel and MACOR,* a machinable glass ceramic, no direct observations of electrode-propellant interaction were possible and such information could only be inferred from the data obtained. Accordingly, a propellant chamber was devised that would permit observation of events taking place within it, and this was incorporated into the design of a diagnostic system capable of time-resolved recording of such events. The development of this device and some illustrative results of its use are the subject of this report.

II. EXPERIMENTAL DESIGN

The ability to obtain the data desired will depend on an adequate design of the three major portions of the experimental components: the propellant chamber, the optical system, and the electronics and recording system needed to produce the timing sequences used in acquiring and recording the data. Each will be discussed briefly.

A. The Propellant Chamber

The propellant chamber was designed to permit controlled variation of a number of the parameters known from past work to affect the response of the propellant to a pulsed electrical stimulus. Such parameters include, but

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[1] N. Klein, F.B. Carleton, and F.J. Weinberg, "Methods for Evaluation of Ignition Stimuli for Liquid Propellants," Proc. 19th JANNAF Combustion Meeting, Vol I, p 505, 1982, CPIA Pub 366.

[2] N. Klein, "Preparation and Characterization of Several Liquid Propellants," ARBRL-TR-02471, Feb 1983, US Army Ballistic Research Laboratories, Aberdeen Proving Ground, MD.

*Corning Glass Works, Corning, NY

are not limited to, placement and geometry of the electrodes, and the size and shape of the vent orifice. As in our previous work, small cavity volumes (30-45 μl) were used. Safety considerations required that propellant volume be small enough that quantities of gas resulting from combustion remain within a safe operating range. As a result, the electrical contribution to the total energy will not be negligible as compared with the chemical potential. Since the primary purpose of the work reported herein was direct observations of the events taking place within the chamber rather than plug design for maximum efficiency, the coaxial electrode system used previously was replaced by an approximately two-dimensional geometry.

The corrosivity and general reactivity of the propellants restrict the selection of materials that may be used in the construction of the reaction chamber and electrodes. Polytetrafluoroethylene (PTFE), glass, and either platinum or tantalum were to be the only materials that would come in contact with propellant or its decomposition products since these materials are known not to interfere with the processes to be studied. The reaction chamber consisted of a PTFE plate, 1.5 mm thick, in which were cut a cavity to hold the propellant sample, a vent, and the electrode channels. Because PTFE deforms due to cold flow, no serious effort need be expended in attempting to machine surfaces and orifices to closely held tolerances. The chamber vent was, therefore, a simple gap, typically 1 mm, forming a rectangular orifice with the plane windows. The electrodes were a pair of platinum wires, 0.4 mm in diameter. In addition to discharges between simple wire terminals protruding through the walls, discharges between an axial electrode at the base and the orifice (to simulate the previously used cylindrical geometry), and between wire electrodes running the length of the walls of a trapezoidal cavity (to encourage current flow at the smallest gap) were among the geometries studied.

The body of the reaction chamber was formed by a pair of glass microscope slides held together by several, small C-clamps. Cleaning and filling was accomplished using hypodermic syringes. The two windows of the assembled reaction chamber act as components of an interferometer and their parallelism and alignment within the optical system could be judged very accurately by minimizing the interference pattern created. Typical propellant chamber geometries are shown in Figure 1.

The reaction chamber electrode power supply and the current and voltage recording system are identical to that previously described[3] and were used without modification. Current measurements, obtained using a differential amplifier recording across a 3.22 ohm resistor in series in the electrode supply circuit gave results identical to those obtained with a Pearson* Model 410 Wide Band Current Transformer, the comparison serving as a verification of the accuracy of the current measurement technique.

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[3]N. Klein, F.J. Weinberg, and F.B. Carleton, "Ignition Phenomena in Energetic Liquids," ARBRL-TR-02514, Aug 1983, US Army Ballistic Research Laboratories, Aberdeen Proving Ground, MD.

*Pearson Electronics, Inc., Palo Alto, CA.

