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DROPLET BURNING AT ZERO G

FINAL REPORT

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F. A. Williams

Department of Applied Mechanics and Engineering Sciences
University of California, San Diego
La Jolla, California 92093

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ABSTRACT

This study has been focused on questions of the importance and feasibility of performing experiments on droplet burning at zero gravity in Spacelab. The underlying interest is to acquire basic information that will advance knowledge of the physics and chemistry of droplet combustion, with attention directed specifically to the chemical kinetics, heat and mass transfer, and fluid mechanics of the phenomena involved. The work was divided into three phases, the justification study, the feasibility study and the conceptual development of a preliminary design. In the first phase, a study was made of the status of the theory of droplet burning, and available experimental results were reviewed, with the intention of defining potential gains from zero-g experiments. In the second phase, experiments on droplet burning were designed and performed in the NASA 2.2-second drop tower, to investigate the feasibility of obtaining the desired information without going into space and to define the unique contributions that experiments in Spacelab may provide. Results from the experiments performed during the second phase revealed a few new facts concerning droplet burning, notably burning rates in excess of theoretical prediction and a phenomenon of flash extinction, both likely traceable to accumulation of carbon produced by gas-phase pyrolysis in the fuel-rich zone enclosed by the reaction surface. However, these experiments also showed that primarily due to timing difficulties,

it was not possible to obtain a stationary droplet burning freely, away from supporting probes in the absence of gravity, for time periods in excess of one second. This work thus supplied evidence favoring performance of the experiments in Spacelab. It also resulted in preliminary designs for items of equipment, notably a remotely operated syringe for forming a droplet and placing it at a desired position, a low-energy, capacitive, ignition system for igniting the droplet with low impulse and a motion-picture system for photographing the combustion process. These results were used in the third phase of the work, the conceptual design of the Spacelab experiment. In addition, during the third phase attention was devoted to studies of methods for remote-controlled droplet positioning in zero gravity. An electrostatic method for droplet positioning was established as being desirable, and electrical parameters of droplets in electrostatic fields, relevant to the design of such a positioning device, were measured. The phase of conceptual design terminated with a specification of equipment and procedures for studying droplet burning at zero gravity in Spacelab. The primary results to be obtained from such experiments appear to be clarification of the role of time-dependent phenomena in droplet burning, establishment of relationships between extinction phenomena and chemical kinetics for various types of droplet extinctions and elucidation of the role of carbon production in droplet burning.

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1. INTRODUCTION

Among the many branches of physics and chemistry whose scientific knowledge might be improved through specifically designed experiments performed in Spacelab, combustion holds an unusual position in that basic information developed in the subject is likely to be comparatively immediately applicable to practical problems such as the efficient and pollution-free utilization of energy resources. The present investigation is concerned with a particular aspect of combustion that is of both basic and applied interest, namely, droplet burning. The objective is to document ways in which Spacelab experiments on droplet burning may be expected to advance basic understanding of the phenomenon, and to define specific experiments, including hardware aspects, having a high probability of producing results of appreciable scientific impact. The study was divided into the three phases identified in the abstract. The presentation that follows is divided into three sections, each corresponding to one of the phases of the study. The presentation in each section will be brief, with technical details contained in appendices. Following the sections on the three phases of the work is a section stating the contributions of the various personnel involved in the project and identifying publications. The final section lists the major, general conclusions reached.

2. JUSTIFICATION

The justification phase of this work addressed the question of ascertaining the need for experimentation on droplet combustion at zero gravity. The main activity during the justification phase involved a detailed review of existing literature on droplet burning. Results of this review are given in Appendix A, where it is indicated that important unknowns currently exist concerning effects of unsteadiness and of finite-rate chemistry on droplet burning and that zero-g experiments provide an attractive means for investigating these effects. The first intention of zero-g experimentation would be to attain spherical symmetry, thereby facilitating interpretation of experimental results. Attempts to achieve spherical symmetry through reduction of pressure generally result in extinction occurring prior to attainment of spherical burning, and attempts to achieve spherical symmetry through reduction of droplet size lead to observational difficulties that generally prevent an individual droplet from being observed for its entire burning history. Reduction in gravity is the only other way to decrease the buoyancy effects that are responsible for departures from spherical symmetry.

Consideration was given to reasons for selecting Spacelab experiments as the means for obtaining essentially zero gravity. The reasoning rests on the observation that droplets on the order of 1 mm in diameter typically burn for times on the order of 10 sec. Drop-tower experiments therefore provide insufficient time for observation of the complete burning histories

of the larger droplets of interest. Moreover, as discovered during the experiments performed in connection with the feasibility portion of this study, there are timing problems associated with drop-tower experiments. The release mechanisms for removing the support from the droplet imparts a motion to the droplet, and the experimental apparatus must be exposed to zero gravity at the instant that normal gravity has reduced this release-induced velocity to zero. Since difficulties in precise timing of the release of a package increases as the weight of the package increases, and since the required weight of the package increases with the zero-g time, there is a limit on the burning times of free, stationary droplets that can be studied in drop towers. These timing problems have been solved for the 50-lb packages needed in obtaining 1 sec of zero gravity but not for the 200-lb packages needed in obtaining 2 sec of zero-g. Thus, timing problems provide the main limitation that prevents drop towers from being employed in the needed experiments on droplet burning at zero gravity.

An alternative approach would be to perform the experiment in an aircraft that flies a zero-g trajectory. Zero-g times on the order of 30 sec can be achieved in this way. Unfortunately there are also timing problems associated with this approach. If the droplet is released automatically as in the drop-tower experiments, then the same timing problems, encountered in the drop tower, would occur, but with accentuated difficulty because of the difficulty of placing the aircraft in the zero-g trajectory at a controllable time. If, instead, the droplet is released after attainment of

zero gravity, then a means for remotely controlling the position of the free droplet is needed, and devices for doing this have positioning times on the order of the zero-g trajectory time. Adding this complication, which gives a low probability of success for any given run, to the necessity for making many runs to obtain the desired data for various droplet and atmospheric compositions, results in a long and expensive program having a low probability of success. Therefore the aircraft approach is not promising.

The only other alternative to performing the experiments in a space vehicle is to employ unmanned rockets, which can provide zero-g trajectories lasting minutes. Because of the remote nature of such a rocket experiment, its design would be formidable and likely would encounter timing and observational problems due to the absence of an on-board observer. In addition, the expense of such a program would be quite substantial. Therefore the rocket approach also was not deemed promising.

The experiments in Spacelab would have the advantage of affording sufficient time to observe the droplet for its entire burning history, even for relatively large droplets in the 1 cm size range. They also would have the advantage of considerably reduced timing problems and of having an observer on board to interpret results and to help in making decisions concerning subsequent runs from results obtained in previous runs. The Spacelab experiments would afford the opportunity to obtain a significant

amount of data for systematically varied conditions of droplet and atmospheric composition. It was therefore concluded that use of Spacelab is the most promising approach for obtaining the needed experimental information.

3. FEASIBILITY

A large part of the feasibility phase of this work involved design and performance of experiments in the 2.2 second drop tower at Lewis Research Center. The objectives of these experiments were first to ascertain whether the required experimental information could be obtained in ground-based facilities and second to develop information concerning key items of equipment needed in the conceptual design of the Spacelab experiment. In all, a total of 67 drops were made in the tower. The apparatus employed is described in Appendix B, which is a technical paper that was prepared on the basis of results of the first part of the testing. A more complete discussion of the details of the apparatus may be found in Appendix C, which is a thesis that was prepared on the basis of the results of this work. Subsequent to completion of the thesis, which involved 37 drops, an additional series of tests, involving 30 more drops, was run at the Lewis Research Center, addressed specifically toward the timing problems. Details of this last series are not reported here because, despite a number of changes motivated by the timing problems, the results essentially confirmed those reported in Appendices B and C.

Certain scientific results were obtained from the drop-tower tests. These are summarized in Appendix D. Additional details may be found in Appendices B and C. Essentially, the new results concerning droplet combustion were observations of enhanced burning rates and of flash extinction, both likely indicative of carbon buildup. The drop-tower tests proved to be worthwhile for obtaining qualitative information on the character of droplet burning at zero gravity, but they demonstrated that the quantitative information needed for studying transient combustion and extinction, as well as for testing the accuracy of quasi-steady droplet burning theory, probably can be obtained only by performing experiments in a space facility.

During the feasibility portion of this study, certain key feasibility questions were identified. These were:

- (a) What specific useful parametric studies of transient and extinction phenomena in droplet burning need to be conducted in Spacelab because of constraints imposed by ground-based facilities?
- (b) What types of apparatus will be useful for establishing a chamber having a controlled atmosphere within which experiments can be performed on droplet burning at zero-g?
- (c) What problems are likely to be associated with deployment of spherical droplets at zero-g in controlled environments?
- (d) What is the optimum ignition system and timing for achievement of reproducible ignitions of spherical droplets with minimum disturbances at zero-g?

(e) What photographic systems are suitable for providing adequate space and time resolution for observing transient burning and extinction of spherical droplets at zero-g?

The resolutions of these key questions, as developed from the results of the feasibility study, may be summarized as follows:

(a) Parametric studies were not attainable in the ground-based experiments attempted. Timing restrictions permitted only qualitative results to be obtained. The work shows that a facility such as Spacelab is needed for obtaining quantitative results. The specific parametric studies will be outlined in the section on Conceptual Design.

(b) The apparatus defined in NASA TM-78910, "Preliminary Concept, Specifications, and Requirements for a Zero-Gravity Combustion Facility for Spacelab," by R. L. DeWitt, Lewis Research Center, Cleveland, Ohio 44135, June 1978, provides an adequate chamber, having a controlled atmosphere, within which the experiments can be performed. Successful interfacing of the droplet burning experiment with this combustion facility is achievable.

(c) A remotely operated syringe, for placing a droplet on the end of a glass fiber in drop-tower tests, is described in Appendices B and C. This same syringe design can be used in designing the instrument to deploy a droplet at a desired position inside the test chamber in the Spacelab combustion facility. A mechanism may be incorporated to impart a small impulse to the syringe, for the purpose of detaching the droplet

from the syringe. Previous experiences, such as those in Skylab, suggest that the problem of droplet deployment is not difficult to solve. A means for controlling the position of the droplet in the chamber after detachment also is needed. An electrostatic positioner appears to be best suited for this task. The design of the electrostatic positioner is discussed in the section on Conceptual Design.

(d) A low-energy, retractable spark appears to represent the optimal means for igniting the droplet in the chamber of the Spacelab combustion facility. The design of the spark is discussed in Appendices B and C. Consideration may be given to release of a small amount of hydrogen at the spark for facilitating reproducible ignition. This refinement may be helpful but does not appear to be essential. Use of larger droplets reduces the disturbance given to the droplet by the ignition process, and the longer burning times associated with larger droplets present no problem in Spacelab experiments, whereas they may prevent observation of extinction in ground-based facilities. While ground-based experiments require automatic timing of the spark, the optimal procedure in Spacelab appears to be manual initiation of the spark, after the manually operated droplet positioner has placed the droplet at the desired location in the chamber.

(e) The 16 mm motion-picture camera operated at speed on the order of 400 frames per second, as discussed in Appendices B and C, appear to represent a suitable photographic means for providing adequate space

and time resolution in acquisition of data on droplet burning. Provisions for backlighting and for filters should be incorporated so that the droplet diameter itself can be resolved, without obscuration by flame radiation. Thought should be given to use of more sophisticated instrumentation in Skylab, namely interferometric photography to obtain density profiles, adsorption spectrometry to obtain concentration profiles and sampling to obtain species concentrations. However, these additional measurements should be viewed as refinements, since the primary data can be obtained by the photographic techniques that have been explored. Color photography for general information, as well as the black-and-white photography for data acquisition, should be considered.

The main general conclusion of the feasibility study is that the feasibility questions appear to be adequately resolved. There appear to be no remaining questions of principle that may prevent the experimental program from being completed successfully. Adequate systems for droplet deployment and for data acquisition have been constructed, while a promising approach to the problem of droplet positioning has been identified.

4. CONCEPTUAL DESIGN

The objective of the third phase of this work was to develop a conceptual design of a Spacelab experiment on spherically symmetrical

burning of fuel droplets in oxidizing environments, with specific attention given to transient effects and to effects of chemical kinetics, the latter involving extinction phenomena and carbon buildup. This objective involves consideration of the hardware needed to make the measurements and to achieve well-controlled, reproducible conditions, sufficient for quantitative testing of the quasi-steady theory of droplet combustion.

As currently envisioned, the experiment designed in this phase of the work will be conducted inside a chamber in which a controlled environment exists. The characteristics of this chamber are described in Ref. 1 of Appendix E. The composition of the gases in the chamber, as well as the pressure and temperature, will be known from measurements made during chamber filling. Interior to the chamber will be (a) a syringe device for droplet deployment, (b) an electrostatic device for droplet positioning, and (c) a retractable spark for droplet ignition. Outside the chamber will be (d) the gaseous and liquid consumables, (e) the plumbing required to fill and evacuate the chamber prior to and after each test, and (f) a motion-picture camera, with lenses and backlighting, to photograph the droplet as it burns. The viewing ports in the chamber will permit photography to be accomplished, as the primary mode of data acquisition, and will allow the experimenter to observe the test and to manually deploy and position the droplet, start the camera and initiate the spark. Prior to discussing experimental procedure, it is desirable to provide additional information on each of the components assigned a letter above.

(a) The syringe system is described in Section 4 of Appendix B and in Section 5 of Appendix C. Figure 4 of Appendix C provides a diagram of the syringe. Although this design was developed for drop-tower testing, it possesses the attributes needed for remotely deploying a droplet in the Spacelab experiment. A method is needed for detaching the droplet from the end of the syringe remotely. The electrostatic positioner can aid in this detachment. It is also desirable to fix an electromagnetically activated spring mechanism to the adjustable support of the syringe, so that the syringe can be retracted rapidly, over a distance of the order of 1 cm, to dislodge the droplet without additional fuel being fed into it by the plunger. This serves to provide an additional degree of control of the initial droplet size.

(b) The electrostatic positioning device is described in Appendix E, where it is also compared with other types of positioning devices. A schematic diagram of the device appears in Figure 20 of Appendix E. Complete testing of the electrostatic positioner in a ground-based laboratory cannot be performed because gravitational forces are comparable with the electrostatic forces and thereby limit residence times of droplets in the field. For this reason, the laboratory studies were focused on measuring design parameters that are critical to the operation of the device. The results of these laboratory investigations are reported in Appendix E. With the data obtained in Appendix E, rational design of the positioner is straightforward. It is of interest that although in theory

the induced dipole moment of the droplet may cause its motion to be sensitive to the field, in practice it appears that net negative charges acquired by the droplet overbalance this effect and provide an enhanced effectiveness of the system. The dual-mode capability of the system is designed to exploit both effects.

(c) The low-energy, retractable spark system, developed in connection with the drop-tower experiments is described in Section 4 of Appendix B and in Section 5 of Appendix C. Part of Figure 3 of Appendix C shows a diagram of the spark, and part of Figure 5 of Appendix C gives the circuit diagram. Some circuitry modifications have been developed but are not shown; the optimal design is not unique. The modified retraction arrangement shown in Figure 1, employing telescoping electrodes, seems preferable for the Spacelab experiment. Electrode extension, spark firing and electrode retraction are to be initiated manually in the Spacelab experiment.

(d) Requirements for liquid fuels are minimal in this experiment because only one droplet is burned in each test. One liter of each of the fuels to be tested is anticipated to be sufficient for the entire test series. Fuels to be studied are heptane, decane, hexadecane, methanol, a 50/50 mixture of heptane and hexadecane, kerosene, a heavy fuel oil, and a water-fuel emulsion, 10% water in hexadecane, with 2% Span 80 as an emulsifying agent. Gases to be employed are nitrogen and oxygen, sufficient amounts of each being needed to fill the chamber to 1 atmosphere

pressure 50 to 100 times, approximately one time for each experimental run.

(e) The plumbing requirements, as well as the requirements for measurement of pressure and temperature in the chamber, are consistent with the chamber design described in Ref. 1 of Appendix E.

(f) The motion-picture camera and its use are described in Section 5 of Appendix C. In the drop-tower testing, controllability was such that only a general view of the entire test area was useful. In Spacelab it is anticipated that a lens system for obtaining magnified photographs of the burning droplet itself will be useful. Also, backlighting of the droplet is required, to aid in measurement of the droplet diameter on the film. Instrumentation having greater sophistication, such as interferometry, absorption spectroscopy and gas sampling, is not studied in this report because it is felt that the major data of use will be the photographic records. In further studies consideration should be given to more elaborate instrumentation, for use after the primary, photographic data is acquired.

The experimental procedure involves first placing the apparatus inside the combustion chamber. The apparatus is to be inserted through the lower port and raised to the observation position, opposite the upper port. The apparatus is supported on a platform which holds the elements of the positioning device, the electrodes for ignition and the syringe for deployment of the fuel. The syringe must be filled in advance with the fuel to be tested. The chamber need not be opened between runs, unless

it is desired to study a different fuel, in which case the syringe must be removed and refilled.

The experiment is initiated by evacuating the test chamber and purging the system with the gas mixture to be used in the test. After gas motion ceases, the droplet is deployed by the syringe mechanism, the syringe is retracted, the droplet is positioned between the electrodes by the positioner, the electrodes are extended, and the data recording system is initiated. The spark is then fired to initiate combustion, and the electrodes are retracted. Tests are to be made both with and without the positioner operating during spark and combustion, to ascertain the effect of the positioner on the burning. After the combustion is completed, the data recording system will be stopped and the chamber evacuated.

The primary parameters to be varied from run to run are the pressure (0.1 to 2 atm), the initial droplet size (0.1 to 1 cm), the atmospheric composition (20% oxygen, 80% nitrogen to 100% oxygen), the fuel type (see (d) above), and the fuel and atmospheric initial temperatures. An individual test will require less than five minutes to run.

A schematic diagram of the apparatus is shown in Figure 1. The electrostatic positioner, the retractable electrodes, the syringe mechanism and the photographic recording system all are indicated in the figure.

The primary items of data to be obtained in these experiments are the flame diameter and droplet diameter as functions of time. This information will be extracted from the film records after return to Earth.

The data will be obtained for the range of conditions outlined above, to test existing theories of quasisteady droplet combustion and to investigate transient effects. Also to be measured are extinction diameters of droplets as functions of these conditions. From information of that type, overall rates of chemical reactions will be extracted by use of newly developed theories. Equally important are qualitative observations of different extinction modes, as well as possibly different burning modes, for different fuels. The extent of carbon buildup, as a function of fuel type and atmospheric composition, can also be obtained qualitatively from the film records and from on-board observation by the experimenter. It is on the basis of analyses of this type that advances in understanding of droplet burning are anticipated to be achieved.

The conceptual design, developed herein, is expected to undergo small modifications during the final design process, if the present experiment is carried out in Spacelab. The intention here has been to provide the concept for the design, not full design details. It appears that on the basis of the concept outlined above, a useful study of droplet burning can be developed and performed in the combustion facility of Spacelab. The potential payoff of such a program is an appreciably improved understanding of processes of droplet combustion.

5. PERSONNEL AND PUBLICATIONS

This work was performed by researchers at the University of California, San Diego, with help from personnel at the Lewis Research Center. The work

was supervised by F. A. Williams. The drop-tower experiments were designed by Brian Knight, who conducted the first series of drop-tower tests. His thesis, which is based on this work, appears in Appendix C. Tom Cochran, John Haggard, Ray Sotos, Gene Farkas and Tom Labus are among the NASA personnel who helped him with the experiments and their interpretation. The second series of drop-tower tests was designed and conducted by John Haggard, whose contributions to this project proved to be invaluable. The studies related to the electrostatic positioning device were performed at the University of California, San Diego by Moshe Kuna. Brian Knight contributed some early ideas to this study, the results of which appear in Appendix E. Klaus G. B. Sulzmann and Tohru Mitani helped in preparation of the final report. The continued helpful interest of Tom Cochran in this work provided an indispensable impetus to the entire study.

A paper on this work was delivered at the 1977 meeting of the Central States Section of the Combustion Institute and is included herein as Appendix B. Additional publications are the thesis of Brian Knight (Appendix C) and the work on the electrostatic positioning device (Appendix E), not yet submitted to a journal. F. A. Williams and Brian Knight are in the process of preparing a paper on the scientific results of the study, to be submitted to a journal for publication.

6. CONCLUSIONS

(a) There exist basic unknowns concerning droplet combustion, notably transient effects and effects of finite-rate chemistry, which can best be studied by performing experiments on droplet burning at zero gravity in Spacelab.

(b) It is feasible to perform the needed experiments on droplet burning in the combustion facility designed for Spacelab.

(c) The conceptual design of the experiment on droplet burning employs a chamber in which the combustion will occur, gas and liquid supplies and associated plumbing for placing the reactants in the chamber, a syringe mechanism for deploying the droplet in the chamber, an electrostatic positioning device for controlling the location of the droplet in the chamber, a spark system for ignition of the droplet, and a photographic system consisting of a motion-picture camera and lighting, for observing the droplet as it burns.

(d) A sequence of droplet-combustion tests in Spacelab, with systematically varied atmospheric and fuel compositions, holds the potential for providing a significant improvement in our understanding of basic mechanisms of droplet burning.

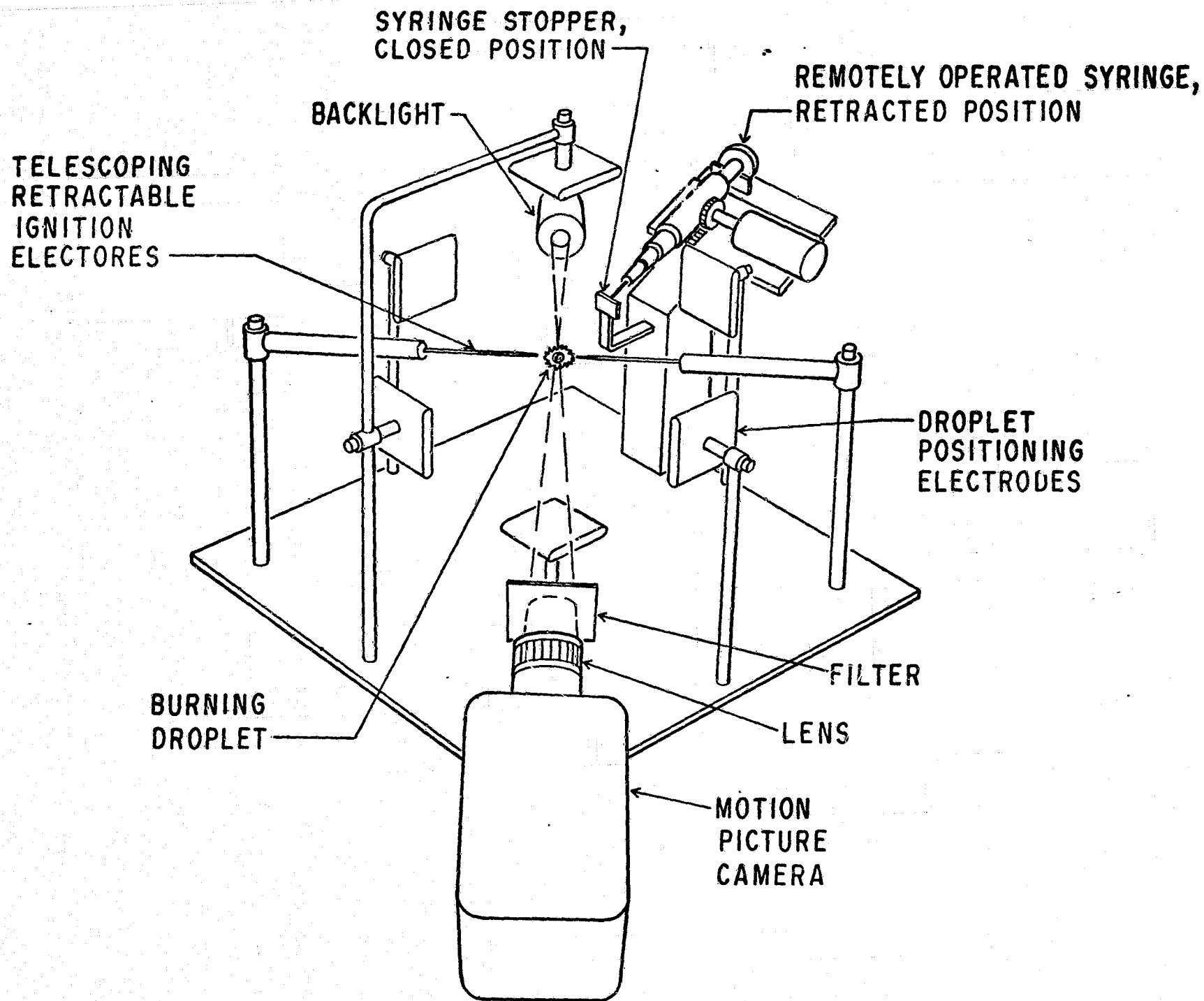


Fig. 1 Schematic Diagram of the Experiment

APPENDIX A

JUSTIFICATION STUDY

1. Introduction

The purpose of the justification phase of the work was to ascertain the need for experiments on droplet combustion at zero gravity through review of the existing literature on droplet burning. That droplet burning has been the subject of extensive study over many years is evident from numerous reviews of the topic [1-4]. Practical motivations for such investigations come mainly from the desire to achieve clean and efficient production of power through the combustion of liquid fuels. The fact that the studies are continuing demonstrates that unknown factors remain at the basis of the subject. Identification of these unknowns, provided by the literature review, points out what types of investigations hold greatest promise for advancing fundamental knowledge.

2. Analytical Background

The conservation equations of fluid mechanics describe droplet combustion, provided that allowance is made for transport phenomena and for chemical transformations in these equations[5]. Two factors prevent solutions to the equations for specific systems from being obtained readily. One is the complexity of the equations themselves in three-dimensional, time-dependent situations, and the other the fact that certain quantities appearing therein, especially chemical kinetic parameters, usually are not known. Theories for droplet burning have circumvented these two difficulties through introduction of approximations. To

the extent that the approximations correspond to reality, the analytical basis exists for calculating histories of droplet combustion.

Critical approximations in the classical theory are (a) that the liquid droplet maintains a constant temperature consistent with evaporative equilibrium at its surface, (b) that steady-state, spherically-symmetrical forms of the conservation equations are applicable in the gas phase and (c) that chemical equilibrium prevails and gives rise to a spherical flame-sheet positioned in the gas. With these and other, less critical, approximations, it is calculated that the ratio of the flame diameter d_f to the droplet diameter d_l remains constant during burning, being given by

$$\frac{d_f}{d_l} = \frac{\ln \{1 + [Q Y_O + \bar{c}_p (T_a - T_l)]/L\}}{\ln(1 + \nu Y_O)} , \quad (1)$$

and that the square of the droplet diameter decreases linearly with time t , according to the "d-square law",

$$d_l^2 = d_{l0}^2 - Kt , \quad (2)$$

where the "burning-rate constant" K is given by

$$K = \left(\frac{8\lambda}{\rho_l \bar{c}_p} \right) \ln \left\{ 1 + \frac{1}{L} \left[Q Y_O + \bar{c}_p (T_a - T_l) \right] \right\} . \quad (3)$$

Here Y_O denotes the mass fraction of oxygen in the ambient gas, ν the stoichiometric ratio of fuel to oxygen on a mass basis, Q is the heat released in the gas-phase combustion per unit mass of oxygen consumed, L the heat absorbed per unit mass of liquid fuel vaporized, $\bar{c}_p$ the mean specific heat

at constant pressure for the gas, T_a and T_l the ambient and liquid temperatures, respectively, λ is the average thermal conductivity of the gas, and ρ_l the liquid density. In essence, diffusion of oxygen through reaction products to the flame sheet, where it reacts to liberate heat that is conducted back to the liquid fuel to cause vaporization, comprises the dominant physical mechanism of droplet burning in the classical model.

3. Experimental Background

Although Eq. (2) often agrees well with experiment, one or more of the approximations on which it is based do not. To study the burning of an individual droplet experimentally, it has usually been necessary to support the droplet on the end of a fine quartz fiber [6-12], and the flows observed are manifestly nonspherical. The hot flame in a gravitational field causes gases to rise buoyantly about the droplet. An axisymmetric flow under natural convection is established, thereby violating approximation (b) above. Although a few attempts have been made to describe the nonspherical flow, the task is difficult since the Grashof numbers typically are neither large nor small. Thus, analysis of the observed flow has proven to be prohibitively difficult to date. Under these conditions, Eq. (2) must be viewed as an empirical correlation applicable over a limited range. Indeed, there is as much as a factor of 2 difference between measured values of K under similar conditions for similar fuels [12].

If droplet combustion influenced by natural convection were of major fundamental or practical concern, then greater effort should be expended on analysis of these experiments. However, aside from interest in phenomena of natural convection per se, it is not clear that studies of burning droplets

influenced by buoyancy would be profitable. From the practical viewpoint, droplets in combustion chambers are relatively small and are not supported by fibers, and influences of buoyancy thereby are reduced by droplet motions. From the fundamental viewpoint, underlying physical and chemical phenomena, such as the effects of chemical kinetics, can be studied more readily if the complicating influence of buoyancy is removed. Therefore there is interest in establishing experimental conditions under which droplet burning is studied in a buoyancy-free environment, not only for seeking a more vigorous validation of Eqs. (1) through (3) but also for obtaining a better understanding of basic combustion phenomena associated with droplet burning.

One method for minimizing buoyancy is to work with very small droplets projected into a hot gas stream [13-15]. This approach has the disadvantage that individual droplets cannot be observed throughout their histories; only overall information such as burning times can be obtained. An alternative approach that has been attempted [12] is to burn droplets at reduced pressure, since the Grashof number decreases with decreasing pressure. Unfortunately it is found that extinction due to finite-rate chemistry generally occurs at pressures above those at which the flame becomes essentially spherical, with suspended droplets. The only other way to decrease effects of buoyancy is to reduce gravity. This approach has been pioneered by Kumagai [16-20], who designed and performed zero-g experiments using drop towers. A means for extracting the fiber support from the droplet prior to ignition was developed [19, 20], and measurements showed that burning rates of free droplets differed somewhat from those of suspended droplets, even at zero g [19], because of disturbing influences, such as heat-sink effects, of the support. The free-droplet data of

Kumagai and co-workers [19,20] certainly are the best available on spherically symmetrical burning of fuel droplets today.

4. Unsteady Phenomena

Although these experiments [19,20] achieved spherical symmetry, the results still do not comply with the approximations of the classical theory. Although the d-square law again is satisfied closely, the ratio of flame to droplet diameter does not remain constant during burning, in contradiction to Eq. (1). Observationally it is evident that the spherically symmetrical gas flow is unsteady. This unsteadiness may be inherent in zero-g combustion, or else it may be attributable to the limited test time of about 1 sec. In none of the tests [19,20] was sufficient time available to follow the droplet to half its initial diameter, even though only small droplets, less than 1.5 mm in initial diameter, were employed. Extrapolation of data [19,20] indicates that more than 2 sec. are needed for the diameter of a 1 mm droplet to decrease to zero. A larger drop tower, such as the 2.2 sec. tower, or possibly the 5 sec. tower at NASA Lewis Research Center, would be needed if such a droplet is to be observed for its entire burning history. Larger droplets, having initial diameters in the range of 1 mm to 1 cm, can be observed for their entire burning history at zero g only in a facility such as Spacelab.

The observation of unsteadiness [19] has prompted further theoretical investigation of time-dependent phenomena in droplet burning. A practical motivation for this is that comparable time-dependent effects also may occur for small droplets in conventional equipment for power production. A basic motivation may be the desire to achieve better understanding of time-varying flows with

heat and mass transfer. The theoretical studies have taken two separate lines. In one [21] it is assumed that the unsteadiness arises from time-dependent responses that occur within the liquid. The gas is treated as quasisteady, and therefore only approximation (a) of the classical theory is omitted. In the second approach [22, 23] approximation (a) is retained but (b) is rejected, it being assumed that the gas flow is unsteady. Asymptotic methods can be employed [22, 23] to account for unsteadiness within the context of manageable theory. Both types of theories [21, 22] exhibit qualitative agreement with the available data [19].

Experiments at zero g in which conditions of initial droplet diameter and of initial liquid temperature are varied over appreciable ranges, would help to define better the regimes in which each of the two types of unsteady theories is likely to be more nearly correct. Some experiments of this type can be performed in drop towers, although greater variations in initial conditions should be achievable in Spacelab. Results of such experimental studies would help to clarify processes of unsteady droplet combustion.

5. Extinction Phenomena

Thus far, the underlying approximation (c) of equilibrium chemistry with a flame sheet has not been addressed here. In effect, this approximation has removed chemical kinetics entirely from Eqs. (1), (2) and (3). To the extent that the classical theory is applicable, droplet burning is a phenomenon of heat and mass transfer and of fluid flow, unaffected by complexities of the underlying chemistry. In fact, the chemistry intrudes in a variety of subtle ways.

For droplets burning in oxygen-enriched atmospheres, the flame temperature is high enough to produce a large amount of equilibrium dissociation of reaction products at the flame. There results the well-known [24] equilibrium broadening of the diffusion flame, which invalidates the flame-sheet approximation. Although there are approximations that lead to Eq. (2) without requiring a flame sheet,² these exclude various phenomena of basic interest, such as preferential diffusion of light and mobile species like hydrogen. Nevertheless, it seems likely that equilibrium dissociation will not contradict Eq. (2) but instead will merely modify K in a manner as yet unknown, while at the same time introducing arbitrariness in the definition of d_f , so that Eq. (1) becomes inapplicable. Effects of equilibrium dissociation can be studied at zero g in Spacelab through experiments on steady burning of porous spheres, by measuring temperature and concentration profiles (for example by thermocouple probing and gas sampling followed by gas chromatographic or other analysis). However, the potential payoff of such experiments is not studied herein.

The second way in which chemistry influences droplet burning is through finite-rate kinetics. Except for subsidiary effects such as the production of oxides of nitrogen, finite-rate chemistry becomes important only at low flame temperatures, where equilibrium dissociation often is unimportant. This occurs in nitrogen-enriched atmospheres, at reduced pressures or for very small droplets. Typically these influences set in suddenly, being negligible in most respects up until a point at which they become overriding and produce extinction.

Extinction of diffusion flames due to finite-rate chemistry is a well-known phenomenon for which an extensive theoretical [24-26] and experimental [27-28] background recently has been developed. The theory has been extended to the problem of droplet burning [29], and experimentally extinction through finite-rate chemistry has been observed for burning droplets [12] (as well as a reduction in burning rate near extinction through finite-rate effects). A droplet burns until reaching a size at which the residence time of reactive species in the flame zone is less than the chemical time for reaction of these species, at which size the flame around the droplet suddenly experiences extinguishment.

The implication of these theoretical and experimental results is that Eq. (2) is valid only for droplet sizes above a critical minimum value. This value is very small for hydrocarbon droplets burning in oxygen-rich atmospheres at atmospheric pressure, and therefore the influence of chemical kinetics is not likely to be significant under these conditions, although even here overall combustion efficiencies and the extent of hydrocarbon emission from combustion chambers can depend on the critical extinction diameters. At reduced pressures or in nitrogen-rich atmospheres, the extinction diameters should be observable. From measurements of extinction diameters as function of ambient conditions, information concerning overall chemical-kinetic parameters of the combustion process can be obtained [30]. Thus, studies of droplet burning in which extinction diameters are measured provide an avenue for approaching underlying chemical kinetics.

Extinction theory, for use in calculating parameters of chemical kinetics, is available only for spherical droplets. Droplets supported on quartz

fibers are highly nonspherical at extinction [12]. A clean experiment for approaching chemical kinetics requires spherical symmetry, which is achieved only at zero g. Drop-tower experiments can be used to measure extinction diameters provided that sufficient time is available for the droplet to burn to an identifiable extinction. The utility of drop towers for this type of study is uncertain at present; experimental exploration is required to ascertain whether drop towers can suffice. On the other hand, it is clear that in Spacelab sufficient time would be available for droplets to burn to extinction at zero g. Experiments on droplet extinction in Spacelab can contribute to the knowledge of the chemical kinetics of hydrocarbon oxidation.

6. Conclusions

From the foregoing it may be seen that the principal impact of Spacelab droplet-burning experiments is likely to lie in the improved scientific knowledge of (a) fluid-dynamical, heat - and mass-transfer aspects of unsteady flows involving combustion and of (b) chemical kinetics of combustion reactions. It is probable that influences of the results on the man in the street will stem from improved efficiency and cleanliness of power production as derived from better understanding of droplet combustion, rather than directly from the scientific results per se. Drop-tower experiments definitely are warranted for the purposes of (a) better defining what information can be obtained only through experiments in space and of (b) discovering experimental information that will aid in designing droplet-burning experiments for Spacelab.

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APPENDIX B
OBSERVATIONS ON THE BURNING OF DROPLETS
IN THE ABSENCE OF BUOYANCY*

1. Short Abstract

The first stages of an experimental study on droplet burning at zero gravity are described. Motivations for the work are to investigate transient burning and effects of finite-rate chemistry, in a geometrical configuration sufficiently simple for theoretical analyses to be performed in detail and checked against experiment. Two possible approaches to the experimental problem are to establish burning in a freely falling chamber in a drop tower and to perform the experiments in a research facility in space, such as spacelab. The work reported here concerns the drop-tower experiments. The design of these experiments is discussed, and preliminary results of the experimental program are given. Critical problems in drop-tower testing of this kind, especially timing problems that arise in this particular experiment, are emphasized.

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2. Extended Abstract

Kumagai has succeeded in observing the burning of fuel droplets in oxidizing atmospheres uninfluenced by buoyancy. His measurements were made in a freely falling chamber which afforded slightly less than one second of negligible gravitational acceleration. This allowed sufficient time for establishment of an essentially constant value for the time rate of decrease of the square of the droplet diameter. However, it did not enable him to observe the burning all the way to the time of its completion. In particular, he found a transient behavior for the ratio of flame to droplet radius, over the entire duration of each of his tests. The present work concerns efforts to extend the time of observation of a free droplet burning in a quiescent atmosphere with negligible influences of buoyancy.

The purpose of designing such experiments is to test the basis of the theory of droplet combustion. Two aspects of the theory are especially involved, viz. (1) quasisteady burning and (2) infinite rates of chemical reaction in the gas phase.

(1) Although Kumagai's results show a perpetually transient flame, there seems to be a tendency for the flame location to approach a quasisteady situation at the end of his testing time. By extending the time of observation, it may be possible to demonstrate that later in its burning history the droplet conforms closely to the basic theory. In addition, if quasisteady behavior does not develop, it may be possible to ascertain, through comparison with recent theoretical advances,

whether the transient behavior stems predominantly from liquid-phase or gas-phase processes.

(2) In recent years it has become well known that a droplet burning in a cool atmosphere must experience extinction prior to achieving zero radius. The reason for this is that the residence time in the flame decreases as the droplet diameter decreases, eventually becoming smaller than the chemical time required for combustion to occur in the reaction zone. The consequent failure of the approximation of infinite-rate kinetics affords the possibility of obtaining information on chemical rates, if the extinction phenomenon can be observed. There is a possibility that, by extending the test time in Kumagai's experiment, systematic measurements on extinction can be made, from which fundamental kinetic data can be extracted.

With these objectives in mind, a program has been initiated to photograph free droplets burning under negligible gravitational acceleration in controlled atmospheres, for longer durations than previously attained. There are two approaches to the problem. One is to employ a freely falling chamber in a drop tower taller than that used by Kumagai, and the other is to perform the experiment in spacelab. Work on the drop-tower approach is discussed here.

The apparatus consists of a chamber within which a controlled atmosphere can be established and containing the fuel and the means for igniting it, a support system for an individual fuel droplet, with

provision for removing the glass-fiber support from the droplet through a spring-type mechanism, a remotely operated syringe system for placing a measured amount of fuel on the supporting fiber, an ignition system incorporating retractable electrodes that can be removed from the vicinity of the droplet after ignition, a data acquisition system consisting of a Millikin camera, focused on the burning droplet and operated at framing rates on the order of 400 per second, and an electronic timing system for synchronizing the camera start-up, the spring-removal mechanism, release of the package in the drop tower and firing of the spark to initiate combustion. The maximum time of negligible gravity in these tests is 2.2 sec.

3. Introduction

In the field of combustion, one particular aspect that is of both basic and applied interest concerns the burning of liquid fuel droplets. That droplet burning has been the subject of extensive study over many years is evidenced by the fact that an early review [1] contains more than sixty references. Practical motivations for such investigations come mainly from the desire to achieve clean and efficient production of power through the combustion of liquid fuels. But certain unknown factors still remain at the basis of the subject.

The conservation equations of fluid mechanics describe droplet combustion provided that allowance is made for transport phenomena and for chemical transformations in these equations [2]. Two factors prevent solutions to the equations for specific systems from being obtained readily. One is the complexity of the equations themselves in three-dimensional, time-dependent situations, and the other the fact that certain quantities appearing therein, especially chemical kinetic parameters, usually are not known. Theories for droplet burning have circumvented these two difficulties through the introduction of approximations, of which three are (a) that spherically-symmetrical forms of the conservation equations are applicable in the gas phase, (b) that quasisteady conditions are achieved, in the sense that steady-state equations can be employed in the gas and droplet properties such as its temperature are invariant with time, and (c) that chemical equilibrium prevails and gives rise to a spherical flame-sheet positioned in the gas. Numerous refinements to the

early theoretical considerations of Godsave [3] include the semiempirical treatment of effects of forced and natural convection [4-8].

The experimental study of an individual droplet, however, has usually necessitated the support of the droplet on the end of a fine quartz fiber [3, 4, 9-13], and flows are manifestly non-spherical. The hot flame in a gravitational field causes gases to rise buoyantly, establishing an axisymmetric flow under natural convection, thereby violating approximation (a) above. Attempts to describe the non-spherical flow have proven difficult since the Grashof numbers typically are neither large nor small. It is not clear that studies of burning droplets influenced by buoyancy would be profitable since from a practical viewpoint the droplets in combustion chambers are relatively small and are not supported by fibers, so influences of buoyancy are reduced by droplet motions. From the fundamental viewpoint, underlying physical and chemical phenomena, such as the effects of chemical kinetics, can be studied more readily if the complicating influence of buoyancy is removed.

Minimizing buoyancy has been achieved by working with very small droplets projected into a hot gas stream [14-16], but individual droplets could not be observed throughout their histories. An alternative approach that has been attempted [4] is to burn droplets at reduced pressure, since the Grashof number decreases with decreasing pressure. It is found, however, that extinction due to finite-rate chemistry generally occurs at pressures above those at which the flame becomes essentially spherical,

with suspended droplets. The only other way to decrease buoyancy effects is to reduce gravity.

Previous experiments by Kumagai [17, 18, 19], in which a freely falling chamber eliminated significant influences of gravity for a short period of time, have been successful at achieving spherical symmetry. However, observations showed that the gas flow was unsteady, and the ratio of flame to droplet diameter did not remain constant during burning, which violates approximation (b) above. This unsteadiness may be attributable to the limited test time of about 1 sec. In none of these tests was sufficient time available to follow the droplet to half its initial diameter, even though only small droplets, less than 1.5 mm in initial diameter, were employed. Since there seemed to be a tendency for the flame location to approach a quasisteady situation at the end of Kumagai's testing time, by extending the observation time it may be possible to demonstrate that later in its burning history the droplet conforms closely to the basic theory. The work reported herein constitutes an attempt to achieve such an extension by use of a drop tower having 2.2 sec of negligible gravity.