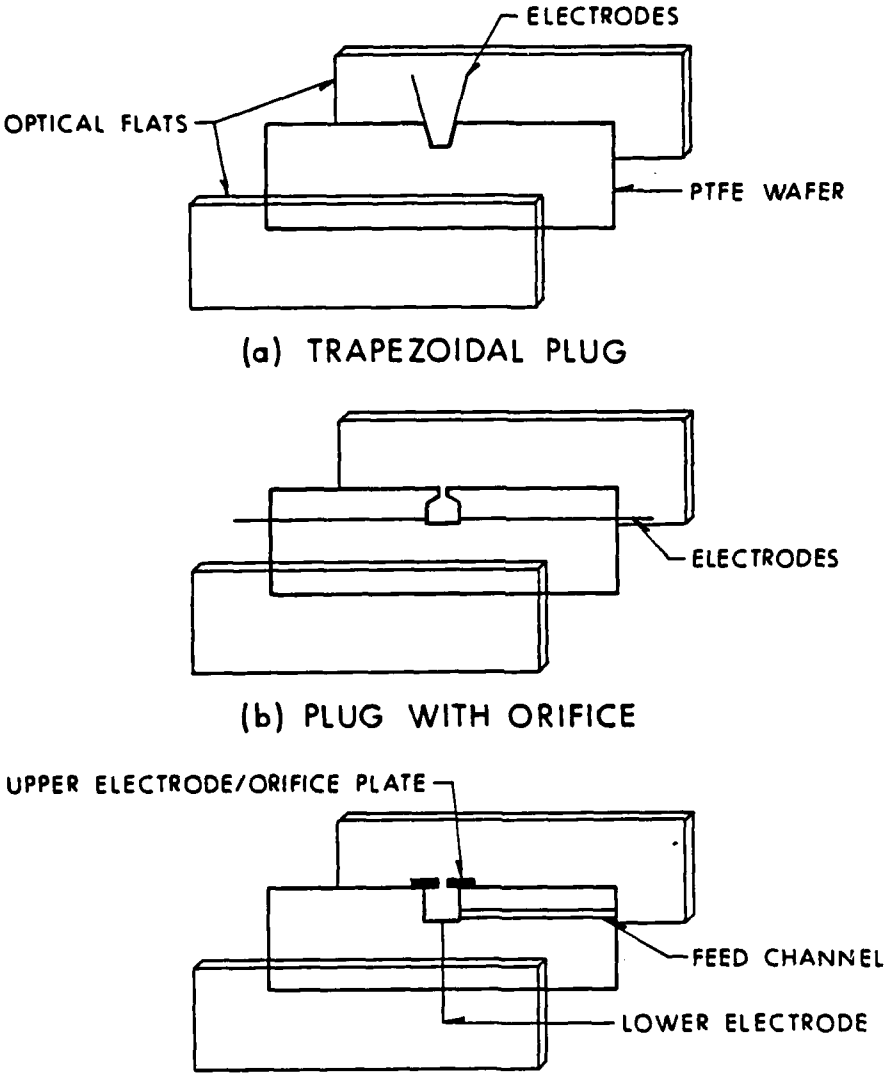

Figure 1. Exploded Views of Various Transparent Cavity Plasma Plug Designs

B. The Optical System

The optical components of the shadow-schlieren system[4] used will be briefly described. The light source was a 4W argon-ion laser, operating continuous wave at 488 nm, followed by a spatial filter, optics for producing and focusing a parallel light beam, knife-edge, and finally, the optical components for projecting a focused image onto the receptor. For shadow recording the knife-edge is removed and the scene is recorded out of focus. The use of monochromatic light simplifies analysis because refractive index is wavelength dependent and the images to be obtained derive from variation in light refraction. For the same reason it also permits the use of simple lenses in the optical system without giving rise to chromatic aberration. It is, however, not an unmixed blessing[5] in schlieren/shadow systems because of the diffraction effects it causes in the vicinity of sharp boundaries. These were not so extreme in the present instance as to call for the corrective procedures discussed in reference 5.

Spatial filtration to obtain uniform beam intensity was achieved by focusing the beam through a 50 μm pin-hole. The expanding beam emerging from the spatial filter was collected by a spherical mirror of 240 cm focal length to give a parallel beam approximately 10 cm in diameter. This transluminated the test space containing the propellant chamber. The emerging beam was refocused in the vicinity of the knife edge (when used) and projected onto photographic film. The required intensity of the light source is dictated by the sensitivity of the film at 488 nm. The duration of the exposure is a matter of some complexity for exposure times signif-. icantly shorter than those employed in conventional photography, since published values for film response (ISO ratings) often do not apply under these conditions. Previous work made use of a rotating prism, high speed cine camera that could accurately record events up to speeds of about 20,000 frames per second. The approximately 50 μs time resolution for this system was not fast enough to record events within the reaction chamber. Judging from current-voltage records and blur in some of the photographs, the characteristic time for events within the plasma plug were in the range of 5-20 μs.

Since rotating prism, moving film, cameras are not adequate for data recording at the time resolution required, a rotating hexagonal mirror camera was selected. From past use[6] this type of camera was shown to be capable of the required time resolution. This camera utilizes a stationary 1 m length of 35 mm film and requires a repetitively pulsed light source. A method for proper sequencing of frames is required, since no more than 100 images can readily be recorded on the 1 m length of film without overlap. The 4W argon-ion laser, a Spectra-Physics* Model 164, was fitted out with a

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[4]F.J. Weinberg, Optics of Flames, Butterworths, London, 1963.

[5]A.K. Oppenheim, P.A. Urtiew, and F.J. Weinberg, Proc. Royal Soc. Lond. A Vol. 291, p 279, 1966.

[6]F.J. Weinberg and J.R. Wilson, Proc. Royal Soc. Lond. A, Vol. 321, p 41, 1971.

Model 344 Cavity Dumper that includes the Model 451 Power Supply and Model 346 Stabilizer, so that a train of light pulses, preselected in duration, intensity, and repetition rate would be available for use. Energy is supplied to the Bragg crystal in the resonant cavity such that a nominal 14 ns light pulse can be obtained over a broad frequency range. In addition, an external mixer input permits the production of light pulses longer than 14 ns. The accessory does not interfere with continuous wave operation of the laser so that rotating prism camera imaging is unaffected by this modification.