The objective in use of the drop tower has been to eliminate significant influences of buoyancy and of nonspherical convection for a period of time long enough to allow observation of the entire burning history. In this manner, the combustion can be recorded down to droplet sizes so small that the chemical conversion time in the gaseous flame become comparable with

the residence time therein. Under these conditions, approximation (c) above is expected to begin to fail, and in a cool atmosphere droplet extinction theoretically should occur [20]. Measurement of extinction diameters as functions of ambient conditions of pressure and of oxygen content in the atmosphere should provide data on overall rates of oxidation and thereby further the understanding of chemical kinetics of hydrocarbon combustion. The current status of our attempts to achieve these goals is covered in the following pages.

4. Experimental Apparatus and Procedure

In order to study the burning rates of spherical droplets, a method must be devised to confine the droplet to a pre-specified location without there being solid objects (e.g., glass fibers) near it to destroy the spherical symmetry. Various methods have been used for positioning liquid droplets, but many require large power supplies, while others require a considerable length of time to accomplish positioning [21]. Kumagai et al. [18, 19] have developed a method which requires little equipment and is fairly reliable, and it is upon this basic design that the present system has been constructed.

The test apparatus consists of a metal framework which contains the test chamber, ignition system, batteries, timers and high-speed motion-picture camera, (Figure 1). The framework, or drop frame, is a 30" x 15" x 15" aluminum structure designed to withstand a 30-g. impact upon completion of each test. During a test, it acts as a completely separate

unit with nothing to interfere with its free-fall condition. The drop frame is used in the NASA-Lewis Research Center's Zero-Gravity facility which is capable of providing 2.2 seconds of free-fall with drag forces to 10^{-6} g.

Since the tests performed involved burning in oxygen or oxygen-nitrogen atmospheres ranging in pressure from 0.1 to 2 atm., it was necessary to build a test chamber within the drop frame containing the droplet deployment and ignition systems. The chamber is a 12.5" x 10" x 11" box made of 3/4" plexiglas, fitted with an o-ring at its base to provide a seal against the chamber bottom. The chamber bottom is rigidly fixed to the drop-frame, and access is gained to the experiment by removal of the box. During a test, the chamber is held against the bottom through the use of 4 stainless steel threaded rods and 2 cross-bars which exert a downward force on the box. The use of clear plexiglas enables the camera to film the burning within the chamber and the operator to see the internal mechanism in order to deploy the droplet.

The droplet release mechanism, located within the chamber, is the heart of the apparatus. As shown in Figure 2, it consists of an aluminum "piston" which is contained in a 0.825" I.D. cylinder. Into the piston are inserted a brass rod, $1\frac{1}{2}$ " long, and a 2 mm max. dia. quartz fiber. The piston rests against a helical spring, and when the spring is compressed, is held in place by an electromagnetically actuated catch. A microswitch on an adjustable support is used to activate the ignition source. The ignition electrodes consist of fine copper wires, spaced about 1.5 mm apart, located

at the tip of the quartz fiber. The electrodes are mounted on a sliding track which is held in place by a second electromagnetically actuated catch against the tension of a stretched helical spring.

The release mechanism uses the motion of the droplet to enable the release of the drop frame to be timed properly. The piston is pushed against the spring and held by the catch. After a droplet is mounted on the fiber, the first relay is energized. The piston begins an upward motion, but its acceleration is great enough to pull the fiber out of the droplet. The droplet continues moving upwards, but is now being decelerated by gravity. The droplet reaches a point where it has no vertical velocity which occurs approximately 0.02 sec. after release. At that time, the piston which was still moving upwards trips the micro-switch which energizes the ignition circuit and the second electromagnet. As a consequence of the resulting catch release, the electrodes then begin moving away from the droplet. At the same instant that the droplet reaches the high point of its trajectory, and zero velocity, the drop frame is released into free fall. In this manner the droplet is stationary with respect to the camera mounted on the drop frame, no physical objects are supporting it, and it is now in free-fall.

Since the release mechanism is located inside the pressure chamber, a means for mounting the droplet by remote control is needed. Figure 3 shows the droplet mounting mechanism. It consists of a hypodermic syringe activated by two small d-c motors. One motor drives the syringe, which can be moved to bring the needle tip in contact with the quartz fiber. The

second motor, through a 9780:1 gear reduction, slowly moves the plunger in to force the liquid fuel out the needle tip. A droplet is then transferred to the fiber. The droplet size can be controlled fairly easily due to the fine control provided by the gear reduction, and by the use of a fine-gauge syringe needle (25 gauge). The syringe is then withdrawn slowly from the fiber and the droplet remains on the fiber. The syringe is then moved back away from the combustion zone.

The ignition system consists of an automotive induction coil for the high secondary voltage needed for the spark gap, a 4300 μF 50 Vdc capacitor which is discharged through the coil's primary windings, and the control switches for the spark timing. Power was supplied by three 15 Vdc nickel-cadmium batteries.

The camera used for the tests was a 16 mm Millikan running at a framing rate of 400 PPS. The camera was controlled through a set of relays in the drop frame control box, and powered by two 15 Vdc nickel cadmium batteries. Lighting was also provided through the control box, as was the power for the droplet release mechanism. Wiring enters the chamber through pressure fittings in the bottom.

At the beginning of a test, the syringe is filled with 0.25 cm^3 of fuel, the piston is "cocked" into position, as are the electrodes, and the syringe is moved forward until the needle tip seals in the R. T. V. stoppers shown in Figure 2. The chamber top is put in place and secured, and the chamber is evacuated. It is then refilled slowly with N_2 until a specified

partial pressure is reached, and then filled with O_2 until the test pressure is reached. The pressure is monitored to ensure that no over-pressure develops if throttling has occurred while filling the chamber.

The drop frame is then placed within an air-drag shield (Figure 1) whose purpose is to shield the frame against air resistance. The drag shield moves separately from the drop frame and during the course of a test will move 8" relative to the frame due to air resistance on the shield. At the base of the drag shield are penetration spikes which are designed to absorb energy as the package strikes the deceleration tank — a 6 ft. deep box of aerated sand. Access doors to the test package are mounted on the drag shield.

The entire test package is hoisted to the top of an eight-story tower where the package release mechanism and timers are located. The package is suspended by an over-stressed music wire which is passed through a cutter assembly. The control box relay switches are connected to the main control console through umbilical cables, and the normally-closed relays on the drop frame control box are energized (Figure 4). All package switches are then placed in the "ON" position but due to the opened relays, no mechanisms operate. The droplet is then mounted by remote control. To initiate the test run, the automatic timers are set on the main control console, and the start button is activated. The camera circuit closes first, starting the camera. After 2.0 sec., the droplet release electromagnet circuit is closed, releasing the droplet. In another

0.02 sec, the cutter is activated which releases the test package into free-fall, and the spark circuit is energized igniting the droplet. After 2.2 sec, the package experiences the 30-g. impact and the control box shuts off power to the various circuits. The package is then retrieved and prepared for the next run.

5. Experimental Observations

The following discussion is based on the results of thirty-six tests performed in the zero-g facility on n-heptane and decane in atmospheres of 20, 25, 30, 50 and 80 percent oxygen diluted with nitrogen at pressures of 1/2 and 1 atmosphere. These tests were designed to determine the performance characteristics of the test apparatus and the critical timing sequence necessary to achieve a stationary droplet.

The droplet mounting and release mechanisms worked to our full expectations, providing a very reliable means of mounting a small (approximately 1 mm diameter) droplet at the end of the quartz fiber, and of releasing that droplet between the spark electrodes. It was found that any disturbances to the surface of the droplet due to the action of the fiber being withdrawn were damped-out completely within 0.1 seconds, and a spherically symmetric droplet was formed. Ignition was achieved in 84% of the tests; the remaining tests did not achieve ignition due to either misalignment of the droplet with the spark electrodes, or malfunction of the spark system itself. Since the electrodes were of low mass and were mounted on a sliding

arrangement, there appeared to be no disturbance to the flame shape due to the ignition system.

The timing system, however, proved to be a critical factor in the performance of the apparatus. Even after the installation of high-precision digital timers (accurate to within ± 0.0005 sec) it took several tests in order to get the timing close to a point at which the droplets were moving with a relatively slow velocity, on the order of 10 cm/sec. With the use of a "zero-g light" which indicated on film when the drop frame entered free fall, the timing was adjusted in increments in an effort to achieve a stationary droplet. However, an irreproducibility in timing of ± 0.01 seconds appeared to be associated with the package release mechanism; subsequent tests at identical timing settings had a difference of 0.02 seconds between the time the fiber began withdrawing from the droplet and the time that free-fall was achieved.

As a result, all but one test had a droplet moving in the field of view of the camera. The test in which the droplet was stationary was one in which the spark system failed to operate. This may indicate that motion of the droplet is induced by the spark discharge, but this point remains uncertain due to the irreproducibility of the relative time to enter zero-g. Because of the small field of view of the camera, approximately 5 cm vertical distance, the moving droplets were in view for a limited time. The motion also brought the droplets in contact with objects within the chamber which distorted the flame, and in oxygen-rich atmospheres

sometimes resulted in ignition of combustible material encountered by a burning droplet. In spite of these difficulties, undisturbed burning occurred in several tests for approximately 0.5 seconds. In general, the period of growth of flame diameter was shorter than previously observed [18], due to the convective effects.

Several interesting observations were made concerning the burning of droplets in the absence of buoyancy with forced convection. Heptane droplets with initial diameters of 1.3-1.4 mm extinguished after ignition when the velocity of the droplet was nearly 16 cm/sec in an atmosphere of 20% O_2 and 80% N_2 at 1 atm total pressure. This result may be compared with corresponding results for suspended heptane droplets, at 1g, in which externally imposed convective velocities above 30 cm/sec were achieved without extinction [1]; the supporting fiber may help to stabilize the flame in these earlier experiments. Heptane droplets of the same approximate size in an atmosphere of 50% O_2 , 50% N_2 at 1 atm total pressure did not extinguish in the present zero-g tests even when a velocity of 43 cm/sec had been reached. This difference is qualitatively consistent with theory in view of the greatly reduced chemical reaction time at the elevated flame temperature of the enriched atmosphere.

Decane droplets burning in 50% O_2 were obscured by their highly luminous flame, preventing observations of droplet diameters by the photographic technique employed. Only at reduced oxygen concentration,

i.e. 25% or 30%, was the droplet visible during the entire burning time on film. Heptane droplets proved to be more visible under the same conditions since their flame was less luminous. The increased luminosity of the flames of the higher alkanes appears to reflect a greater tendency toward carbon formation.

Under forced convection, the flame shape was distorted from the spherical, with the expected elongation forming in the wake of the droplet. Interestingly, during the first 0.1 seconds of burning while the droplet surface is oscillating from the fiber withdrawal, the flame seemed to eject glowing particles in its wake. This behavior was most prevalent with decane droplets.

In one test, a decane droplet with an initial diameter of 1.4 mm rebounded from objects in the chamber and was therefore in the camera field for the full 2.2 seconds of free-fall. It had not extinguished at the completion of the test, which was conducted at 1 atm total pressure with 50% O_2 , 50% N_2 composition. This indicates that for extinction to be observed, smaller droplets must be used, and conversely a longer test time must be available to observe extinction due to finite-rate chemistry in larger droplets.

Two tests with decane in 50% O_2 , 50% N_2 at 1 atm revealed a phenomenon whose explanation remains uncertain. Satellite droplets were ejected from the primary droplet at ignition and were observed to

burn to very small diameters. After the flame had become barely visible, a luminous flash was recorded on one frame of the film (at a framing rate of 400 PPS), then combustion apparently ceased. The flash was reminiscent of microexplosions observed with emulsified fuels [22]. The phenomenon appears likely to necessitate the presence of impurities in the decane whose relative concentrations increase during burning. Whether these impurities are present initially or accumulate during burning remains unclear. The phenomenon suggests that over certain ranges of atmospheric conditions it may be difficult to observe quiet extinctions of the type from which overall chemical kinetic parameters of the gas-phase combustion can be extracted.

6. Conclusions

(a) Reproducibilities in timing of about 2 msec are needed for all components in drop-tower testing if stationary, burning droplets are to be observed under zero-g conditions in experiments of this type.

(b) Quantitative data on histories of droplet and flame diameters and on droplet extinction are not likely to be obtainable in the experiments reported herein without improved timing.

(c) Unexpected qualitative observations of new phenomena, such as flash extinctions, may be obtained in experiments of this type, even if quantitatively reproducible conditions are not achieved.

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8. List of Figures

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DROP ASSEMBLY

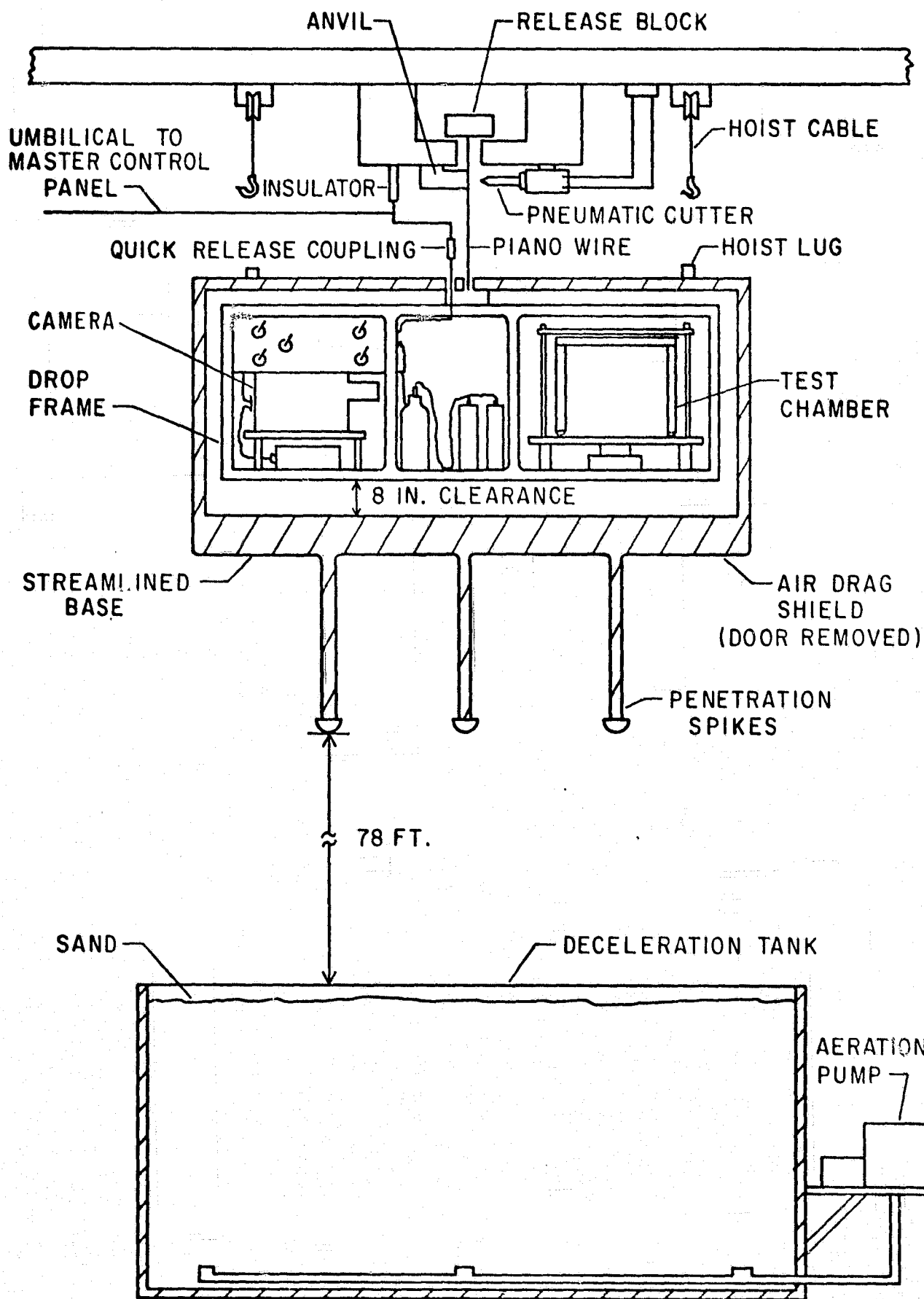


Fig. 1

DROPLET RELEASE MECHANISM

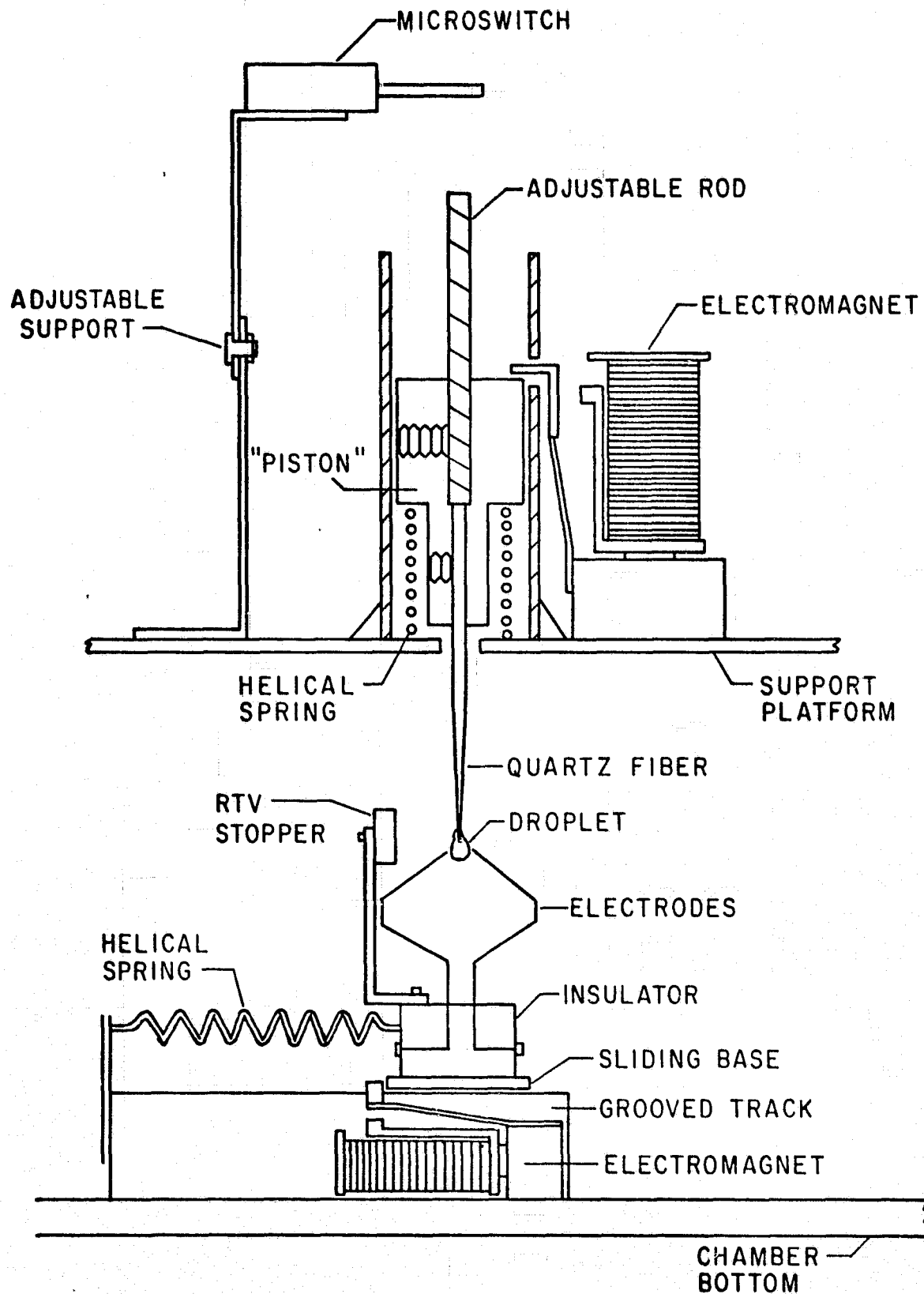


Fig. 2

DROPLET MOUNTING MECHANISM

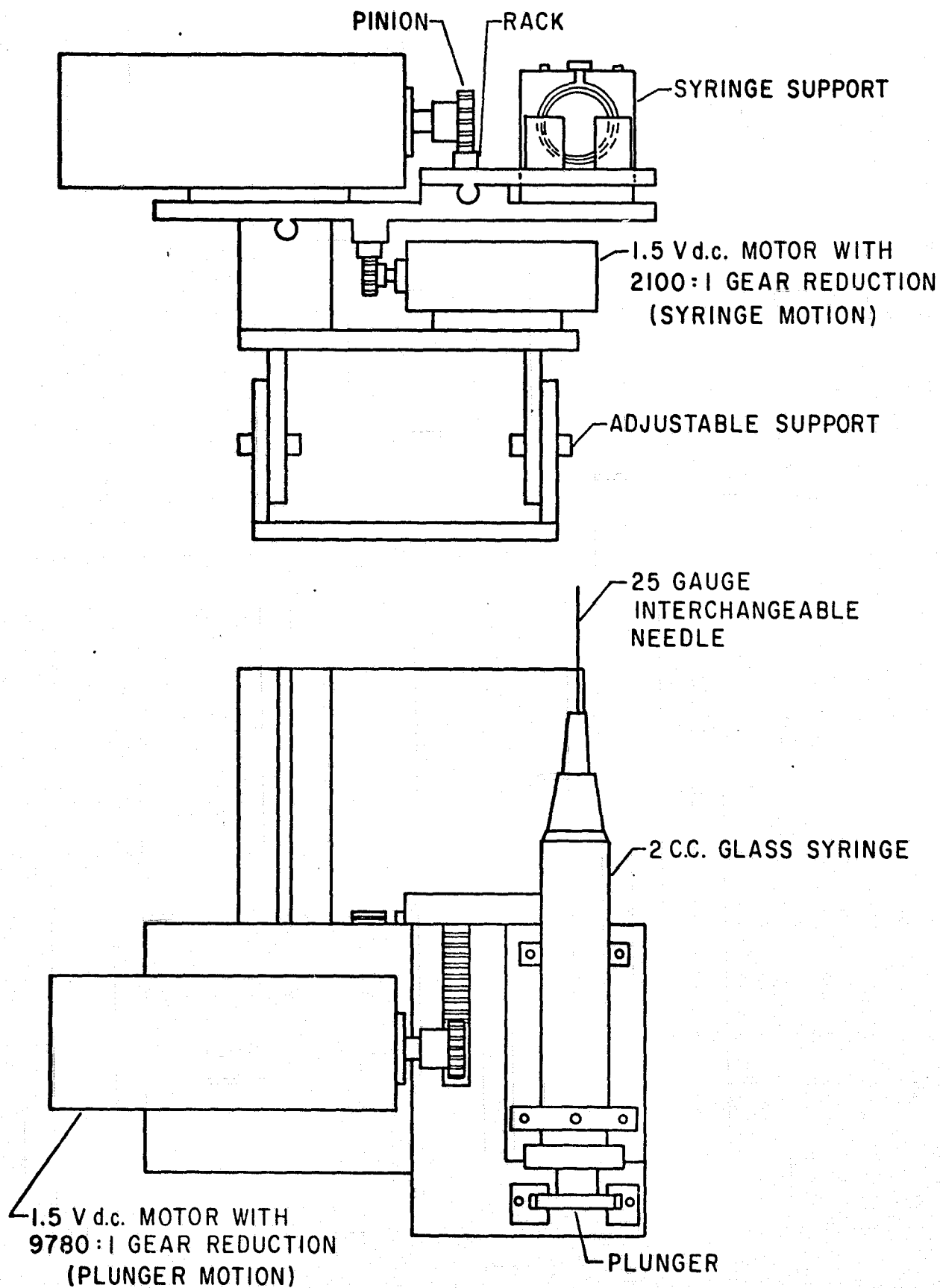


Fig. 3

CONTROL SYSTEM

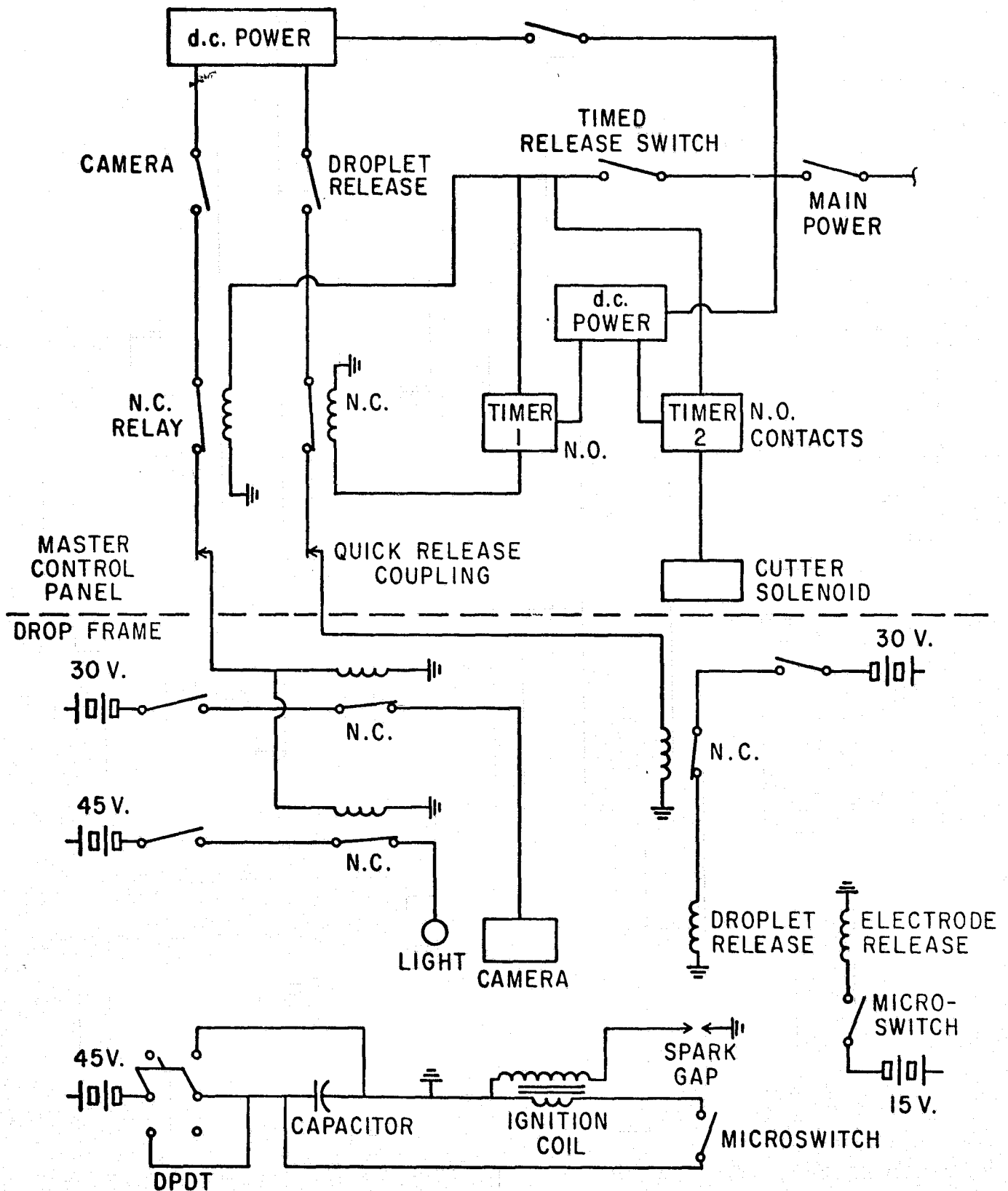


Fig. 4

APPENDIX C

COMBUSTION OF HYDROCARBON DROPLETS
IN THE ABSENCE OF BUOYANCY*

1. Abstract

An experimental apparatus was designed to study the burning of single hydrocarbon fuel droplets in the absence of buoyancy. A series of experiments upon n-heptane and decane droplets burning in atmospheres consisting of 20, 25, 30, 50 and 80 percent oxygen diluted with nitrogen at pressures of $1/2$ and 1 atmosphere were performed at the NASA-Lewis Research Center's Zero-Gravity Facility between August 1976 and April 1977. Data from the experiments were obtained from a 16 mm high speed motion picture camera which recorded the events that occurred in each of the thirty seven tests performed.

The objective of this experiment was to eliminate the significant influences of buoyancy and of nonspherical convection upon a single burning droplet in order to determine the effects of variations in the ambient gas constituents upon the burning rate. Ultimately it was hoped that burning could be observed down to droplet sizes so small that chemical kinetic effects would predominate and extinction would be observed, thereby

* The technical content of a thesis submitted by Brian Alan Knight, in partial satisfaction of the requirements for the degree of Master of Science in Applied Mechanics, University of California, San Diego, 1977.

providing information upon the kinetics of hydrocarbon oxidation.

However, it was not possible to obtain stationary droplets so that burning could not be observed in the absence of forced convective effects.

The experimental burning rate constant for n-heptane burning in air was determined by a linear regression analysis of the data to be $K = 1.775 \times 10^{-2} \text{ cm}^2/\text{sec}$. The theoretical value corrected for forced convective effects was $K = 1.083 \times 10^{-2} \text{ cm}^2/\text{sec}$ based upon the classical constant property theory. This heptane droplet was observed for 2.16 seconds in the absence of buoyancy.

The burning rate constant was not determined experimentally for decane but the droplet diameter obtained from one test film had a general tendency to decrease more rapidly than the theoretical value of $K = 1.516 \times 10^{-2} \text{ cm}^2/\text{sec}$. The extreme brightness of the flame obscured the droplet surface in all other tests filmed.

The ratio of the flame to droplet diameter was determined experimentally for heptane in air to vary between 6.2 and 7.2, compared with the theoretical value from a constant property approach of 35. The assumption made in the theoretical approach which resulted in an acceptable value for the burning rate based upon this and other experiments may be less valid when determining flame position.

A phenomenon termed "flash extinction" was observed in two tests with decane. Minute satellite droplets were observed to burn to very small diameters at which point they seemed to explode. These tests were performed in atmospheres consisting of 50% O_2 - 50% N_2 by mass at 1 atmosphere pressure.

The most probable explanation for the flash extinction and the discrepancy between the theoretical and experimental values for the burning rate is the accumulation of hot carbon at or near the surface of the droplet through dissociation of the reaction products. The carbon production would be most severe in the tests with decane where the flash extinction occurred and where experimental-theoretical differences are the greatest. Ignition of the hot carbon at the droplet surface could cause the flash, whereas the increased heat transfer from the carbon to the droplet could increase the burning rate.

Carbon production can be seen then as possibly being a crucial factor in this type of combustion. The burning time, fuel used, and atmospheric content are all factors which affect the production of carbon, whose effect upon the burning rate has not yet been fully demonstrated.

2. List of Symbols

B	Pre-exponential reaction-rate constant
$C_{p,i}$	Specific heat at constant pressure for species i
D_{ij}	Binary diffusion coefficient for species i and j
$D_{T,i}$	Thermal diffusion coefficient for species i
E	Activation energy for a reaction
f_i	External force per unit mass on species i
h_i	Specific enthalpy of species i
h_i^o	Standard heat of formation per unit mass for species i at temperature T^o
m_i	Chemical symbol for species i
N	Total number of species present
P	Pressure tensor
R_o	Universal gas constant
T	Temperature
V_i	Diffusion velocity for species i
W_i	Molecular weight of species i
X_i	Mole fraction of species i
Y_i	Mass fraction of species i

Greek

δ	Flame thickness
----------	-----------------

λ_i	Thermal conductivity of species i
μ_i	Coefficient of viscosity for species i
ν_i	Stoichiometric coefficient for species i appearing as a reactant
ν'_i	Stoichiometric coefficient for species i appearing as a product
ρ	Density
<u>Subscript</u>	
b	Evaluated at boiling conditions
F	Fuel
f	Evaluated at flame conditions
mix	Mixture
O	Oxidizer
∞	Evaluated at ambient conditions

3. Introduction

In the field of combustion, one particular aspect that is of both basic and applied interest concerns the burning of liquid fuel droplets. That droplet burning has been the subject of extensive study over many years is evidenced by the fact that an early review [1] contains more than sixty references. Practical motivations for such investigations come mainly from the desire to achieve clean and efficient production of power through the combustion of liquid fuels. But certain unknown factors still remain at the basis of the subject.

The conservation equations of fluid mechanics describe droplet combustion provided that allowance is made for transport phenomena and for chemical transformations in these equations [2]. Two factors prevent solutions to the equations for specific systems from being obtained readily. One is the complexity of the equations themselves in three-dimensional, time-dependent situations, and the other the fact that certain quantities appearing therein, especially chemical kinetic parameters, usually are not known. Theories for droplet burning have circumvented these two difficulties through the introduction of approximations, of which three are (a) that spherically-symmetrical forms of the conservation equations are applicable in the gas phase, (b) that quasisteady conditions are achieved, in the

sense that steady-state equations can be employed in the gas and droplet properties such as its temperature are invariant with time, and (c) that chemical equilibrium prevails and gives rise to a spherical flame-sheet positioned in the gas. Numerous refinements to the early theoretical considerations of Godsave [3] include the semiempirical treatment of effects of forced and natural convection [4-8].

The experimental study of an individual droplet, however, has usually necessitated the support of the droplet on the end of a fine quartz fiber [3, 4, 9-13], and flows are manifestly non-spherical. The hot flame in a gravitational field causes gases to rise buoyantly, establishing an axisymmetric flow under natural convection, thereby violating approximation (a) above. Attempts to describe the non-spherical flow have proven difficult since the Grashof numbers typically are neither large nor small. It is not clear that studies of burning droplets influenced by buoyancy would be profitable since from a practical viewpoint the droplets in combustion chambers are relatively small and are not supported by fibers, so influences of buoyancy are reduced by droplet motions. From the fundamental viewpoint, underlying physical and chemical phenomena, such as the effects of chemical kinetics, can be studied more readily if the complicating influence of buoyancy is removed.

Minimizing buoyancy has been achieved by working with very

small droplets projected into a hot gas stream [14-16], but individual droplets could not be observed throughout their histories. An alternative approach that has been attempted [4] is to burn droplets at reduced pressure, since the Grashof number decreases with decreasing pressure. It is found, however, that extinction due to finite-rate chemistry generally occurs at pressures above those at which the flame becomes essentially spherical, with suspended droplets. The only other way to decrease buoyancy effects is to reduce gravity.

Previous experiments by Kumagai [17, 18, 19], in which a freely falling chamber eliminated significant influences of gravity for a short period of time, have been successful at achieving spherical symmetry. However, observations showed that the gas flow was unsteady, and the ratio of flame to droplet diameter did not remain constant during burning, which violates approximation (b) above.

This unsteadiness may be attributable to the limited test time of about 1 sec. In none of these tests was sufficient time available to follow the droplet to half its initial diameter, even though only small droplets, less than 1.5 mm in initial diameter, were employed.

Since there seemed to be a tendency for the flame location to approach a quasisteady situation at the end of Kumagai's testing time, by extending the observation time it may be possible to demonstrate that later in its burning history the droplet conforms closely to the basic theory. The work reported herein constitutes an attempt to achieve

such an extension by use of a drop tower having 2.2 sec of negligible gravity.

The objective in use of the drop tower has been to eliminate significant influences of buoyancy and of nonspherical convection for a period of time long enough to allow observation of the entire burning history. In this manner, the combustion can be recorded down to droplet sizes so small that the chemical conversion time in the gaseous flame become comparable with the residence time therein. Under these conditions, approximation (c) above is expected to begin to fail, and in a cool atmosphere droplet extinction theoretically should occur [20]. Measurement of extinction diameters as functions of ambient conditions of pressure and of oxygen content in the atmosphere should provide data on overall rates of oxidation and thereby further the understanding of chemical kinetics of hydrocarbon combustion.

4. Analytical Background

A. Constant Property Theory

We consider then the problem of a liquid fuel droplet burning in an oxidizing atmosphere, as shown in Figure 1(a). Outwardly diffusing fuel vapor reacts stoichiometrically with the inwardly diffusing oxidizer gas at an infinitesimally thin flame front according to the one-step chemical reaction



whose reaction rate is given by

$$\omega = k \prod_{i=1}^N C_i^{\nu_i}$$

and whose specific reaction rate is given by the Arrhenius expression

$$k = B e^{-E/R_o T}$$

The chemical heat released during this reaction is conducted inward to sustain vaporization and transported outward to the ambient, thereby creating two diffusive-convective regions separated by the flame sheet.

The general conservation equations of fluid mechanics are given by Williams [2] as

$$\partial \rho / \partial t + \nabla \cdot (\rho \vec{v}) = 0 \quad \text{Overall continuity} \quad (2)$$

$$\rho \partial u / \partial t + \rho \vec{v} \cdot \nabla u = -\nabla \cdot \vec{q} - \vec{p} : (\nabla \vec{v}) + \rho \sum_{i=1}^N Y_i f_i V_i \quad \text{Energy} \quad (3)$$

$$\partial Y_i / \partial t + \vec{v} \cdot \nabla Y_i = w_i / \rho - [\nabla \cdot (\rho Y_i \vec{V}_i)] / \rho \quad \text{Continuity of species} \quad (4)$$

where the diffusion velocity $\vec{V}_i$ is determined from

$$\begin{aligned} \nabla X_i = & \sum_{j=1}^N \left(\frac{X_i X_j}{D_{ij}} \right) (\vec{V}_j - \vec{V}_i) + (Y_i - X_i) \left(\frac{\nabla p}{p} \right) \\ & + \left(\frac{p}{p} \right) \times \sum_{j=1}^N Y_i Y_j (f_i - f_j) \\ & + \sum_{j=1}^N \left[\left(\frac{X_i X_j}{\rho D_{ij}} \right) \left(\frac{D_{T,j}}{Y_j} - \frac{D_{T,i}}{Y_i} \right) \right] \left(\frac{\nabla T}{T} \right) \quad i=1, \dots, N \end{aligned} \quad (5)$$

For droplet combustion in a falling chamber, several effects contained in these equations appear to be unimportant and will be neglected, including

(a) Body forces (f_i terms)

(b) Soret and Dufour effects ($D_{T,i}$ terms)

(c) Pressure gradient diffusion $[(Y_i - X_i) \nabla p / p]$

By further assuming low speed, steady flow, equal binary diffusion coefficients for all pairs of species, and that the Lewis number is unity, we obtain the Shvab-Zeldovich form of the conservation equations [21,22].

$$\nabla \cdot (\rho \vec{v}) = 0 \quad \text{Overall continuity} \quad (6)$$

$$\begin{aligned} \nabla \cdot \left[\rho \vec{v} \left(\int_{T_0}^T C_p dT \right) - \frac{\lambda}{C_p} \nabla \left(\int_{T_0}^T C_p dT \right) \right] \\ = - \sum_{i=1}^N h_i^0 W_i \quad \text{Energy} \quad (7) \end{aligned}$$

$$\nabla \cdot (\rho \vec{v} Y_i - \rho D \nabla Y_i) = W_i \quad i = 1, \dots, N \quad \text{Continuity of species} \quad (8)$$

where

$$C_p \equiv \sum_{i=1}^N Y_i C_{p,i}$$

By expressing equations (7) and (8) in the form

$$L(\alpha) = \omega$$

where

$$L(\alpha) = \nabla \cdot [\rho \vec{v} \alpha - \rho D \nabla \alpha]$$

and defining a coupling function β_T as

$$\beta_T \equiv \alpha_T - \alpha_O$$

where

$$\alpha_T \equiv \frac{\int_{T_o}^T C_p dT}{\sum_{i=1}^N h_i^o W_i (\nu_i' - \nu_i'')}$$

$$\text{and } \alpha_O = Y_O / W_O (\nu_O'' - \nu_O')$$

the solution of the equation

$$L(\beta_T) = 0 \tag{9}$$

will result in an expression for the burning rate of the droplet, $\dot{m}$, which appears as an eigenvalue of (9).

This analysis, as performed by Williams [2], assumes that the liquid droplet reaches a temperature that is determined by the phase equilibrium condition that the partial pressure of the fuel at the surface must equal the equilibrium vapor pressure of the fuel. This is very nearly equal to the boiling temperature of the liquid. The flame-sheet approximation is introduced which assumes that all

of the oxidizer is consumed instantaneously at the spherical flame surface where the reaction rate is infinite so that $Y_O \equiv 0$ inside the flame, and $Y_F \equiv 0$ outside the flame. Constant C_p and unity Lewis number are also assumed. The mass burning rate is then given as

$$\dot{m} = \left(\frac{4\pi\lambda r_\ell}{C_p} \right) \ln \left\{ 1 + \frac{1}{L} \left[iQ_{F\infty} Y_{O\infty} + C_p (T_\infty - T_b) \right] \right\} \quad (10)$$

where the terms used are described below.

The mass-conservation condition that

$$\dot{m} = -4\pi r_\ell^2 \rho_\ell \frac{dr_\ell}{dt}$$

can be used to express the rate of change of the droplet diameter

$$d(d_\ell^2)/dt = -2\dot{m}/\pi\rho_\ell r_\ell \quad (11)$$

Previous experiments with burning droplets [3, 9, 10] have shown that the square of the droplet diameter decreases linearly with time according to the "d-square law"

$$d_{\ell,o}^2 - d_\ell^2 = K_o(t - t_o) \quad (12)$$

Combining equations (10), (11) and (12) then results in an expression for the burning rate constant K_o ,

$$K_o = \left(\frac{8\lambda}{\rho_l C_p} \right) \ln \left\{ 1 + \frac{1}{L} \left[iQ_F Y_{O\infty} + C_p (T_\infty - T_b) \right] \right\} \quad (13)$$

where i = stoichiometric mass ratio of fuel to oxidizer

Q_F = heat of combustion per unit mass of fuel (lower heating value)

L = heat absorbed per unit mass of liquid fuel vaporized at T_b

C_p = mean specific heat for the fuel vapor

ρ_l = liquid density at T_b

λ = thermal conductivity.

A similar analysis for the flame-surface conditions utilizing equation (9) and $Y_O = 0$ for $r < r_f$, $Y_F = 0$ for $r > r_f$ results in an expression for the ratio of flame-to-droplet diameter,

$$\frac{d_f}{d_l} = \frac{\ln \left\{ 1 + [iQ_F Y_{O\infty} + C_p (T_\infty - T_b)]/L \right\}}{\ln \left\{ 1 + i Y_{O\infty} \right\}} \quad (14)$$

The results of these analyses show that for a given fuel burning in an atmosphere with given initial composition, K_o and the ratio d_f/d_l should remain constant once steady-state conditions are attained. The only question that remains is in the definitions of λ and C_p . Law and Williams [4] have developed a

physically motivated empiricism giving an explicit prescription for evaluating these quantities which gives good agreement with previous experiment. For λ they employed the formula $\lambda = X\lambda_F + (1 - X)\lambda_{\text{mix}}$, where λ_F is the thermal conductivity of the fuel vapor and λ_{mix} is that of the gas mixture excluding the fuel, and X is related to the average fuel mole fraction between the droplet and the flame sheet. The value $X = 0.4$ was chosen for experimental agreement. Here λ and C_p are evaluated at a temperature $T = (T_b + T_F)/2$ where dissociation is taken into account in evaluating T_f . These approximations are used in the work presented here.