Beam intensity requirements are determined by image magnification, pulse duration, and the photometric sensitivity of the recording medium, which in this case was a fine-grained, black-and-white, photographic film such as Ilford* HP-5. The light source described was quite adequate for 0.5 μs film exposures. The camera mirror was rotated slowly enough for the pulse train, whose duration was as long as 1 ms, not to multiply expose any portion of the film. Used in this manner, the optical system is capable of recording images at framing rates of from 25,000 to 500,000 frames per second. The optical components are shown in Figure 2.

C. Electronics

The system described consists of a number of separate components all of which must function in a coordinated and proper sequence in order to obtain the data desired. The rotating mirror camera contains an electric motor capable of 75,000 revolutions per minute, the speed of which is controlled by a variable voltage transformer. Although the speed obtained at a given transformer setting is quite reproducible, the time required to achieve such speed is fairly long, typically 20-30 seconds. The long and somewhat variable time required to bring the camera to operating speed mandated that the camera be the system component that would initiate the experimental sequence. The active components of the system consisted of a power supply that delivered the electrical energy needed for the plasma arc discharge, a storage oscilloscope that recorded the discharge current and voltage data, and the laser cavity dumper. All of these components had external triggering capability although the individual trigger signals needed were quite different from one another.

The camera was equipped with a mechanical shutter that, when opened, enabled a -5 V signal to be delivered to the external trigger input of a Venner** Model TSA628 Pulse Generator. This signal was routed through a synchronization unit that coordinated the trigger signal train with a particular position of the rotating, hexagonal mirror in the camera via an infrared light emitting diode and photodiode pair. The synchronization unit was needed to achieve position reproducibility in the film records since operation of the mechanical shutter is manual and totally independent of other, needed conditions. After a delay of 15 μs, the Model TSA628

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*Ilford Ltd., Basildon, Essex, England.

**Venner Electronics, Ltd., New Maldin, Surrey, England.

generator produced a pulse 1.0 ms in duration with an amplitude of +5 V which is delivered to the start pulse input of a Farnell* Modular Pulse Generating System. The 15 μs delay was chosen to ensure that any variation in operation of the mechanical shutter would not affect operation of the remainder of the system. Pulse duration was kept sufficiently long to ensure that the only trigger signal supplied was the positive, leading edge of the pulse since it had been observed that the Modular Pulse Generator could also be triggered by the descending step that ended the pulse. This generator was set to deliver 90 pulses at a 100 kHz repetition rate and is equipped with 2 output channels. The first delivered a pulse 100 ms long and -5 V in amplitude to the external trigger input of the propellant chamber electrode power supply after a 30 μs delay. The synchronous output of this channel is connected to the external trigger input of a Tektronix** Model 7613 Storage Oscilloscope equipped with a Model 7A18 Amplifier, Model 7A22 Differential Amplifier, and Model 7B53A Time Base. The 30 μs delay permitted the oscilloscope, triggered by the prompt synchronous output, to record and display the baseline of both of its channels before the arrival of current and voltage data. The long trigger pulse delivered to the electrode power supply ensures that the voltage and current data recorded are controlled exclusively by events taking place within the propellant chamber. Any perturbation caused by external electrical signals will occur long after event recording has been completed. The second channel, after a 1 μs delay, delivered 90 pulses at 10 μs intervals, each 0.5 μs wide and +1.5 V in amplitude to the external mixer input of the laser cavity dumper thus creating the string of light pulses to be recorded by the camera. Two light pulses should precede the delivery of current to the propellant chamber, so as to ensure a record of the earliest current-propellant interaction. All signals were carried by 50 ohm coaxial cable in order to minimize reflections and distortion of pulse shape. A block diagram of the electronics array is shown in Figure 3.

The electronic signal sequence as described produces a camera framing rate of 100,000 frames per second, the framing rate being established by the frequency selected for the programmable pulse generator. Exposure time is established by the duration of the pulse delivered to the cavity dumper, 0.5 μs in the example cited. This allows reasonable freedom in selecting framing rate and exposure time independently. There are, however, limitations, the most important of which is that the duty cycle of the cavity dumper shall not exceed 50%. Thus, for example, a 1 μs exposure limits the maximum framing rate to 500,000 frames per second. Exposure time is constrained by the sensitivity of the film at short times and by the amount of light energy that can be stored in the laser resonant cavity at longer times. The intensity of the light pulse can not be made to duplicate the shape of the trigger pulse if the pulse is much longer than 1 μs, the intensity attenuation being due to depletion of energy stored in the cavity. This property of the laser resonant cavity means that a longer pulse does not result in a significant increase in light intensity. The use of film more sensitive than Ilford HP-5 is required if light pulses shorter than 0.5 μs are to be used.

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*Farnell Instruments Ltd., Sandbeck Way, Wetherby, West Yorkshire, England.