One of the effects excluded by the assumptions given above is that of forced convective motion of the gas relative to the droplet. This effect, first analyzed by Frössling for a non-burning vaporizing droplet [24], resulted in the ratio of the vaporization rate $\dot{m}$ to be given as

$$\dot{m}/\dot{m}_0 = 1 + 0.276 \text{Re}_\infty^{1/2} \text{Sc}_\infty^{1/3} \quad (15)$$

Agoston et al. [7] have suggested using Pr_∞ instead of Sc_∞ and further assuming $\text{Pr}_\infty^{1/3} \approx 1$. When applied to (13) this results in $K = K_0(1 + 0.276 \text{Re}_\infty^{1/2})$ and droplet motion can be accounted for in the burning rate.

B. Extinction Criteria

One of the assumptions leading to (10) was that of infinite reaction rates. However, if one wishes to study the effects of finite-rate chemistry and determine reaction orders, the parameter of primary interest is the Damköhler number D_f . As shown in Appendix (C1) this is defined as

$$D_f = \frac{\delta^2 \rho_f^{m+n-1} B}{D_{O_f} W_O^{m-1} W_F^n} \exp \left(\frac{-E}{R_o T_f} \right) \quad (16)$$

and is the ratio of the flow time of oxygen through the diffusion flame of thickness δ , to the chemical time necessary for the oxygen to react to liberate heat. It can be seen that if $D_f \leq 1$, the oxygen will have insufficient time to react and a flame can no longer be sustained. For a burning droplet, this can occur whenever the droplet radius becomes very small so that the flow time (diffusion time) of oxygen becomes small compared with the chemical reaction time. Theoretically then, if a droplet can be observed to burn to a very small radius, extinction could be observed and the system properties used in (16) to determine the reaction orders and chemical kinetics. By varying the system pressure and observing extinction, the overall reaction order could be determined from the pressure dependence of D_f . By varying the O_2/N_2 ratio the overall activation energy E could be determined, since the Damköhler number is

fairly sensitive to the flame temperature which is affected by the concentration of oxygen in the ambient gas. In order to determine the flame thickness δ , one can use the result [8]

$$\delta = (d/2) D_1^{-1(m+n+1)} \quad (17)$$

where

$$D_1 = \frac{d^2 \rho_\infty^{m+n-1} B_\infty}{4 D_{O_\infty} W_O^{m-1} W_F^n} \left(a + b \frac{W_F}{W_O} \right) \quad (18)$$

in which a and b denote stoichiometric coefficients with respect to oxidizer and fuel for the overall reaction. By systematically varying the gas pressure and flame temperature (through the variation of oxygen concentration in the ambient gas), the quantities of interest in (16) can then be determined.

C. Variable Property Theory

With the assumption of infinite reaction rates, so that the chemical time $\tau \rightarrow 0$, it is seen that the flame thickness $\delta \rightarrow 0$ and $D_1 \rightarrow \infty$. When this limit is applied to the properly non-dimensionalized exact governing equations, it leads to a singular perturbation problem. The droplet combustion problem was analyzed by Kassooy and Williams [25, 26] using this technique in order to determine the effects of thermal variations upon the gas properties and the subsequent effects upon the droplet burning rate. Only the

assumptions and results will be summarized here.

The chemical kinetic process is described by a one-step irreversible reaction whose rate is given by an Arrhenius expression as in eq. (1). An average specific heat for all chemical species is used, but the thermal conductivity of the mixture and the binary diffusion coefficient of the i^{th} species into the product of combustion species D_i' , as well as the Lewis number $Le_i = \rho' D_i' C_p' / \lambda'$ in which these quantities appear, are functions of temperature and concentration. The flow field is divided into three regions, denoted by the superscripts $(\sim)$ for the region interior to the flame, $(\wedge)$ for the region exterior to the flame, and $(*)$ for the flame zone itself.

After performing the proper matching for the solutions in these three zones, an additional assumption is made for the functional variation of the transport properties. Thus, the case

$$\hat{\lambda} = \hat{a}' T'^s \quad \tilde{\lambda}' = \tilde{a}' T'^s \quad \lambda'_{\infty} = a'_{\infty} T'^s \quad s = 1$$

is considered. The ratio $\tilde{a}' / \hat{a}'$ is assumed to be 0.8 to account for the presence of fuel vapor in the inner region and its effect upon the thermal conductivity of the mixture. The primed variables indicate dimensional quantities, whereas unprimed variables have been non-dimensionalized with respect to values at ambient (∞) , with the exception of the radial coordinate r which is constructed by division

by the droplet radius. The results of this analysis give the burning rate and flame position as

$$K_o = \frac{8\lambda'_\infty}{\rho'_l C'_{p\infty}} \left\{ \frac{\hat{a}}{a_\infty} (1 - T^*) + \frac{\tilde{a}}{a_\infty} [T^* - \tilde{T}_o(1)] \right. \\ \left. + \hat{C}_2 L\hat{e} \frac{\hat{a}}{a_\infty} \left[1 - \frac{\tilde{C}_2 \tilde{a}}{\hat{C}_2 \hat{a}} \right] \ln(h) + \frac{\tilde{C}_2 \tilde{a}}{a_\infty} \ln(z) \right\} \quad (19)$$

$$r^* = 1 + \frac{(\tilde{a}/a_\infty)[T^* - \tilde{T}_o(1)] - \tilde{C}_2 Le_\infty (\tilde{a}/\hat{a}) \ln(h) + \tilde{C}_2 (\tilde{a}/a_\infty) \ln(z)}{(\hat{a}/a_\infty)[1 - T^* + \hat{C}_2 L\hat{e} \ln(h)]} \quad (20)$$

where

$$T^* = 1 + [1 - \tilde{T}_o(1) + \bar{L} + \alpha_T/\alpha_F][h^{-L\hat{e}} - 1]$$

$$z = [-\alpha_T/\alpha_F(h^{L\hat{e}} - 1) + 1 - \tilde{T}_o(1) + \bar{L}]/\bar{L} \quad \bar{L} = L/C'_p T'_\infty$$

$$h = (\alpha_F/\alpha_o Y_{O_\infty} + 1) \quad \alpha_F = bW_F/(aW_O + bW_F) \quad \alpha_o = 1 - \alpha_F$$

$$\alpha_T = \frac{\Delta H^o/C'_p T'_\infty}{aW_O + bW_F} \quad -\Delta H^o = \text{heat of combustion}$$

$$\tilde{C}_2 = \tilde{T}_o(1) - \bar{L} \quad \hat{C}_2 = \tilde{C}_2 - \alpha_T/\alpha_F \quad \tilde{T}_o(1) = \text{droplet surface temp.}$$

As stated above, it was necessary to make the assumption

of a constant coefficient in the power law expression for the thermal variation of λ , where the constant can have different values in the two diffusive regions. One deficiency existing in this theory is that the concentration dependence is suppressed, which could be an unrealistic assumption for a gas mixture consisting of species with widely different molecular structures and weights as is the case with hydrocarbon droplet combustion.

A theory which accounts for the temperature and concentration dependence of the various coefficients has been developed [27] in which a reduced diffusion coefficient ρD , as well as λ are functions of a non-dimensionalized radial distance and of temperature. The thermal dependence is assumed to be of a simple power-law form. With the further assumptions that the specific heat of each species is temperature dependent, and that frozen equilibrium exists at the flame sheet, a general expression for the mass burning rate and flame position was developed. However, the solution of the expression involves numerical evaluation of a complex integral, which was performed by Law and Law with the simplification that the concentration dependence of C_i was suppressed, for the case of a heptane droplet burning in air. A constant coefficient was also assumed for the power-law dependence of ρD , and the coefficient was treated as a free parameter by allowing it to vary within reasonable bounds.

The results of this theory for the specific case of a heptane droplet burning in air gave a flame temperature which was uniformly too high for the assumption of frozen equilibrium to be valid. The difference between the theory and previous experimental results [18] is highly dependent upon the value chosen for coefficient of the thermal dependence of ρD .

Because this theory is not entirely general in that the approximations cited above have been made, and because application of the theory would be appreciably more complex, in the present work effects of variable properties are studied on the basis of the theory of Kassoy and Williams [25, 26]. Although the resulting analysis may lack the desired degree of generality, it should at least provide an indication of the influence of variable properties.

One cannot verify both the constant property and the variable property theories, but one can determine which theory seems to best describe the phenomenon, based upon experimental results. In order to do this, it is necessary to observe the actual process in an environment which can be carefully controlled to accurately define the quantities appearing in the expressions above.

5. Experimental Apparatus and Procedure

In order to study the burning rates of spherical droplets, a method must be devised to confine the droplet to a pre-specified location without there being solid objects (e.g., glass fibers) near it to destroy the spherical symmetry. Various methods have been used for positioning liquid droplets, but many require large power supplies, while others require a considerable length of time to accomplish positioning [21]. Kumagai et al. [18, 19] have developed a method which requires little equipment and is fairly reliable, and it is upon this basic design that the present system has been constructed.

The test apparatus consists of a metal framework which contains the test chamber, ignition system, batteries, timers and high-speed motion-picture camera as shown in Figure 2. The framework, or drop frame, is a 30" x 15" x 15" aluminum structure designed to withstand a 30-g. impact upon completion of each test. During a test, it acts as a completely separate unit with nothing to interfere with its free-fall condition. The drop frame is used in the NASA-Lewis Research Center's Zero-Gravity facility which is capable of providing 2.2 seconds of free-fall with drag forces to 10^{-6} g.

Since the tests performed involved burning in oxygen or oxygen-nitrogen atmospheres ranging in pressure from 0.1 to

2 atm., it was necessary to build a test chamber within the drop frame containing the droplet deployment and ignition systems. The chamber is a 12.5" x 10" x 11" box made of 3/4" plexiglas, fitted with an o-ring at its base to provide a seal against the chamber bottom. The chamber bottom is rigidly fixed to the drop-frame, and access is gained to the experiment by removal of the box. During a test, the chamber is held against the bottom through the use of 4 stainless steel threaded rods and 2 cross-bars which exert a downward force on the box. The use of clear plexiglas enables the camera to film the burning within the chamber and the operator to see the internal mechanism in order to deploy the droplet.

The droplet release mechanism, located within the chamber, is the heart of the apparatus. As shown in Figure 3, it consists of an aluminum "piston" which is contained in a 0.825" I.D. cylinder. Into the piston are inserted a brass rod, $1\frac{1}{2}$ " long, and a 2 mm max. dia. quartz fiber. The piston rests against a helical spring, and when the spring is compressed, is held in place by an electromagnetically actuated catch. A microswitch on an adjustable support is used to activate the ignition source. The ignition electrodes consist of fine copper wires, spaced about 1.5 mm apart, located at the tip of the quartz fiber. The electrodes are mounted on a sliding track which is held in place by a second electromagnetically actuated catch against the tension of a stretched helical spring.

The release mechanism uses the motion of the droplet to enable the release of the drop frame to be timed properly. The piston is pushed against the spring and held by the catch. After a droplet is mounted on the fiber, the first relay is energized. The piston begins an upward motion, but its acceleration is great enough to pull the fiber out of the droplet. The droplet continues moving upwards, but is now being decelerated by gravity. The droplet reaches a point where it has no vertical velocity which occurs approximately 0.02 sec. after release. At that time, the piston which was still moving upwards trips the micro-switch which energizes the ignition circuit and the second electromagnet. As a consequence of the resulting catch release, the electrodes then begin moving away from the droplet. At the same instant that the droplet reaches the high point of its trajectory, and zero velocity, the drop frame is released into free fall. In this manner the droplet is stationary with respect to the camera mounted on the drop frame, no physical objects are supporting it, and it is now in free-fall.

Since the release mechanism is located inside the pressure chamber, a means for mounting the droplet by remote control is needed. Figure 4 shows the droplet mounting mechanism. It consists of a hypodermic syringe activated by two small d-c motors. One motor drives the syringe, which can be moved to bring the needle tip in contact with the quartz fiber. The second motor, through a 9780:1

gear reduction, slowly moves the plunger in to force the liquid fuel out the needle tip. A droplet is then transferred to the fiber. The droplet size can be controlled fairly easily due to the fine control provided by the gear reduction, and by the use of a fine-gauge syringe needle (25 gauge). The syringe is then withdrawn slowly from the fiber and the droplet remains on the fiber. The syringe is then moved back away from the combustion zone.

The ignition system consists of an automotive induction coil for the high secondary voltage needed for the spark gap, a 4300 μF 50 Vdc capacitor which is discharged through the coil's primary windings, and the control switches for the spark timing. Power was supplied by three 15 Vdc nickel-cadmium batteries.

The camera used for the tests was a 16 mm Millikan running at a framing rate of 400 PPS. The camera was controlled through a set of relays in the drop frame control box, and powered by two 15 Vdc nickel cadmium batteries. Lighting was also provided through the control box, as was the power for the droplet release mechanism. Wiring enters the chamber through pressure fittings in the bottom.

At the beginning of a test, the syringe is filled with 0.25 cm^3 of fuel, the piston is "cocked" into position, as are the electrodes, and the syringe is moved forward until the needle tip seals in the R.T.V. stopper shown in Figure 3. The chamber top

is put in place and secured, and the chamber is evacuated. It is then refilled slowly with N_2 until a specified partial pressure is reached, and then filled with O_2 until the test pressure is reached. The pressure is monitored to ensure that no over-pressure develops if throttling has occurred while filling the chamber.

The drop frame is then placed within an air-drag shield (Figure 2) whose purpose is to shield the frame against air resistance. The drag shield moves separately from the drop frame and during the course of a test will move 8" relative to the frame due to air resistance on the shield. At the base of the drag shield are penetration spikes which are designed to absorb energy as the package strikes the deceleration tank — a 6 ft. deep box of aerated sand. Access doors to the test package are mounted on the drag shield.

The entire test package is hoisted to the top of an eight-story tower where the package release mechanism and timers are located. The package is suspended by an over-stressed music wire which is passed through a cutter assembly. The control box relay switches are connected to the main control console through umbilical cables, and the normally-closed relays on the drop frame control box are energized (Figure 5). All package switches are then placed in the "ON" position but due to the opened relays, no mechanisms operate. The droplet is then mounted by remote control. To initiate the test run, the automatic timers are set on the main control console,

and the start button is activated. The camera circuit closes first, starting the camera. After 2.0 sec., the droplet release electromagnet circuit is closed, releasing the droplet. In another 0.02 sec, the cutter is activated which releases the test package into free-fall, and the spark circuit is energized igniting the droplet. After 2.2 sec, the package experiences the 30-g. impact and the control box shuts off power to the various circuits. The package is then retrieved and prepared for the next run.

6. Experimental Results

A. General Observations

Droplet burning in the absence of buoyancy was achieved in twenty-two tests using the apparatus described above. In tests where decane was used as the fuel, a highly luminous flame surrounded the droplet, which at oxygen mass fractions equal to or greater than 30% was so intense that the surface of the droplet was not visible. Tests with heptane produced a much less luminous flame but the droplet surface was still obscured when the oxygen mass fraction exceeded 50 %.

The release of the droplet by the withdrawal of the quartz fiber caused the droplets to oscillate, but these oscillations damped out within approximately 0.1 seconds. However, during the period of oscillation the larger droplets, with diameters greater than 1.5 mm, appeared to expel minute glowing particles which traveled beyond the flame sheet until they lost their incandescence and were no longer visible.

The spark discharge used for ignition was of such high intensity that it ejected some material from the main droplet and formed a small satellite droplet in three tests using decane. These small satellites were observed to burn to very small diameters without experiencing extinction in the manner described above.

Because of the highly critical timing needed in this experiment in order to obtain a stationary droplet, it was anticipated that many tests would result in droplets moving through the chamber until the timing was properly adjusted so that the drop frame was released into free-fall whenever the droplet had zero velocity relative to the frame itself. In the thirty-seven tests performed, it was not possible to obtain the timing accuracy needed so that every test performed resulted in a droplet moving with a given velocity through the chamber. This in turn distorted the flame shape such as is shown in Figure 1(b), and in extreme cases where the timing error was very large the droplet attained a velocity where burning could no longer be sustained.

The droplet burning described above occurred in atmospheres consisting of 20, 25, 30, 50 and 80 percent oxygen diluted with nitrogen at pressures of $1/2$ and 1 atmosphere. In fifteen other tests, ignition was not achieved due to malfunction of the ignition or release systems. Because of the droplet motion relative to the camera, only five tests provided film of droplets burning for periods up to 0.5 seconds. In two other tests, the droplets bounced off of the chamber walls so that they remained in the camera field of view for most of the 2.2 seconds of free-fall. When passing near any object in the chamber, the flame was distorted and appeared less intense than the flame with no solid objects nearby, indicating the action of the object as a heat sink which reduced the temperature of the flame

in that particular region. One of these tests provided data with no object close to the flame for 2.16 seconds.

B. Method of Data Reduction

The film of each test was viewed under a Vanguard Motion Picture Analyzer which is equipped with cross-hairs which are moveable and can be placed over any position on the viewing screen. A vernier scale on the analyzer corresponds to the cross-hair position, so that the size of an object can be recorded by lining the cross-hair over the proper position on the screen and recording the scale reading.

Since the droplets were small, and obscured by the flame in many tests, four readings were taken at each data point corresponding to the maximum probable diameter and the minimum probable diameter. The ambiguity in the flame or droplet position was enhanced by the graininess of the film under the high magnification used for viewing. The vernier scale reading from the analyzer was then converted to true size by using a fixed reference on the film, the syringe needle, as a scaling constant. The time reference was provided by the framing rate of the camera, 400 PPS, and verified by timing marks on the film edge. In this manner, the droplet or flame diameter could be plotted as a function of time.

Data from two tests on the droplet diameter variation with

time were plotted using the maximum and minimum points, and a linear regression analysis was used to determine an experimental slope of the square of the droplet diameter versus time. Data from one test on the variation of the flame diameter with time was also plotted using the same technique to determine the maximum and minimum probable diameter. In this case, the diameter was actually the minor axis of the ellipsoid formed by the flame due to the forced convective effect.

C. Theoretical Burning Rate

In order to determine which of the theories presented best describes the droplet burning problem, it is necessary to compute the theoretical burning rates from the expressions given above and compare them to the rate determined from the corresponding test. The evaluation of the quantities appearing in Eqs. (13) and (15) are described in Appendix (C2), with the results that the burning rate from Eq. (13) are:

$$K_o = 7.448 \times 10^{-3} \text{ cm}^2/\text{sec} \quad \text{n-heptane burning in air}$$

$$K_o = 1.516 \times 10^{-2} \text{ cm}^2/\text{sec} \quad \text{decane burning in 50\% O}_2\text{-50\% N}_2$$

The evaluation of the quantities appearing in Eq. (19), the result of the variable property approach, proved to be highly sensitive to the temperature at which those properties were evaluated.

The burning rate was evaluated for the two cases shown above by computing $L\hat{e}$ at eight different temperatures which could be found in the region exterior to the flame from T'_∞ to T'_f . For each of these cases, the quantities λ'_∞ , $C'_{p\infty}$ were evaluated at T'_∞ and at $T' = (T'_\infty + T'_f)/2$. Then $L\hat{e}$ was evaluated at T'_∞ while C'_p was evaluated at $T' = (T'_b + T'_f)/2$ and at T'_∞ and these quantities used in Eq. (19) to determine the burning rate. Finally, $C'_{p\infty}$ was chosen to be that of the ambient gas mixture and not an average for all species, and the burning rate computed again. This provided a total of 19 cases to be evaluated for each fuel in order to determine the temperature at which agreement between the theory and experiment would be best. The maximum and minimum values for K_o using this type of approach are:

$$\left. \begin{array}{l} K_o = 6.139 \times 10^{-1} \text{ cm}^2/\text{sec} \\ K_o = 2.123 \times 10^{-2} \text{ cm}^2/\text{sec} \end{array} \right\} \text{decane burning in 50\% O}_2 \text{ - 50\% N}_2$$

$$\left. \begin{array}{l} K_o = 7.263 \times 10^{-2} \text{ cm}^2/\text{sec} \\ K_o = 2.193 \times 10^{-2} \text{ cm}^2/\text{sec} \end{array} \right\} \text{heptane burning in air}$$

It can be seen that there is a substantial variation in the burning rate using this approach depending upon the evaluation of $L\hat{e}$, λ'_∞ , $C'_{p\infty}$. As will be shown, the constant property results

appeared to agree more closely with the experimental results of the present work based upon the limited amount of data available. Previous experiments [4, 17, 18, 19] achieved fairly close agreement with the constant property theory, and it is felt that at present this theory provides the best basis for theoretical comparison. The theoretical results shown below will be based upon the constant property theory.

D. Experimental Burning Rates

Figure 7 shows the theoretical and experimental results for n-heptane burning in air at 1 atmosphere pressure and a temperature of 20 °C. The experimental value of $K = 1.775 \times 10^{-2} \text{ cm}^2/\text{sec}$ is based upon a linear regression of the maximum and minimum data points shown in the figure. The time shown is the period from 1.36 to 2.16 seconds after ignition so that the gas phase reaction has had sufficient time to reach a quasi-steady condition. The droplet was moving through the chamber with a constant velocity of 3.6 cm/sec, which caused the flame to appear as in Figure 1(b).

In order to determine the theoretical value of the burning rate with the correction for forced convection, the Reynolds number was evaluated at each data point. The diameter of the droplet was then computed using the expression

$$d_l^2 = d_o^2 - K_o \left(1 + 0.276 \operatorname{Re}_\infty^{\frac{1}{2}} \right) t$$

The theoretical value was computed in this manner at each point using the experimental value of the droplet diameter to compute Re_∞ . A "theoretical point" was obtained corresponding to each data point, and a linear regression analysis performed in order to obtain the slope of this curve. This slope was the theoretical burning rate shown, $K = 1.083 \times 10^{-2} \text{ cm}^2/\text{sec}$.

Figure 8 shows the results for a decane droplet burning in a 50% O_2 - 50% N_2 mixture at 23.5 °C and 1 atmosphere pressure. This droplet was a minute satellite ejected from the main droplet by the spark discharge, and was moving with a velocity of 0.8 cm/sec. The satellite existed for only 0.01 seconds before the flame was no longer visible; hence the few number of data points. Also because the droplet was so minute (0.6 mm maximum diameter), it was very difficult to determine the location of the droplet surface within the flame due to graininess of the film. The apparent curvature of the data points may indicate that the droplet was in an unsteady heating stage since the data begins less than 0.01 seconds after ignition. However, it is also possible that there is extreme scatter in the points due to the difficulty in determining the droplet size from the film. No experimental burning rate is presented for decane because

the droplet was visible in only this test, and the factors mentioned above make an experimental determination highly subjective.

E. Flame Diameter

The flame diameter shown in Figure 9 for a decane satellite burning in a 50% O_2 - 50% N_2 mixture is actually the minor axis of the ellipsoid formed by the flame. The data covers the period from 0.1 to 0.34 seconds after ignition. The satellite was moving with a velocity of 10.9 cm/sec, but the flame was not highly distorted due possibly to the low Re_∞ associated with this droplet of 0.383.

As predicted by Eq. (14), the ratio of the flame to droplet diameter should remain constant once quasi-steady state conditions are achieved. The curvature evidenced in Figure 9 may indicate that the droplet was not at this state. In the test with heptane presented in Figure 7, the ratio of the flame to droplet diameter varied between 6.2 and 7.2, based upon the minor axis. The value predicted by Eq. (14) is 35, which is substantially greater than the experimental observation. The ratio of the major axis to the droplet diameter varied between 10.0 and 12.1 which is still appreciably lower than the theoretical value. In previous experiments on n-heptane burning in air in the absence of buoyancy [18], the ratio of the flame to droplet diameter was found to vary between 4.5 and 6.8, which agrees fairly well with the present result upon the minor

axis.

F. Forced Convective Extinction

Because of the timing error in the package release into free-fall, many droplets achieved substantial velocities relative to the drop frame. At a large velocity, burning could not be sustained and the flame extinguished. Figure 10 shows $(Re_\infty)^{\frac{1}{2}}$ at extinction for n-heptane at various atmospheric compositions. It is interesting to note that the velocity at extinction for $Y_{O_2}/Y_{N_2} = 0.25$ at 1 atmosphere pressure was 16 cm/sec, whereas corresponding results for suspended heptane droplets at 1 g achieved imposed convective velocities above 30 cm/sec without extinction [1]. The supporting fiber may help to stabilize the flame in the earlier experiments. The increase in Re_∞ at extinction as Y_{O_2}/Y_{N_2} increases is qualitatively consistent with theory in view of the greatly reduced chemical reaction time at the elevated flame temperature of the enriched atmosphere.

G. Flash Extinction

As stated above, no droplet was observed to experience extinction in the classical sense. However, two tests with decane burning in a 50% O_2 - 50% N_2 mixture resulted in an interesting phenomenon which we have termed "flash extinction".

Figure 11 shows a detail from the film of one of those tests, which corresponds to the flame diameter data of Figure 9. As can

be seen, the satellite burned to a small diameter and then seemed to explode. A phenomenon of somewhat similar appearance was observed by Dryer [22] in his work with emulsified fuels, which he termed microexplosions. This behavior was not observed with heptane in any test, including burning in an 80% O_2 - 20% N_2 mixture, in the present set of experiments. It can be seen from Figure 9 that the diameter of the flame decreased sharply immediately prior to the flash extinction. This type of phenomenon appears likely to necessitate the presence of impurities in the decane whose relative concentrations increase during burning; these impurities may be present initially or perhaps more likely may accumulate during the combustion process.

The presence of hot carbon particles in and around the droplet surface could facilitate this type of phenomenon. In the fuel-rich region interior to the flame, solid carbon particles are produced in the combustion process through pyrolysis reactions at the elevated temperatures found at the flame sheet. The production of hot carbon would increase as the flame temperature increased and as the number of carbon atoms initially present increased, which is the situation occurring whenever a heavier hydrocarbon fuel with a high heat of combustion burns in an oxygen-rich atmosphere. In our tests this corresponds to decane burning in a 50% O_2 - 50% N_2 mixture. This increase in hot carbon production is qualitatively consistent

C-2

with the increased luminosity observed with the decane burning since the presence of carbon is primarily responsible for the flame's luminosity.

The hot carbon particles could be transported by thermophoresis towards the cooler regions near the droplet surface. The collisions between the gas molecules in the region interior to the flame and the carbon particle would result in a transfer of heat to the gas. The elevated temperature associated with this heat transfer could increase the mass evaporation rate of the droplet since if the droplet is at a constant uniform temperature the heat conducted into the droplet must equal that needed to vaporize the fuel leaving the droplet. If the distance the carbon must travel before reaching the droplet is decreased, the heat transfer to the droplet through this effect would increase. As predicted by Eq. (14), the distance the carbon would have to travel between the flame sheet and the droplet, would decrease as the droplet radius decreased or as Y_{O_∞} increased. The flash extinction occurred with minute decane satellites with $Y_{O_\infty} = 0.5$ which correspond to a minimum distance between the flame and droplet for all the tests performed. The sharp decrease in the flame diameter mentioned could result from a rapid decrease in droplet radius which would only serve to increase the heat transfer to the droplet since the flame stand-off distance would decrease. Eventually a point could be reached where the droplet temperature

would increase beyond the normal boiling point and the droplet would superheat, causing the explosion.

A more likely explanation for the flash extinction could be the result of hot carbon reaching the droplet surface itself. As the combustion process continued, the concentration of hot carbon in the droplet would increase. As the flame diameter decreased, the heat transfer by radiation and conduction to the carbon would increase, increasing the mass evaporation rate of the droplet. The energy transfer to the carbon could become sufficient to ignite the particles themselves if enough oxygen were present for combustion to occur. With the flame sheet approximation that $Y_O \equiv 0$ inside the flame, it would not appear likely that the carbon would ignite close to the surface of a large droplet. But for a small droplet where the flame sheet is positioned physically not far from the droplet and with the assumption of finite-rate kinetics so that oxygen can diffuse into the region interior to the flame as described in section 4-B, it would be possible for the carbon particles to ignite. This ignition of the carbon would result in a violent superheating of the droplet which would occur very suddenly whenever the mass fraction of oxygen present at the flame's interior reached a value sufficient for the carbon particles, generally sub-micron in size, to burn. The close proximity of the flame would enhance this ignition process.

7. Summary and Conclusions

A. Explanation of Experimental Deviations from Theory

The data presented in Figures 7 and 8 show an experimental burning rate that is greater than the constant-property theoretical rate, a difference which seems greater for decane in an oxygen-rich atmosphere than for heptane in air. To account for this trend it would appear that a phenomenon is occurring which depends upon the energy involved in the reaction, hence the fuel and atmosphere, the droplet size, and the length of burning time.

Kumagai [18, 19] achieved very good agreement with theory in his experiment with heptane burning in air where he measured a burning rate constant of $7.8 \times 10^{-3} \text{ cm}^2/\text{sec}$. His time available for burning however was only 0.5 seconds which is less than the time at which the data in Figure 7 was begun. The burning time may then be a crucial point in determining a burning rate since there is a difference between the observed burning rates in these experiments which cover two different time periods.

This behavior appears to be consistent with the plausible explanation for the flash extinction which was accumulation of hot carbon at or near the droplet surface. The accumulation of hot carbon would depend upon the burning time and upon the fuel itself. The disagreement between theory and experiment is most severe in

the decane test in the oxygen-rich atmosphere, where one would expect the carbon production to be most severe. For heptane burning in air, the rate of carbon production is much less than that of decane in a 50% O_2 - 50% N_2 mixture, but if a heptane droplet burns for a considerable length of time a substantial amount of carbon could accumulate in the region near the droplet. The high carbon production as well as the small droplet size in the decane test discussed are conditions where the effects upon the burning rate would be most severe. It is in precisely that test where the experimental burning rate exceeds the theoretical rate by the greatest amount. The increased heat transfer to the droplet would increase the mass evaporation rate and hence the burning rate. The carbon production could be the reason then for both the increased burning rate observed and the flash extinction.

At the elevated temperatures found interior to the flame sheet, pyrolysis of the fuel vapor may become a factor affecting the burning rate of the droplet. As described in various works upon hydrocarbon pyrolysis [29, 30, 31], typical reaction times for paraffin pyrolysis range from 0.01 to 1 seconds with some reactions occurring in a time scale on the order of $10 \mu s$. This is within the diffusion time associated with the fuel vapor in the inner region where diffusion coefficients are typically $1.3 \text{ cm}^2/\text{sec}$ and distances are typically 0.01 cm^2 .

Linear saturated hydrocarbons decompose by chain reactions

initiated either by unimolecular decomposition of the parent molecule into two radicals or by oxygen attack on the hydrocarbon, in each case followed by other elementary reactions. As the hydrocarbon becomes larger, the number and variety of radicals increase. Although these radicals are generally more reactive than the parent molecule, the assumption of infinite reaction-rates used in the derivation of Eq. (13), which seems to be a reasonable approximation during most of the burning history, precludes any effect of the radicals upon the reaction rate. However since the process is diffusion controlled, the presence of species different from the parent molecule can change the thermal conductivity and specific heat of the mixture, and hence K_o . The pyrolysis would once again be most severe at the higher temperatures associated with decane combustion, and would serve to increase K_o through an increase in λ and a decrease in C_p . This could contribute to the difference between the experimental and theoretical values of the burning rate, but would not explain the flash extinction.

It seems probable then that hot carbon production through dissociation is primarily responsible for the experimental-theoretical difference as well as the flash extinction. The carbon would accumulate in time and therefore its effects would increase in time. This would explain why the data of Kumagai agreed with theory for his test with heptane, since his burning time was short and there was little

time for carbon to accumulate. The data presented here began much later after ignition so that much more carbon could have accumulated which would affect the burning rate as described above.

The variable property theory described above did not agree as well as the constant property theory, and in view of the difficulties encountered with this analysis it was not used. It would seem however that a theory which could account for the variation in gas properties due to thermal and concentration gradients, and yet not be prohibitively complex, is desired. However it does not seem possible at this time that one can have both; by necessity a theory which can account for most effects would be quite complex. The variable property analysis shown above demonstrated the wide variation in the burning rate possible through allowing thermal variations alone.

It is seen that the burning rate increases for a given fuel as the oxygen content in the ambient gas increases, according to Eq. (13). Although it was not possible to observe the droplet surface on tests at different atmospheric constituents for a given fuel, the results obtained showing decane in 50% O_2 with a higher burning rate than heptane in air is in qualitative agreement with this theory.

The effect of ambient pressure upon the burning rate was also not demonstrated, but its effect was seen upon the extinction Reynolds number as shown in Figure 10, where a reduction in ambient pressure was accompanied by a reduction in the Reynolds number.

This is consistent with theory as evidenced in Appendix A where the chemical reaction time is increased as the pressure is reduced so that extinction would occur earlier at the reduced pressure.

The flame diameters observed were substantially less than the theoretical value, but were in agreement somewhat with the data of Kumagai [18]. It would appear that the assumptions made in the analysis which led to Eq. (14) may lead to an overestimate of the flame diameter. It is interesting to note that the ratio of flame to droplet diameter predicted by Eq. (20), the variable property result, for heptane in air is 10.2 which agrees more closely with the observed value. The region around the flame where the temperatures are maximum would be the region where thermal effects would be greatest. The assumption of unity Lewis number in the determination of the flame position as well as the assumption of constant properties could be responsible for the overestimate.

B. Suggestions for Future Work

The work described herein is really only the beginning of a more extensive test program aimed at further understanding this phenomenon. But several results useful for future work have been shown. In order to effectively study the effects as described above, a purely stationary droplet must burn in the camera field for the full 2.2 seconds of free-fall. To observe extinction in that time, one

must begin with a droplet whose initial diameter is 0.5 mm or less. The critical problem in achieving a stationary droplet with the apparatus described is in the timing accuracy in the release system. At the present time, the release system for the droplet is accurate to within ± 3 milliseconds; the package release system is accurate to within ± 33 milliseconds. Reproducibilities in timing of about 2 milliseconds are needed if stationary, burning droplets are to be observed.

The droplet mounting system seems very reliable and is an efficient method of producing and placing single droplets on the fiber. The ignition system however supplies too much energy as presently designed and consequently induces motion in the droplet. A reduced spark discharge properly timed could be used to arrest the droplet motion.

Because of the brightness of the flames associated with burning in oxygen rich atmospheres, the use of filters is suggested in order to view the droplet surface. This is extremely important since the lighting conditions without filters may make viewing the droplet impossible.

A systematic method of varying the pressure and O_2/N_2 ratio of the ambient gas would provide much useful data on reaction orders and overall activation energies. A program covering a grid of data points for different fuels, to clearly demonstrate the effect of

each parameter upon each fuel, could greatly enhance the understanding of droplet combustion.

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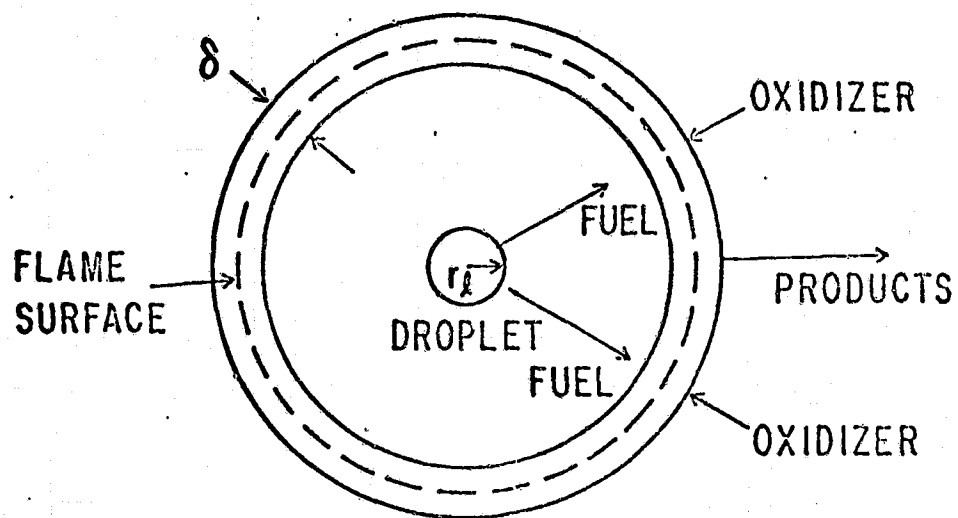
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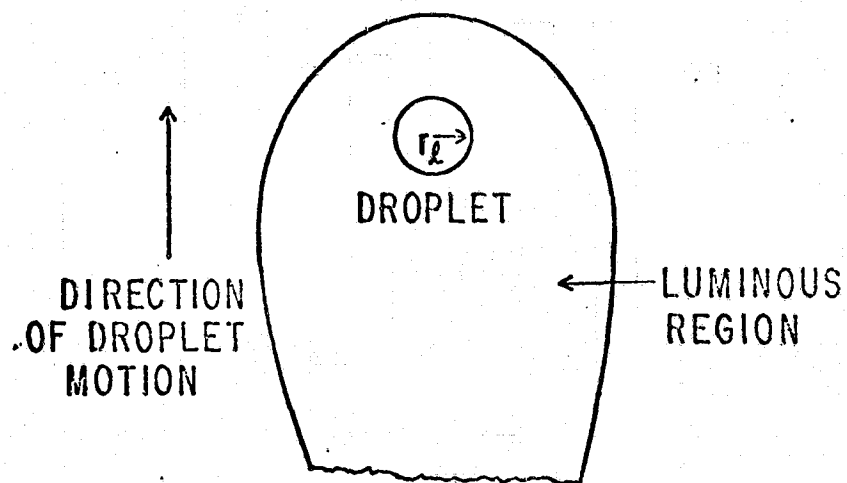
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9. List of Figures

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(a) MODEL OF DROPLET COMBUSTION