**Tektronix, Inc., Beaverton, OR.

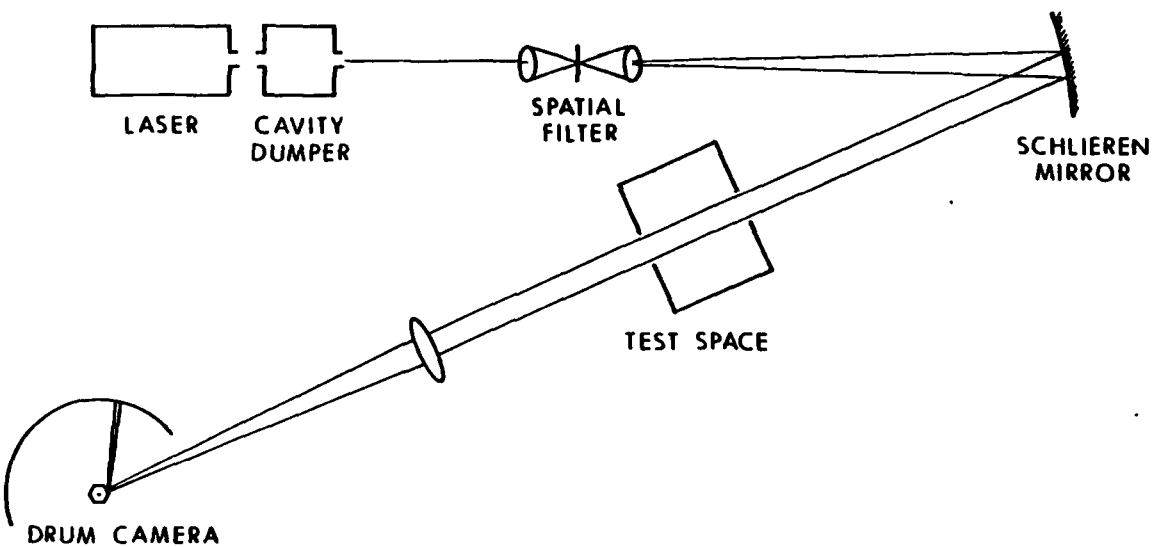

Figure 2. Optical System

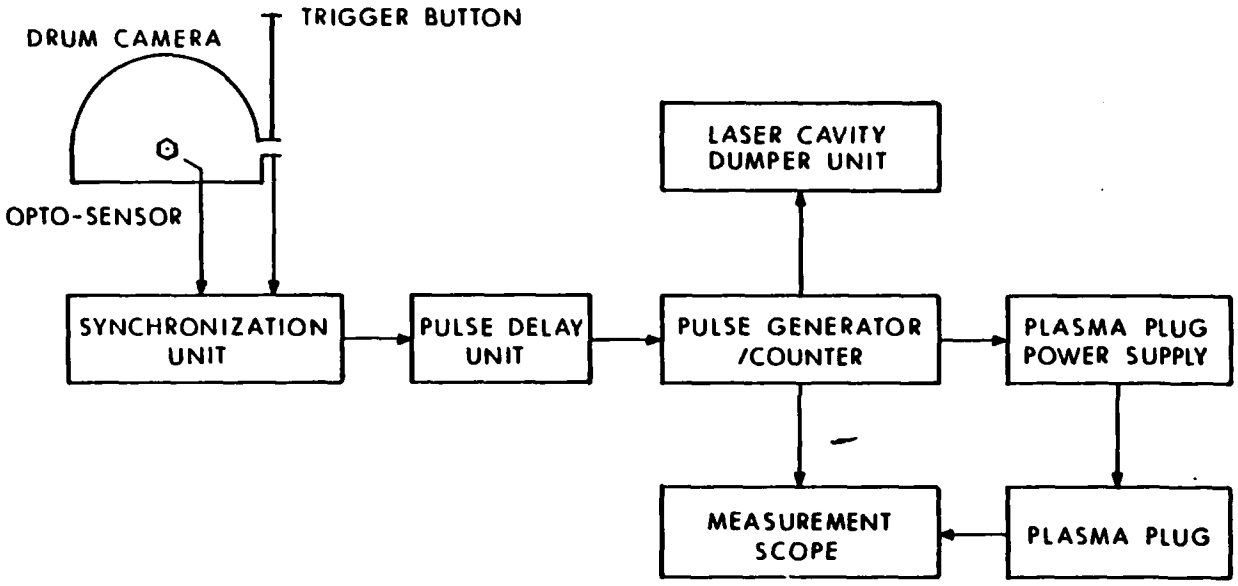

Figure 3. Block Diagram of Electronics Array

III. RESULTS AND DISCUSSION

Optical recording of data from these investigations ranges from qualitative visualization to quantitative measurements using self-luminosity of the test object, as well as a range of optical techniques based on external illumination by the laser system discussed above. Due to the wide scope of this investigation and the purpose of this report, which is to describe the development and use of transparent reaction chambers, results presented here are limited to illustrative examples.

In previous investigations[3] two distinct groups of venting plumes were observed. In one, the emerging material consisted primarily of hot, reacting gases with occasional droplets of liquid entrained in the expanding plume. In the other group, the emerging material consisted of a jet of unreacted propellant, followed by a spray of fine droplets, and ending in a small puff of hot, expanding gas. Characteristic current-voltage traces accompanied these two kinds of events, the first typical of a high-current, low-voltage arc discharge, and the second, characteristic of a low-current discharge, the original, high voltage being maintained throughout the discharge sequence.