(b) FLAME AROUND MOVING DROPLET

Fig. 1

DROP ASSEMBLY

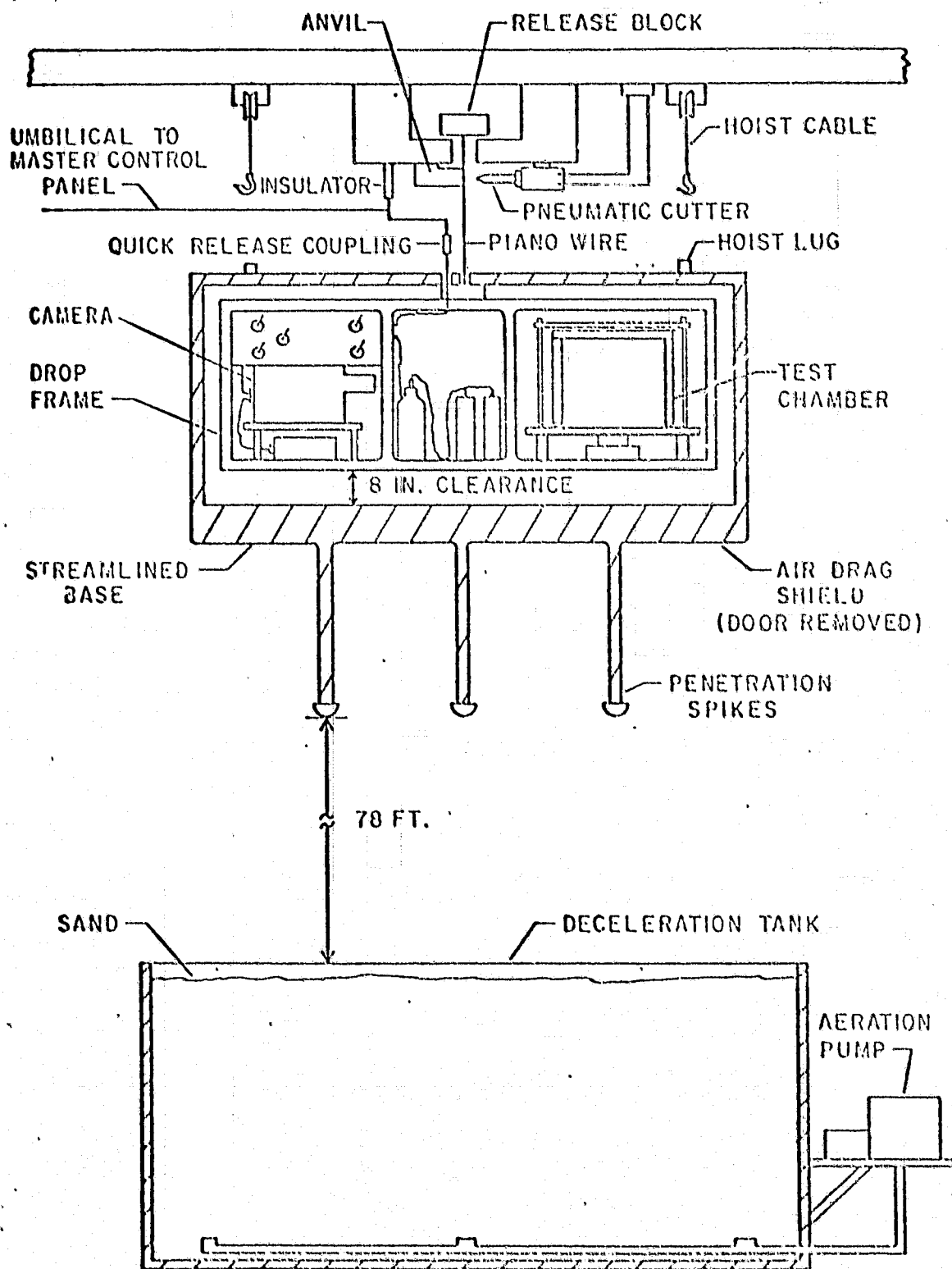


Fig. 2

DROPLET RELEASE MECHANISM

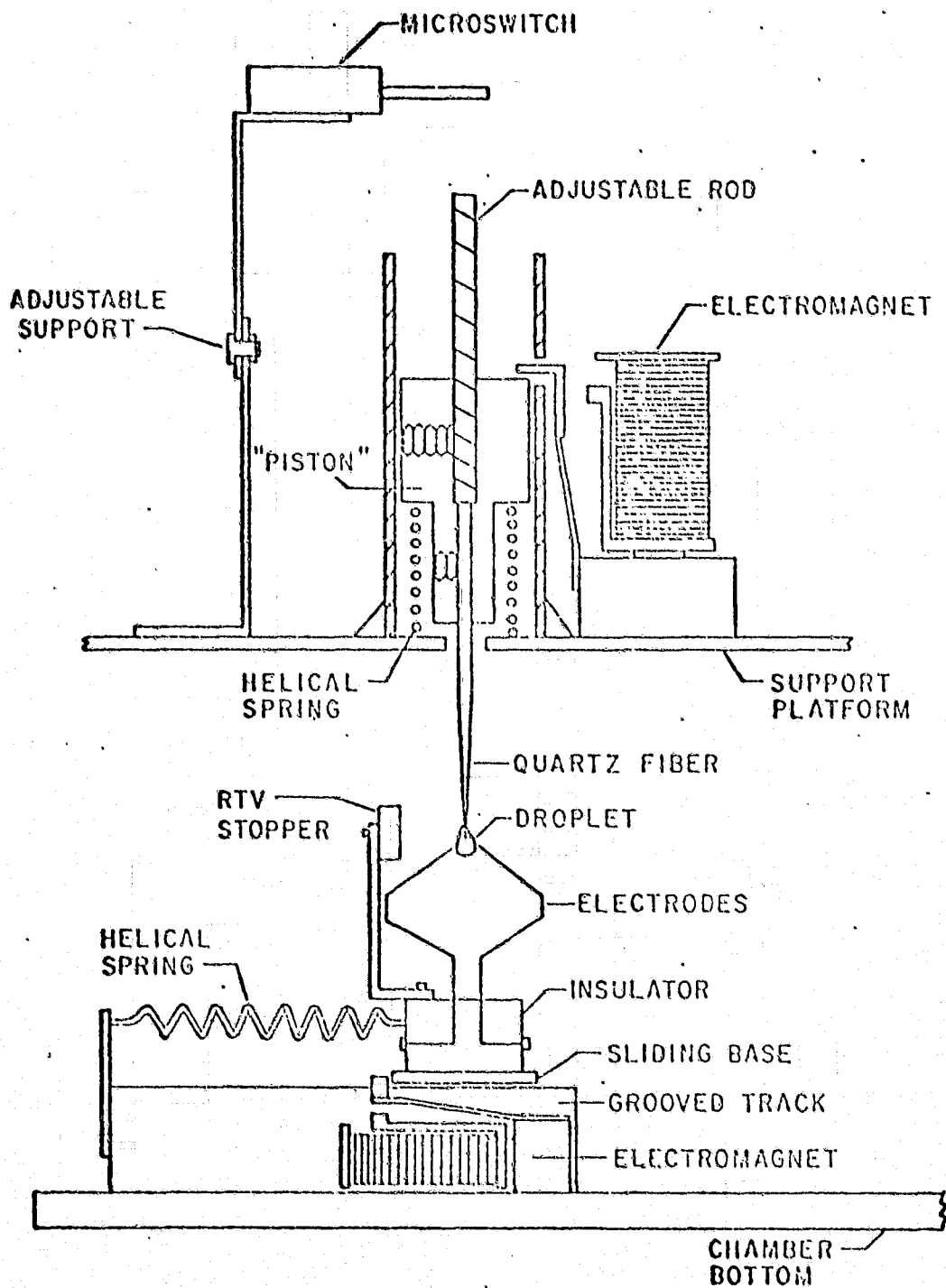


Fig. 3

DROPLET MOUNTING MECHANISM

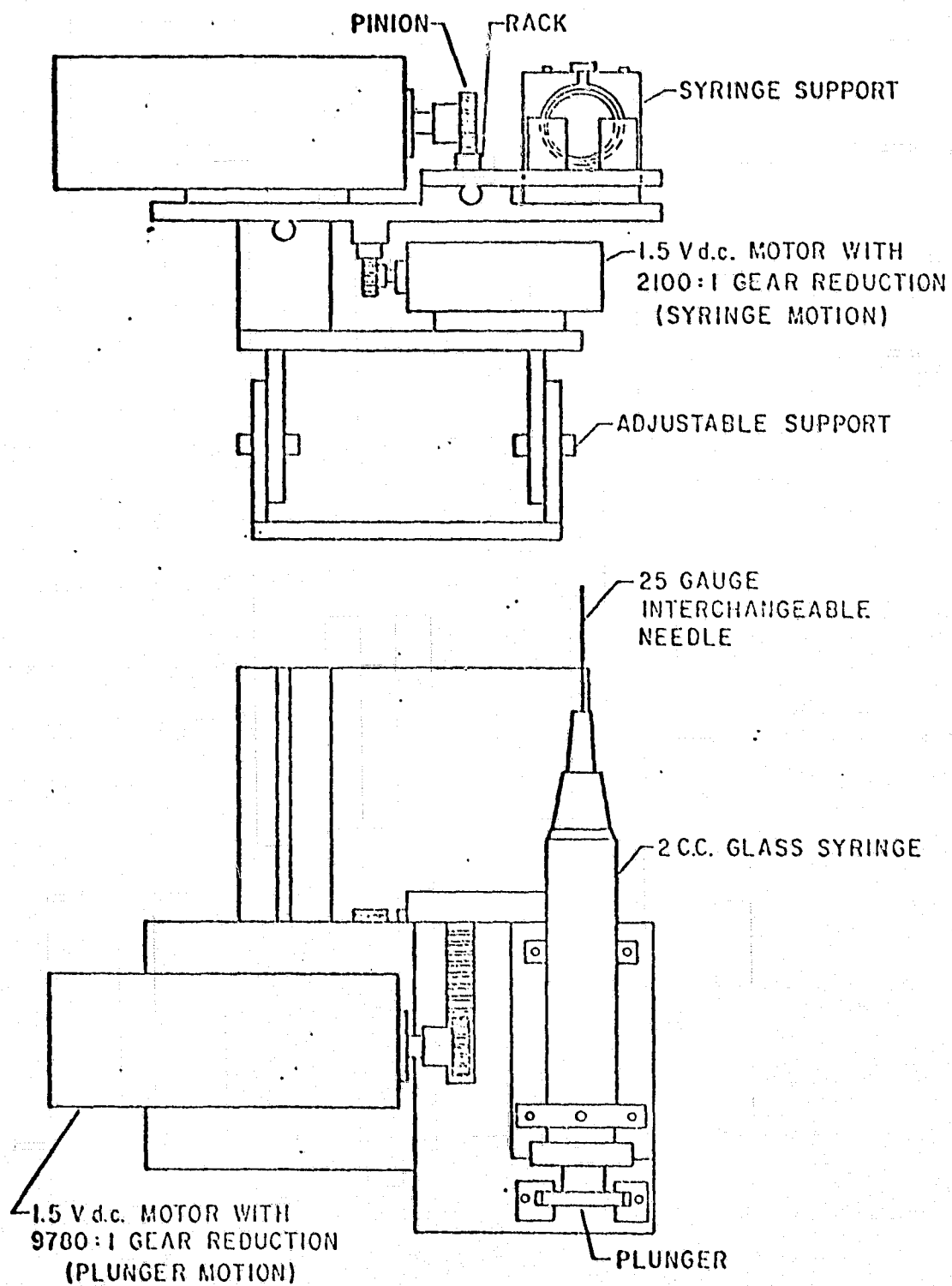


Fig. 4

CONTROL SYSTEM

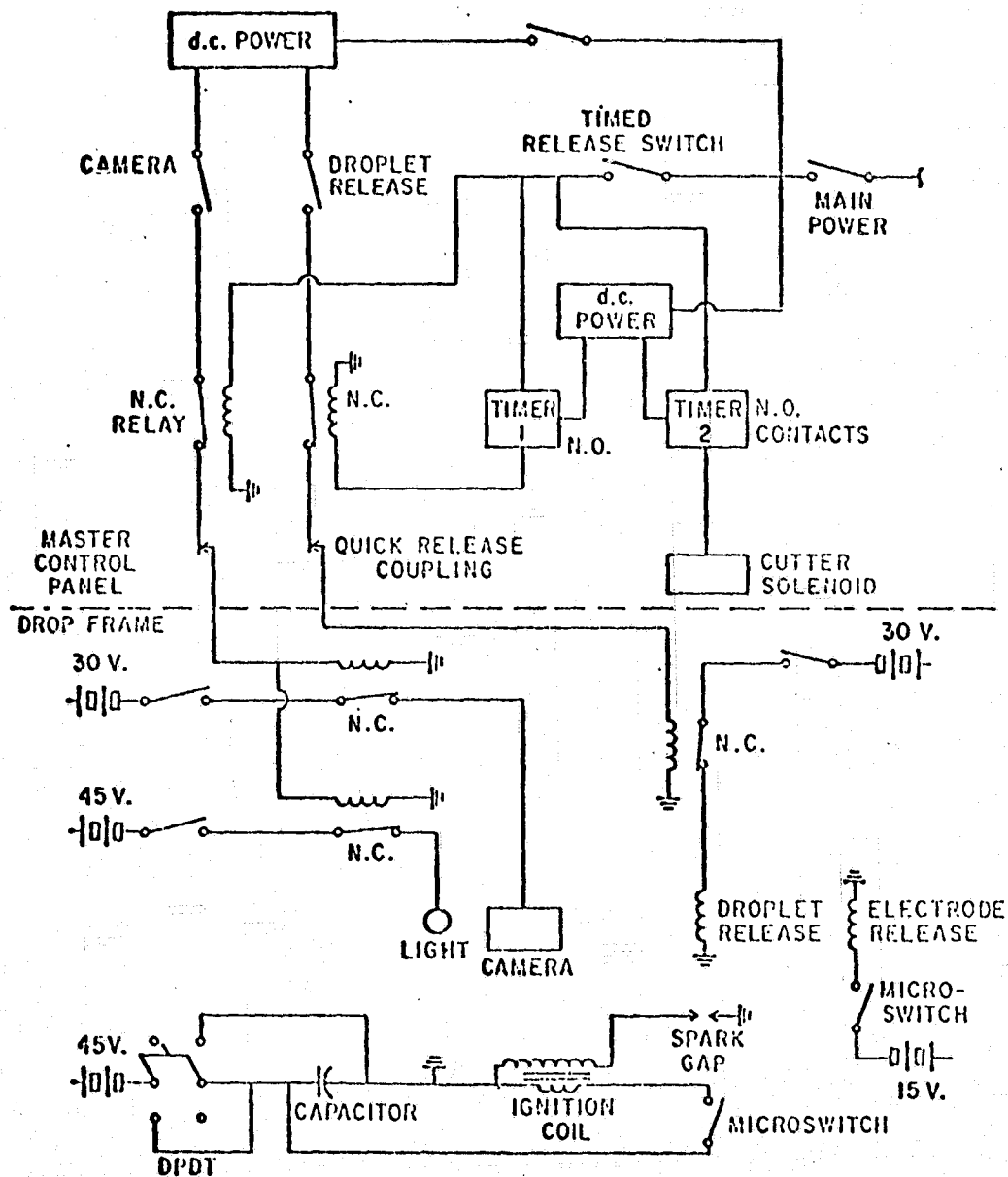
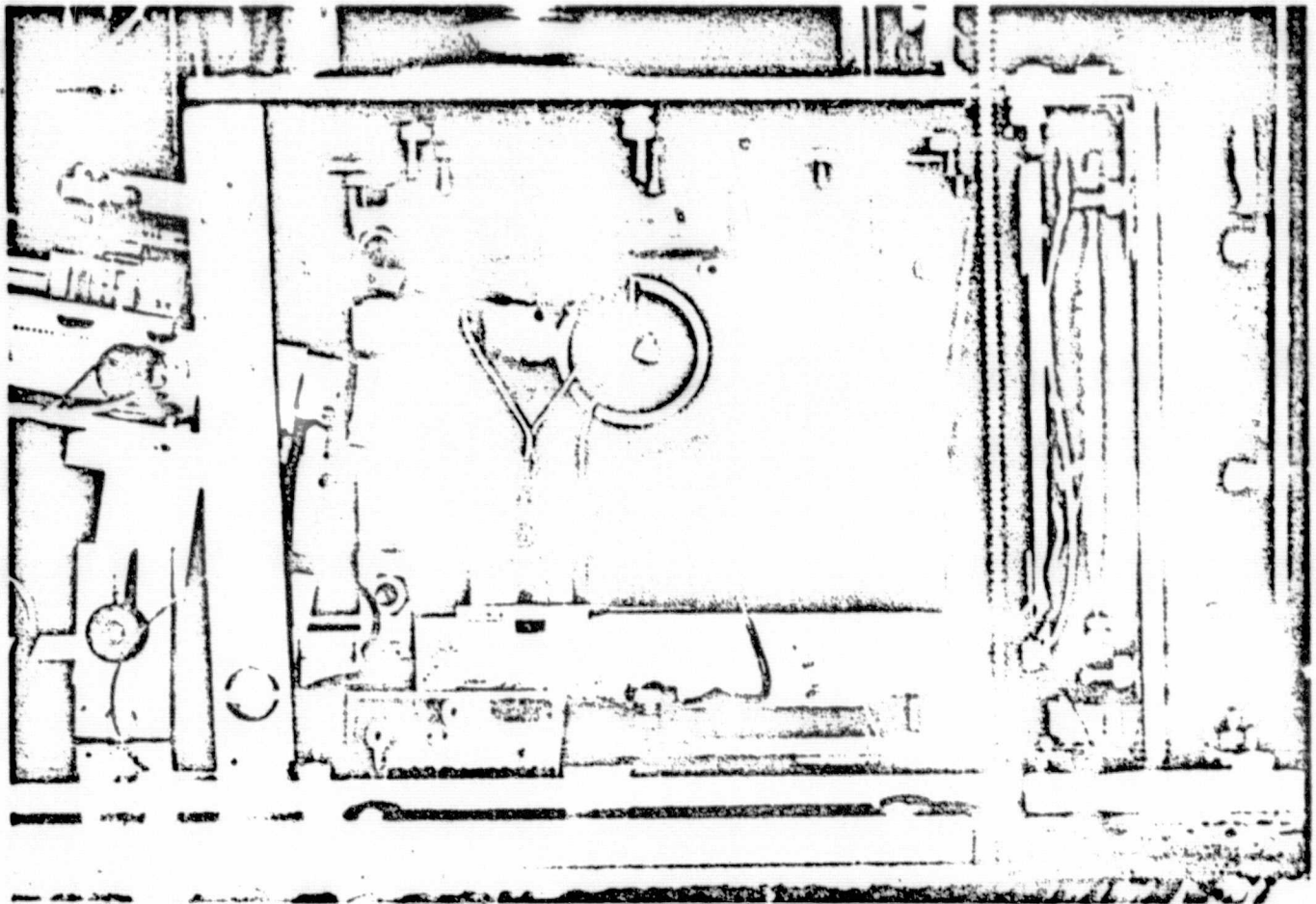


Fig. 5



VIEW INSIDE COMBUSTION CHAMBER LOOKING TOWARD
CAMERA SHOWING DROPLET MOUNTED ON QUARTZ FIBER
PRIOR TO TEST. DROPLET MOUNTING MECHANISM
AND IGNITION ELECTRODES ARE ALSO SHOWN.