Evaluation of the data led to the suggestion[7] that the group of observations involving a large plume and a high-current, low-voltage discharge were produced by an arc discharge generating a plasma near the top of the reaction chamber. Reaction of the propellant contained in the cavity is then initiated by the hot gases produced by the arc, resulting in partial burning, cigarette fashion, with the generation of gases and occasional droplets until the entire sample is consumed. The second type of event was thought to involve initiation of reaction, primarily by ohmic heating and electrolysis of the electrically conducting propellant, at or near the bottom of the reaction chamber. A small amount of hot, gaseous products resulting from these reactions expels the liquid contents of the chamber, thus producing the jet and droplet spray that, according to the schlieren photographs, are at or near room temperature.

The transparent plug allows the evolution of reactions within the plug including the development of luminosity to be followed in time and space. Figure 4 illustrates relevant sequences obtained with the trapezoidal plug (Figure 1A). This configuration was chosen for two reasons. The complete absence of a venting orifice eliminates its possible effect on reactions within the cavity and may thus simplify data interpretation, and secondly, the electrode structure was designed to encourage current flow at the lower part of the chamber (the smallest gap).

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[7] N. Klein, "Liquid Propellant Ignition Studies," Proc. 20th JANNAF Combustion Meeting, Vol I, p 473, 1983, CPIA Pub 383.

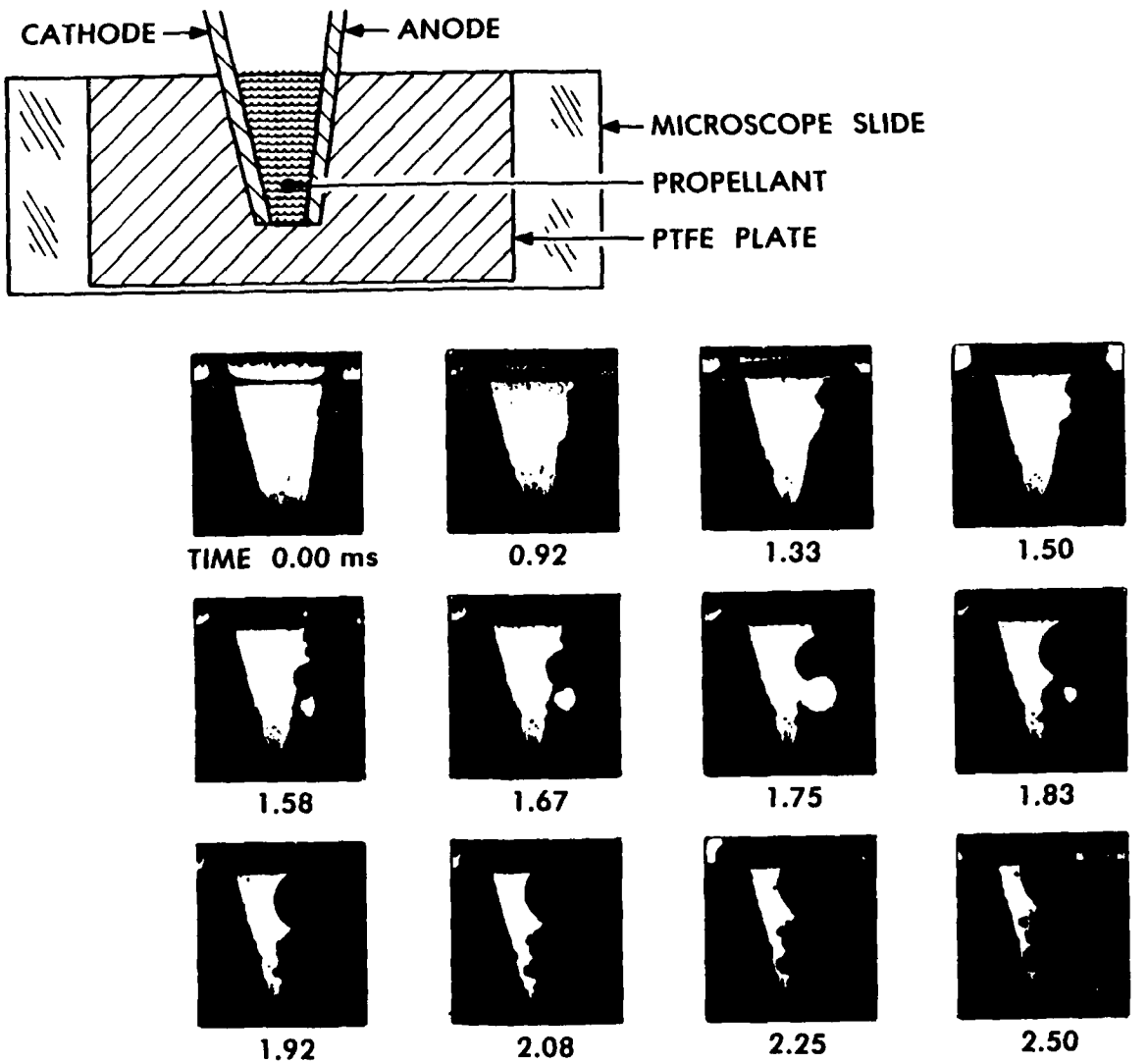

Figure 4. Reaction Sequence in Trapezoidal Plug

A number of interesting and potentially important features are seen in the Figure 4 data. Initiation of reaction is seen along the full length of the anode and is not greater at the bottom of the chamber where electrical resistance is presumably lower. Luminosity appears 1.58 ms after the onset of current flow and is restricted to the gap between anode and propellant, a space containing gaseous, intermediate reaction products that are non-conducting. Since the propellant is both a conductor and an energetic material capable of self-sustaining, exothermic reaction, several plausible mechanisms for reaction initiation resulting from the input of electrical energy can be advanced, the simplest of which is ohmic heating. In contradistinction would be an electrolysis scheme in which ions of appropriate charge would be transported toward electrodes, the onset of reaction being due either to ion-electrode reactions or to an increase in specific ion concentration in the vicinity of the electrodes.

On the premise that the reaction process is initiated by ohmic heating, a fairly uniform onset of luminosity along the current channel would be expected. Heat loss to the electrodes should presumably result in luminosity initially occurring away from these regions. The contrary is observed to occur. Gas formation and luminosity within it invariably occurs first close to the anode. It is thought likely that this is occasioned by local concentrations of ions which promote the process.

This does not rule out the importance of ohmic heating since any redistribution of electrical conductivity will result in a non-uniform pattern of current flow. Even in an essentially unidimensional system, any net space charge due to the predominance of one ion in the electrode region will cause a field gradient leading eventually to breakdown. The fact that such breakdown is observed only at the anode could be due to a difference in size of the positive and negative ions involved. Such difference would affect the severity of the field gradient, with the smaller ion achieving the conditions necessary for breakdown sooner.

Further studies, both of reaction initiation and plume venting are illustrated in Figure 5 using the coaxial plug (Figure 1C); the bottom electrode is the anode. As in Figure 4, reaction is observed prior to luminosity and only at a later stage of the sequence is the luminosity seen to move toward the catnode. A jet of unreacted propellant, at or near room temperature, is seen emerging from the vent, driven by the gases produced at the anode.

The contribution of chemical energy release cannot be readily ascertained from the Figures 4 and 5 data and the experiments were repeated with a saline solution of the same electrical conductivity as the propellant. NaCl solution, although conducting and capable of electrolysis, is not an energetic material and, therefore, cannot contribute chemical energy to the sequence observed. Figure 6 illustrates the results obtained.

FRAMING RATE
10000 pps

Figure 5. Reaction Sequence in Coaxial Plug (Propellant)

Figure 6. Reaction Sequence in Coaxial Plug (Saline Solution)

The same breakdown pattern is observed and luminosity seems greater, moving more rapidly to the cathode than in Figure 5. This may be deceptive since sodium is an intense emission source in that portion of the visible spectrum where film sensitivity is the greatest. A comparison of the emerging jets in Figures 5 and 6 shows interesting and possibly important differences. In the Figure 5 data, taken with propellant, the jet breaks up into a fine spray well before luminosity arrives at the cathode, whereas the Figure 6 data (saline solution) show no indication of such break up.

Since the windows of the transparent plug are equi-spaced and precisely parallel, the area occupied by the propellant image accurately represents its volume. Using simple translumination by a parallel beam, the transparent propellant yields an illuminated area. The droplets and gases which remain in regions from which the propellant has been expelled or burned scatter light, yielding sharp boundaries to the volume of unperturbed reactant. Using the high time resolution of the optical system, it is therefore possible to follow the rate of disappearance - whether due to consumption or expulsion - of the propellant within the cavity. Figure 7 shows the results of such a study taken at a framing rate of 50,000 frames per second and exposure time of 0.5 μs per exposure.

The data were obtained using a planimeter on projected images such as those shown in Figure 8. The Figure 8 data were obtained with a plug with integral orifice (Figure 1B). Electrode locations are such that reaction should begin in a lower corner of the chamber.

A photometric method working on the projected light would be more rapid and convenient as is a scanning video system currently under test.

While two-dimensional geometry applies to the contents of the plug, the ejected plume has every appearance of being axially symmetric, as might have been expected. Since simultaneous recording of events in the plume and in the cavity requires only a small shift in the light beam, it is possible to compare the volume disappearing from the cavity with that appearing in the plume. This is not the trivial exercise it might appear at first sight since, even in the absence of any reaction, atomization of liquid and entrainment of air beyond the orifice will manifest itself as an increase in apparent volume. The reason is that scattering of light by droplets and/or refraction by the boundaries of the jet produce the same effect on the transluminating beam. The change of plume volume with distance downstream allows us to judge the extent of air entrainment as a function of the axial coordinate in the plume and hence also the plume density, data required for momentum and energy calculations further discussed below.

It is of course possible to fill the plug only partially thus creating an air gap above the propellant. This air gap can be used to ensure initiation by a plasma arc by placing the electrodes within the gas. However, the presence, initially, of a gas layer complicates the process further. A series of observations were made with partially filled plugs under low voltage conditions so as to avoid the formation of an arc discharge. The plug, of 43 μl capacity, vented vertically upward. The effect of partial filling on ejection delay, ejection front velocity, and duration of current flow is shown in Figure 9.

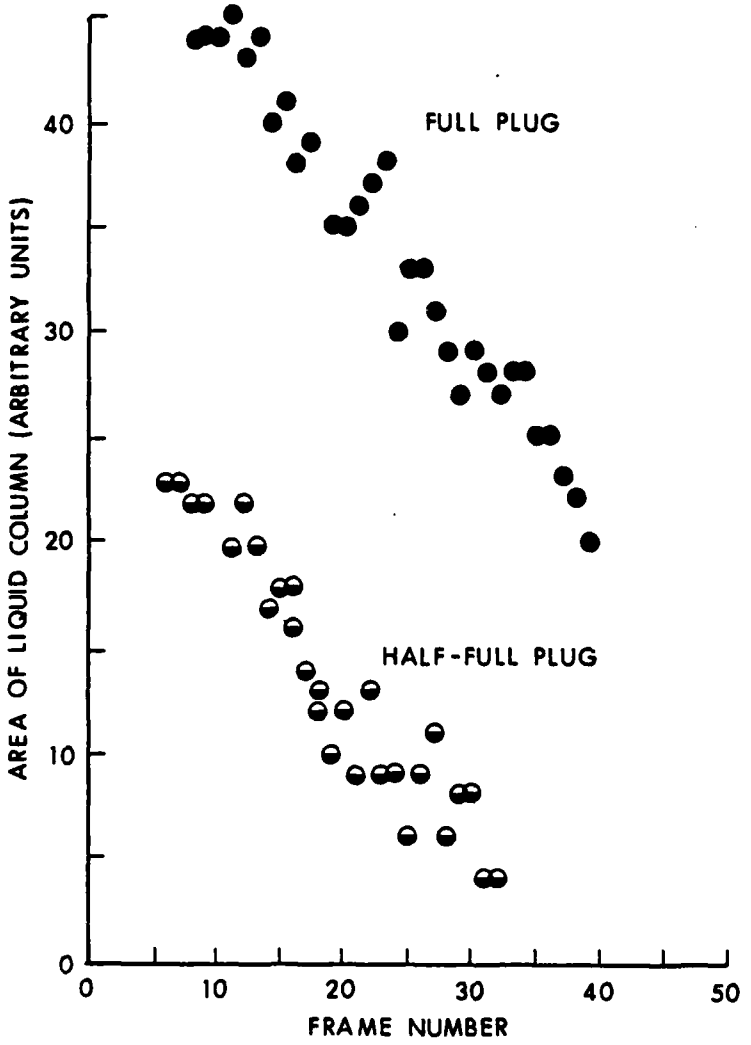

Figure 7. Expulsion/Consumption of Liquid From Plug

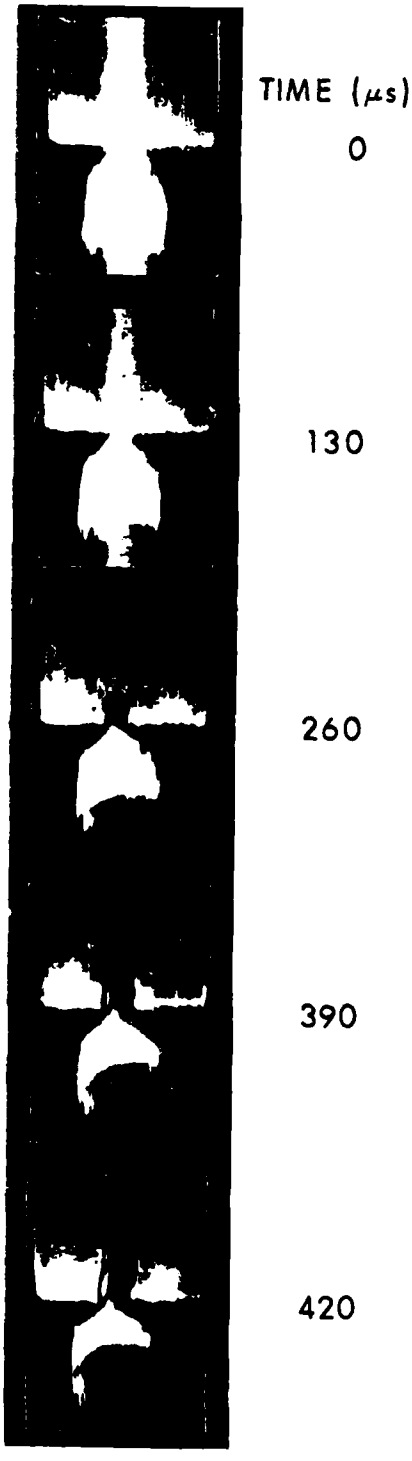

Figure 8. Plume Venting From Plug with Orifice

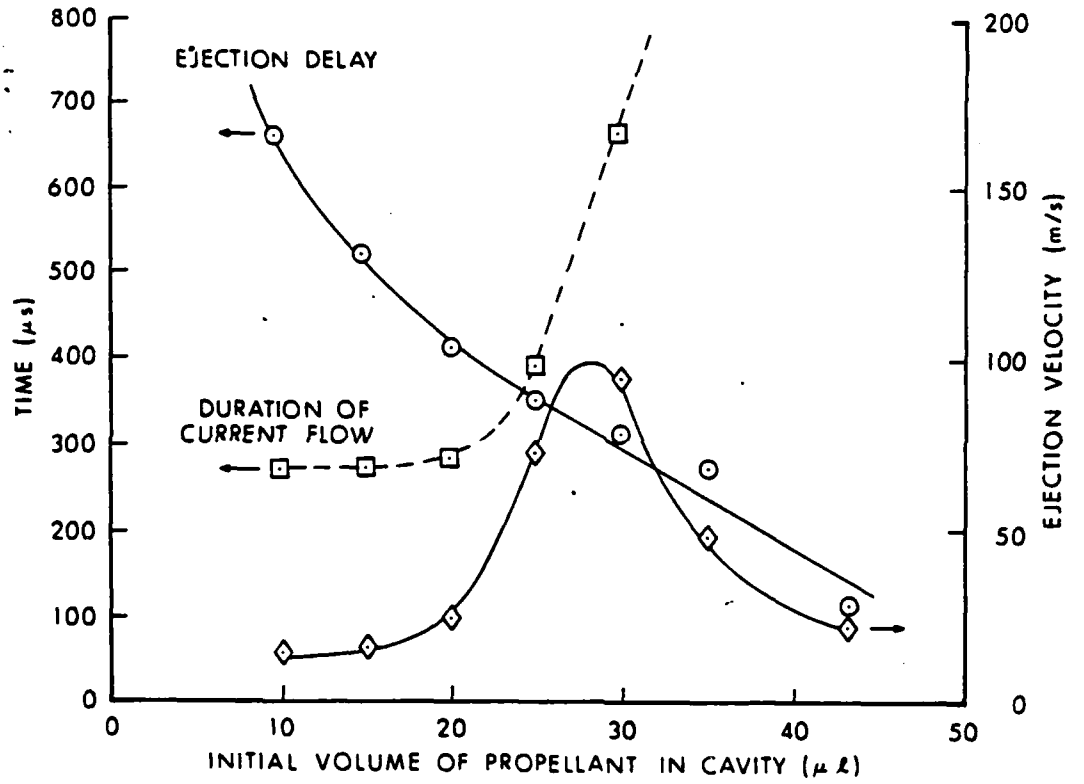

Figure 9. The Variation of Plug Performance With Volume of Propellant

The observed large decrease in ejection delay with increasing volume of fill is accounted for by the pressure drop in the orifice that controls the outflow of gas above the liquid surface in the cavity. The consequently increased time of current flow serves to increase driving energy for ejection. Velocity of the front of the ejected plume is seen to go through a pronounced maximum at around 28 μl filling. This does not, however, define mass movement of the propellant, since the plume rapidly changes shape. Analysis of the optical records associated with the Figure 9 data reveals a number of surprising features - features which do not necessarily apply to the arcing case and, more generally, to large energy releases. Firstly, no air is entrained in the jet, at least in its early stages - up to 400 μ s. The volume of the propellant disappearing from the cavity during this time, exactly equals the volume increase of the plume. This implies that no atomization occurs as the liquid leaves the orifice. Also the densities of the plume and the liquid within the cavity are the same. This observation is of use in attempts to correlate mass movement with its driving force. Since only the electrical energy input is measured, any chemical contribution to the driving mechanism would have to be inferred from the optical records of the propellant motion. Now the driving force, F, at any time, must be equal to the pressure within the bubble, P, multiplied by the area of its interface with the liquid, A, minus the drag force at the orifice, D. A is calculable from the length of the bubble boundary measured on the optical record and D from the mass flow velocity and a knowledge of the orifice area. Since the plume does not break up, the propellant may be treated as a continuous body of liquid outside and within the cavity. From its known shape, both its mass and the motion of its center of gravity (i.e. the combined centers of gravity of the two-dimensional slab within and the axially symmetrical plume outside the cavity) can be deduced. In the course of such an analysis of results it is found that there is virtually no acceleration of the mass after the first 200 μs. A balance of forces suggests that pressures in excess of 13 atmospheres must have occurred in the bubble during its early stages. It may well be that, under the conditions used, the controlling step is the formation of the initial bubble. The surface tension of the propellant requires a high pressure to produce a small initial bubble. Since the liquids used are very clean, nuclei for bubble formation are in short supply and very large forces may be involved in the first step of this process.

IV. CONCLUSIONS

The purpose of this report is to describe methods for detailed observation of electrical energy-propellant interaction. The use of plasma plugs with transparent propellant cavities and a sufficiently fast, reproducible optical recording system permit such observations to be made. Although some data are presented and briefly discussed, they are considered only as illustrations of the success of the methods developed. It is clear from the illustrations that these methods provide a powerful tool for elucidating, by using both simple visualization and quantitative measurements on the records, the underlying reaction mechanism. The phenomena observed appear interesting and revealing, although each possibly applies to only one particular reaction regime. The mechanism of the overall reaction initiation process will be discussed further in subsequent publications.

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