Fig. 6

Fig. 7

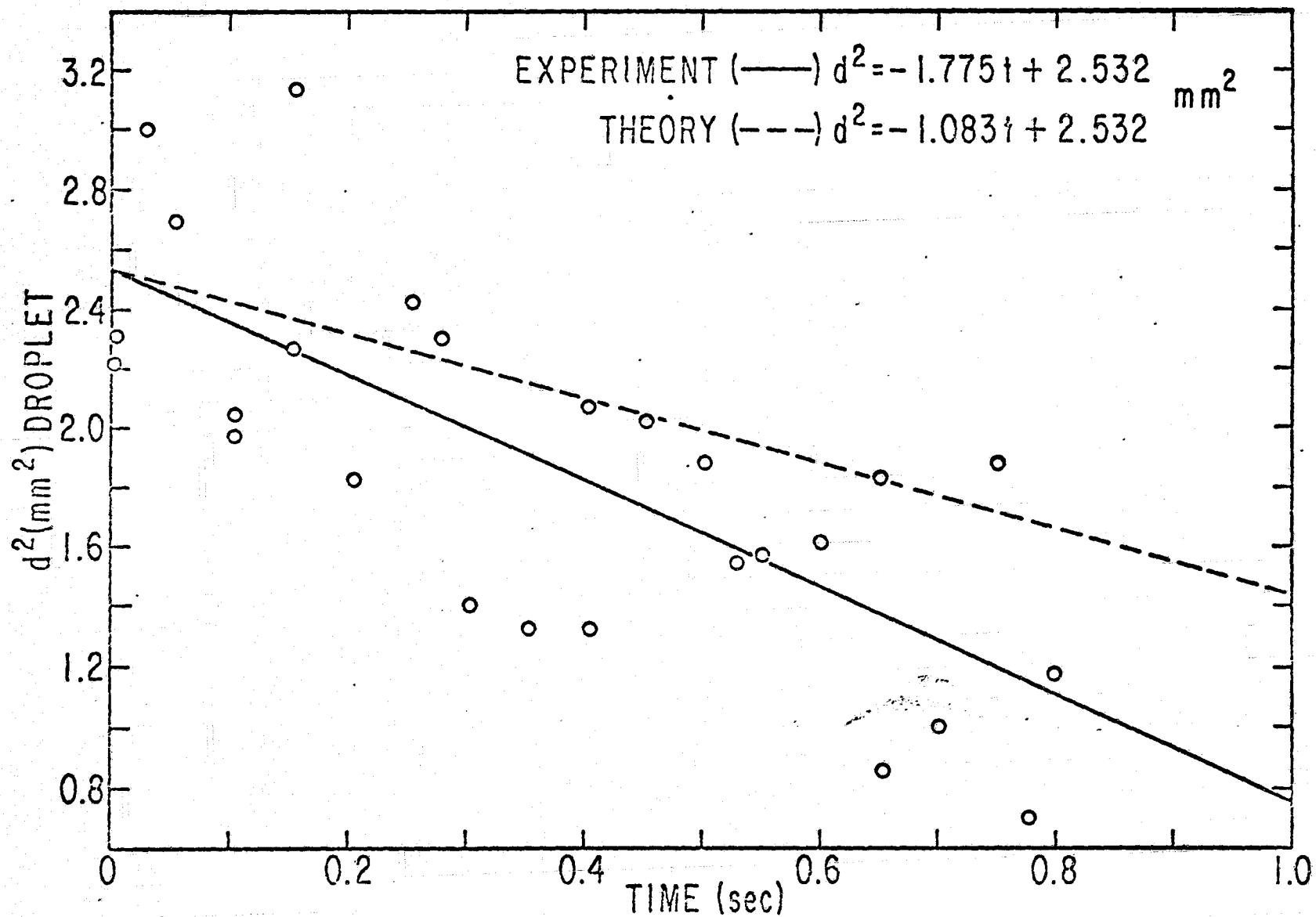


Fig. 8

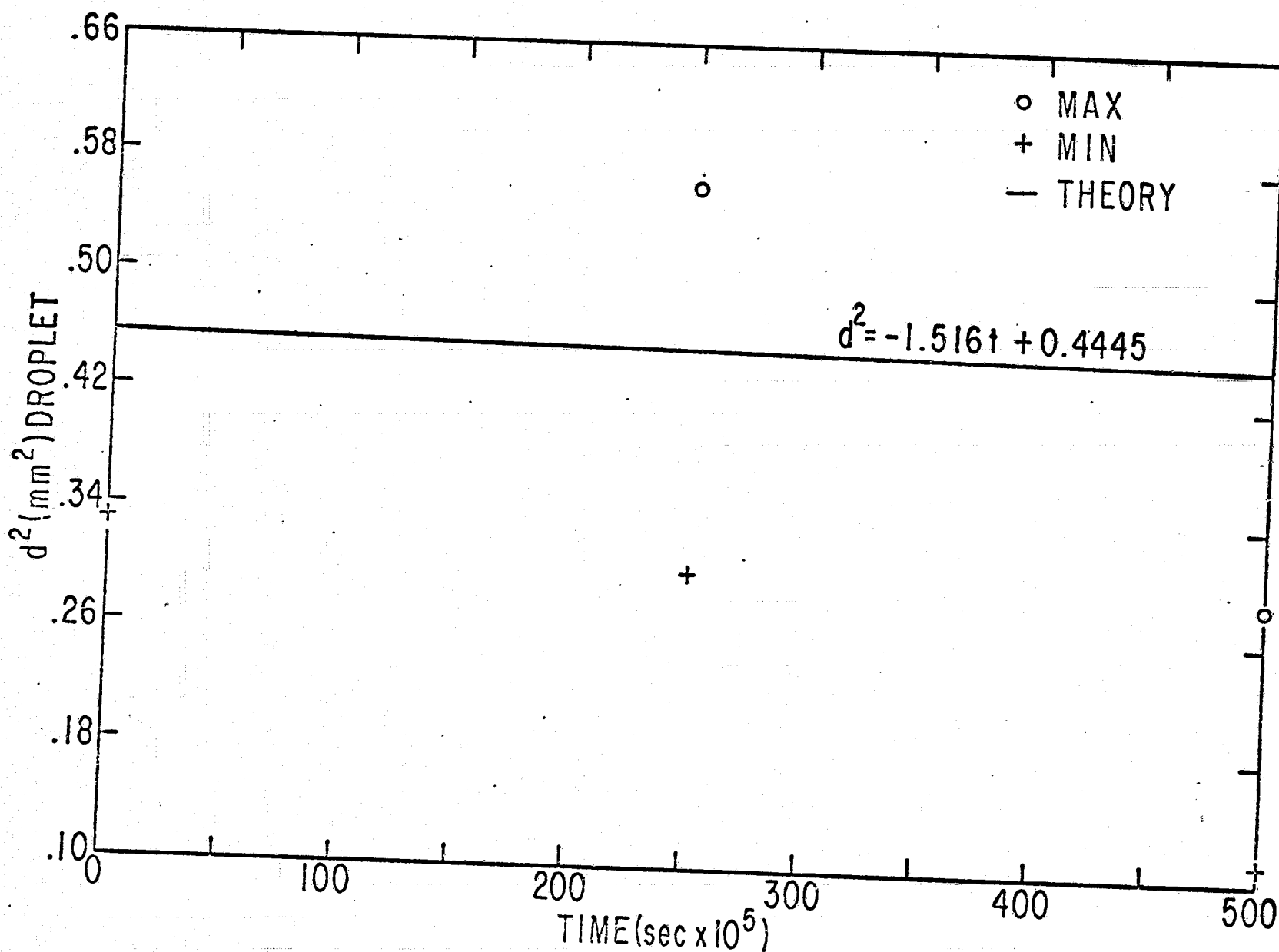


Fig. 9

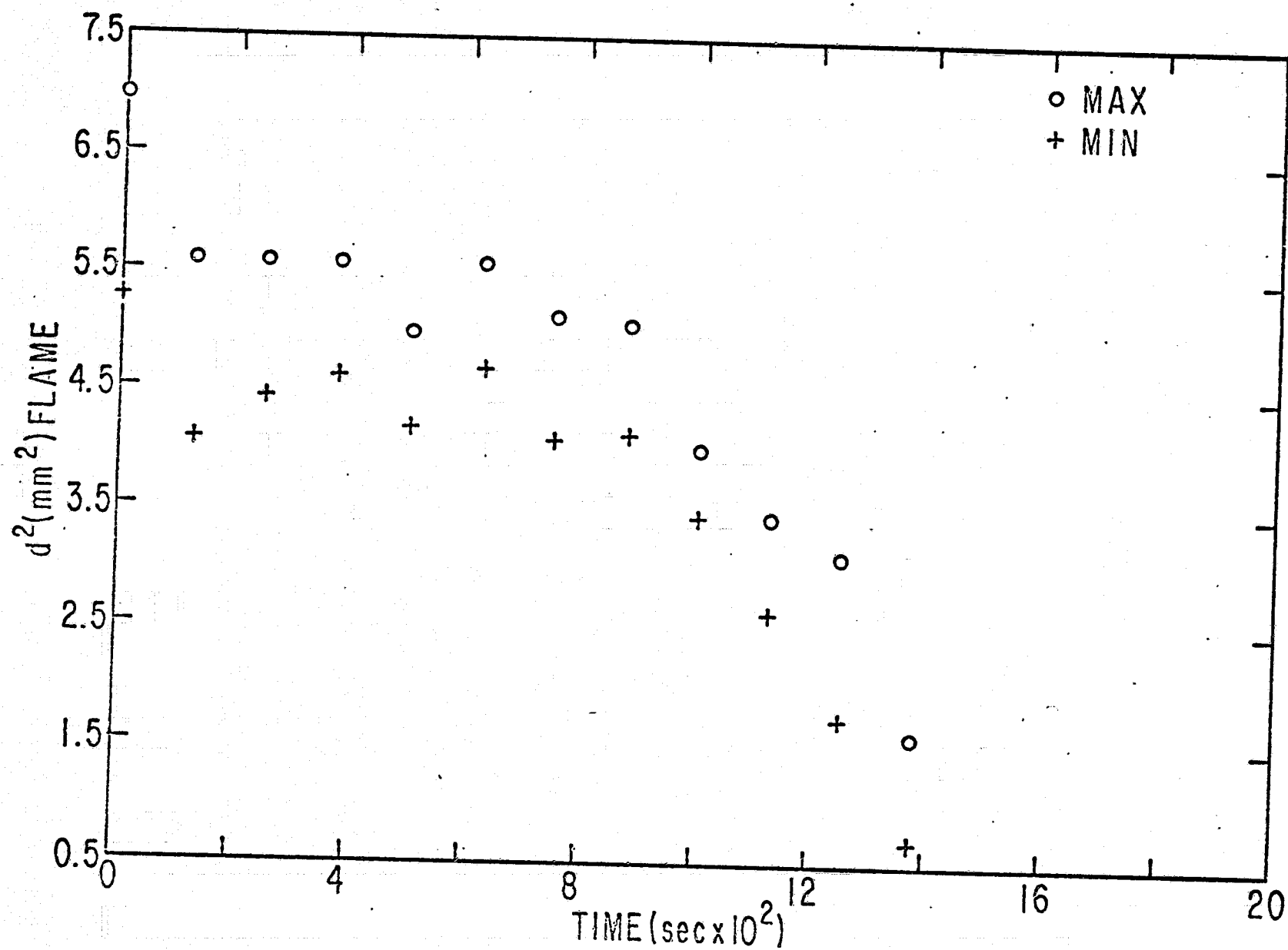
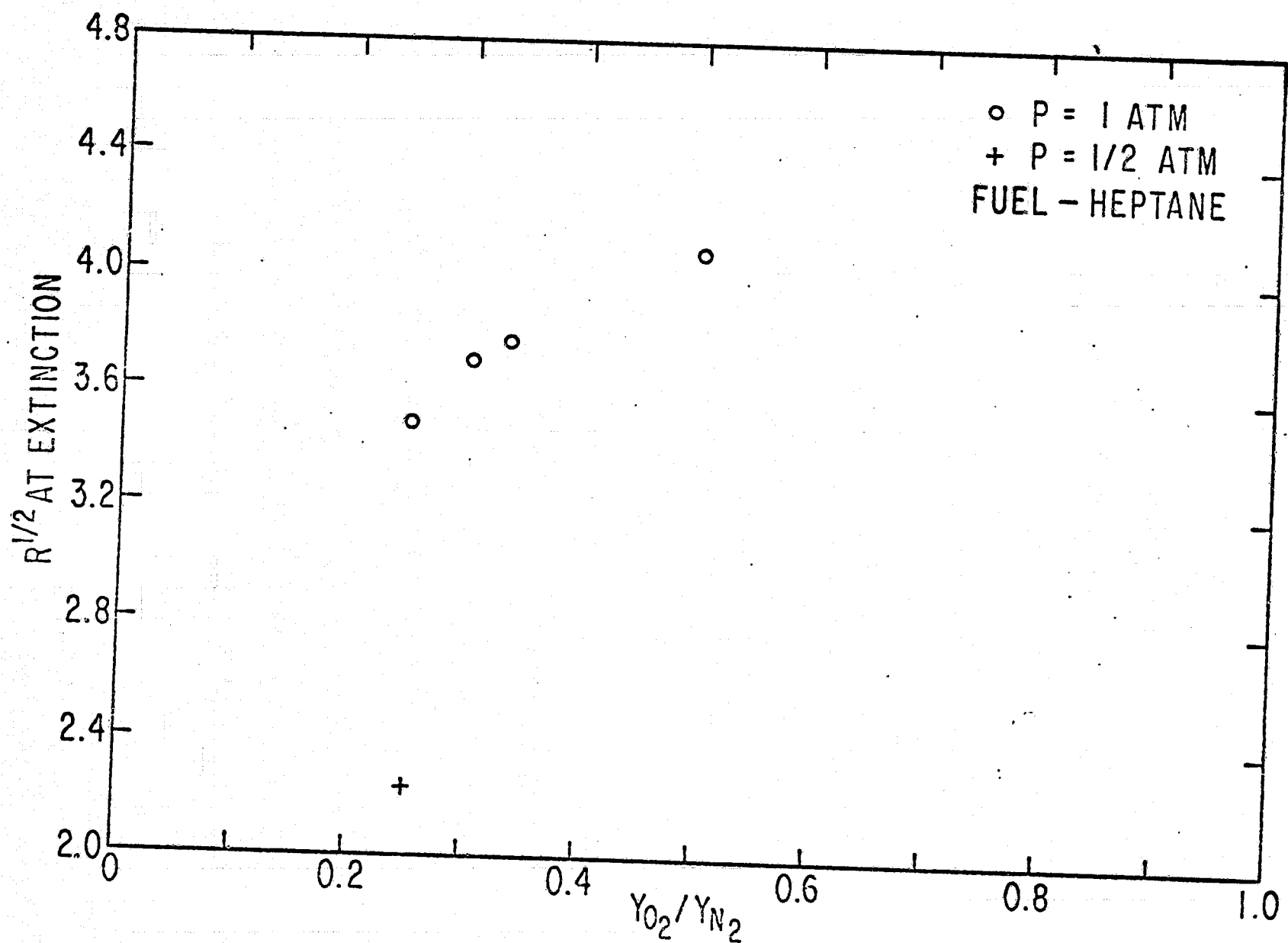
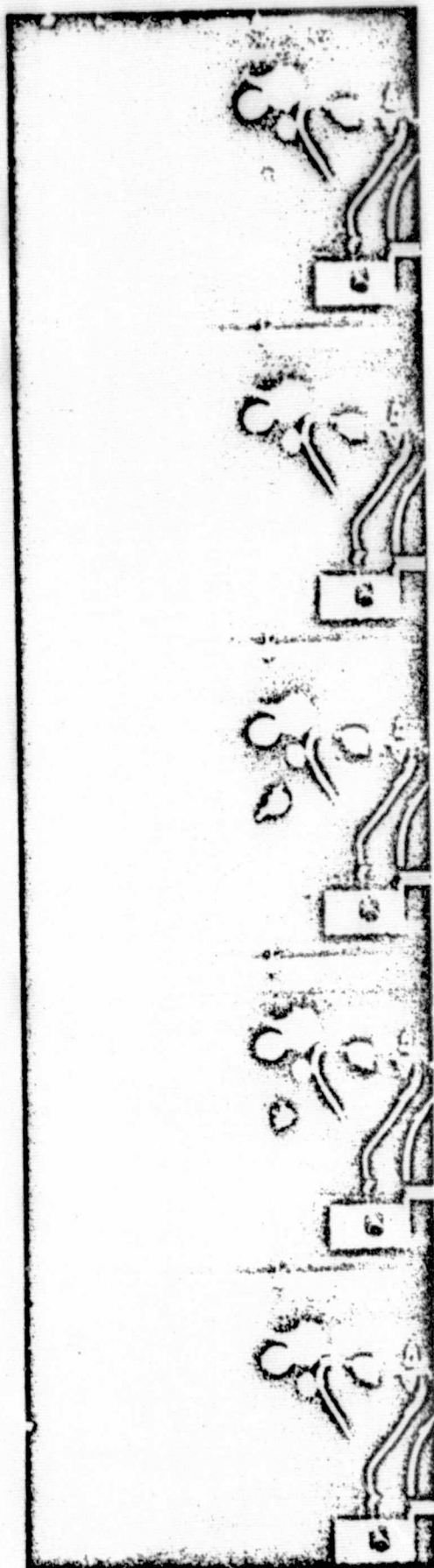


Fig. 10





DECANE SATELLITE
EXPERIENCING
"FLASH EXTINCTION"

$$Y_{O_{2\infty}} = 0.5$$

$$Y_{N_{2\infty}} = 0.5$$

$$T_{\infty} = 23.5^{\circ}\text{C}$$

$$P = 1 \text{ Atm.}$$

$$d_f = 1.5 \text{ mm}$$

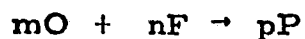
400 FRAMES/SEC.

Fig. 11

Appendix C1

Derivation of the Formula for the Damköhler Number

We assume a one-step reaction



whose specific reaction rate is given by $k = Be^{-E/R_o T}$. Using the definitions provided by Williams [2], we proceed as follows:

$$\sum_{i=1}^N \nu'_i m_i \rightarrow \sum_{i=1}^N \nu''_i m_i$$

$$dC_i/dt = [\nu''_i - \nu'_i] k \prod_{j=1}^N C_j^{\nu'_j}$$

$$X_i = (Y_i/W_i) / \sum_{j=1}^N Y_j/W_j$$

$$P/R_o T = p \sum_{i=1}^N Y_i/W_i \quad \text{for an ideal gas}$$

$$(P/R_o)(X_i) = p(Y_i/W_i)$$

$$\rho = \Sigma M/V$$

$$(P/R_o T)(X_i) = [\Sigma M/V][M_i/\Sigma M]/W_i$$

$$= M_i/W_i V = \text{moles per volume} \equiv C_i$$

$$C_i = (P/R_o T)(X_i)$$

In terms of oxygen in a diffusion flame, we then have

$$dC_O/dt = -mk C_O^m C_F^n = -mk \left[\frac{PX_O}{R_o T_f} \right]^m \left[\frac{PX_F}{R_o T_f} \right]^n$$

$$\text{let} \quad \zeta = \left[\frac{PX_O}{R_o T_f} \right] \quad \beta = \left[\frac{PX_F}{R_o T_f} \right]$$

$$d\zeta/dt = -mk \zeta^m \beta^n$$

Solving the equation and letting τ = chemical reaction time

$$\int \zeta^{-m} d\zeta = -mk\beta^n \int dt \quad m \neq 1$$

$$\frac{\zeta^{-m+1}}{1-m} = -mk\beta^n \tau$$

$$\tau = \zeta^{-m+1} / [1-m] [-mk\beta^n]$$

In terms of original variables

$$1/\tau = -m(1-m) k \left[\frac{P}{R_o T_f} \right]^{m+n-1} [X_O]^{m-1} [X_F]^n \quad m > 1, 1/\tau > 0$$

The Damköhler number D_f is defined as flow time/chemical time

Flow time = δ^2/D_{Of} where δ = flame thickness

$$D_F = \frac{\delta^2}{D_{Of}} k \left[\frac{P}{R_o T_F} \right]^{m+n-1} \left[\frac{Y_O}{W_O} \right]^{m-1} \left[\frac{Y_F}{W_F} \right]^n \left[\frac{1}{\sum_{j=1}^N (Y_j/W_j)} \right]^{m+n-1}$$

$$D_f = \frac{\delta^2}{D_{Of}} f \frac{[\rho_f]^{m+n-1} [Y_O]^{m-1} [Y_F]^n}{[W_O]^{m-1} [W_F]^n}$$

For an order of magnitude calculation, we can assume that

$Y_O = Y_F = 1$ so that we obtain

$$D_f = \frac{\delta^2 \rho_f^{m+n-1} B}{D_{Of} W_O^{m-1} W_F^n} \exp \left(\frac{-E}{R_o T_f} \right)$$

Appendix C2**Evaluation of Gas Properties**

In order to determine the theoretical burning rate, the factors appearing in Eqs. (13), (15) and (19) had to be evaluated. Since the gas properties were to be evaluated for (13) at $T = (T_b + T_f)/2$, the adiabatic flame temperature needed to be calculated. This was accomplished using the NASA Adiabatic Flame Temperature program, which allowed for dissociation and determined the equilibrium composition. The output of this program for all the test cases is shown in Appendix C.

The properties of each species were then evaluated using the following formulae from [32]. Temperatures are in $^{\circ}\text{K}$.

$$10^5 \lambda (\text{DECANE})(\text{cal/sec cm } ^{\circ}\text{C}) = -1.43507 + 9.06309 \times 10^{-3} T \\ + 1.78134 \times 10^{-5} T^2$$

$$C_p (\text{DECANE})(\text{cal/g } ^{\circ}\text{K}) = 0.0560393 + 1.41072 \times 10^{-3} T \\ - 6.96289 \times 10^{-7} T^2 + 1.31224 \times 10^{-10} T^3$$

$$C_p (\text{HEPTANE})(\text{cal/g } ^{\circ}\text{K}) = 0.982018 + 1.30398 \times 10^{-3} T \\ - 5.92804 \times 10^{-7} T^2 + 1.01054 \times 10^{-10} T^3$$

$$C_p (\text{O}_2)(\text{cal/g } ^{\circ}\text{K}) = 0.177100 + 1.49509 \times 10^{-4} T \\ - 8.44940 \times 10^{-8} T^2 + 1.83236 \times 10^{-11} T^3$$

$$C_p (\text{N}_2)(\text{cal/g } ^{\circ}\text{K}) = 0.201678 + 1.08013 \times 10^{-4} T \\ - 3.32212 \times 10^{-8} T^2 + 2.45228 \times 10^{-12} T^3$$

$$C_p(\text{AIR})(\text{cal/g } ^\circ\text{K}) = 0.208831 + 7.71027 \times 10^{-5} T \\ - 8.56726 \times 10^{-9} T^2 - 4.75772 \times 10^{-12} T^3$$

$\lambda(\text{O}_2, \text{N}_2, \text{AIR}, \text{HEPTANE})$ were obtained from tables in [32].

From [33], the properties of the fuels, ρ_L and ΔH_F° , were obtained which were also used in the NASA program to compute the flame temperature. From [34], the heat of vaporization and boiling temperatures of the fuels were also obtained. The conversion factors used were: $1 \text{ J/cm } ^\circ\text{K} = 4.1868 \text{ cal/cm-sec-} ^\circ\text{C}$, $1 \text{ BTU/lbm } ^\circ\text{F} = 4.184 \text{ J/g } ^\circ\text{K}$, $1 \text{ cal} = 4.186 \text{ J}$, and $1 \text{ lb/sec ft.} = 0.0671969 \text{ gm/cm-sec}$. The heats of combustion of the fuels were obtained from [35].

The computation of λ_{mix} , the thermal conductivity of the $\text{O}_2 - \text{N}_2$ gas mixture, was accomplished using the formula from [36] p. 258

$$\lambda_{\text{mix}} = \sum_{i=1}^N \frac{X_i \lambda_i}{\sum_{j=1}^N X_j \Phi_{ij}}$$

where

$$\Phi_{ij} = \frac{1}{\sqrt{8}} \left(1 + \frac{W_i}{W_j} \right)^{-\frac{1}{2}} \left[1 + \left(\frac{\mu_i}{\mu_j} \right)^{\frac{1}{2}} \left(\frac{W_j}{W_i} \right)^{\frac{1}{4}} \right]^2$$

λ_i and μ_i were evaluated at $T = (T_b + T_f)/2$, and the values

$W_{O_2} = 31.998$ gm/mole, $W_{N_2} = 28.013$ gm/mole were used;
 μ_i derived from expression $\mu_i = \lambda_i / \left(\frac{15}{4} \frac{R_o}{M_i} \right)$ as
 described in [36].

The effective thermal conductivity was then defined as

$$\lambda = 0.4 \lambda_F + 0.6 \lambda_{mix}$$

There were ten cases which needed to be evaluated since for each gas mixture and fuel, the flame temperature was different. These results are shown in Table 1, which also includes the density of the gas mixture, defined as

$$\rho_{mix} = X_{O_2} \rho_{O_2} + X_{N_2} \rho_{N_2}$$

The densities were evaluated at $23.5^\circ C$, an average ambient temperature, from the ideal gas law. The viscosities of the mixtures were obtained from [36], p. 24

$$\mu_{mix} = \sum_{i=1}^N \frac{X_i \mu_i}{\sum_{j=1}^N X_j \Phi_{ij}}$$

where Φ_{ij} is defined as above. These viscosities were evaluated

at 23.5 °C also, for use in the correction for forced convection (15).

In the evaluation of (19), several methods were used in defining the quantities therein. The gas properties were evaluated using the formulae above and the binary diffusion coefficient was evaluated from [36] p. 22 using

$$D_{ij} = \frac{0.0018583 \sqrt{T^3 \left(\frac{1}{W_i} + \frac{1}{W_j} \right)}}{P \sigma_{AB}^2 \Omega_{AB}}$$

As stated in 6-B above, the quantities in Eq. (19) were defined in several different manners and at several different temperatures. Each quantity was evaluated using the definitions below Eq. (20), and the value of K_o evaluated. The heat of combustion used was the lower heating value corresponding to gaseous products of combustion. The results were summarized in 6-B.

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FUEL	y_{O_2} %	y_{N_2} %	P " Hg	f F/O	T _{AD, Flame} °K	T °K	μ_{mix} g/cm-sec	C_p^{fuel} J/g °K	λ^{fuel} J/cm-s-°K	λ_{O_2} J/cm-s-°K	λ_{N_2} J/cm-s-°K	λ_{mix} J/cm-s-°K	λ J/cm-s-°K	ρ_c g/cm ³	ρ_{min} g/cm ³
HEPTANE	20	80	29.4	.2847	2160	1265.7	1.812 $\times 10^{-4}$	4.201	1.40 $\times 10^{-3}$	0.854 $\times 10^{-3}$	0.740 $\times 10^{-3}$	0.7590 $\times 10^{-3}$	1.0134 $\times 10^{-3}$	0.6120	1.1e02 $\times 10^{-3}$
HEPTANE	20	80	14.7	.2847	2147	1249.0	1.812 $\times 10^{-4}$	4.179	1.33 $\times 10^{-3}$	0.845 $\times 10^{-3}$	0.732 $\times 10^{-3}$	0.7508 $\times 10^{-3}$	1.0025 $\times 10^{-3}$	0.6144	5.3012 $\times 10^{-4}$
HEPTANE	25	75	29.4	.2847	2385	1378.2	1.825 $\times 10^{-4}$	4.326	1.60 $\times 10^{-3}$	0.910 $\times 10^{-3}$	0.787 $\times 10^{-3}$	0.8128 $\times 10^{-3}$	1.1277 $\times 10^{-3}$	0.6120	1.1e77 $\times 10^{-3}$
HEPTANE	30	70	29.4	.2847	2538	1454.7	1.837 $\times 10^{-4}$	4.402	1.75 $\times 10^{-3}$	0.949 $\times 10^{-3}$	0.821 $\times 10^{-3}$	0.8533 $\times 10^{-3}$	1.2120 $\times 10^{-3}$	0.6120	1.1752 $\times 10^{-3}$
HEPTANE	50	50	29.4	.2847	2856	1613.7	1.890 $\times 10^{-4}$	4.531	2.05 $\times 10^{-3}$	1.027 $\times 10^{-3}$	0.901 $\times 10^{-3}$	0.9571 $\times 10^{-3}$	1.3943 $\times 10^{-3}$	0.6120	1.20e5 $\times 10^{-3}$
DECANE	20	80	29.4	.2869	2159	1303.1	1.812 $\times 10^{-4}$	4.194	1.7008 $\times 10^{-3}$	0.873 $\times 10^{-3}$	0.755 $\times 10^{-3}$	0.7746 $\times 10^{-3}$	1.1451 $\times 10^{-3}$	0.6033	1.1e02 $\times 10^{-3}$
DECANE	25	75	29.4	.2869	2384	1415.6	1.825 $\times 10^{-4}$	4.310	1.9716 $\times 10^{-3}$	0.929 $\times 10^{-3}$	0.804 $\times 10^{-3}$	0.8302 $\times 10^{-3}$	1.28e8 $\times 10^{-3}$	0.6033	1.1e77 $\times 10^{-3}$
DECANE	30	70	29.4	.2869	2538	1492.6	1.837 $\times 10^{-4}$	4.380	2.1678 $\times 10^{-3}$	0.956 $\times 10^{-3}$	0.839 $\times 10^{-3}$	0.8713 $\times 10^{-3}$	1.3599 $\times 10^{-3}$	0.6033	1.1752 $\times 10^{-3}$
DECANE	50	50	29.4	.2869	2856	1651.6	1.890 $\times 10^{-4}$	4.510	2.6010 $\times 10^{-3}$	1.046 $\times 10^{-3}$	0.922 $\times 10^{-3}$	0.9775 $\times 10^{-3}$	1.6269 $\times 10^{-3}$	0.6033	1.20e5 $\times 10^{-3}$
DECANE	80	20	29.4	.2869	3056	1751.6	1.977 $\times 10^{-4}$	4.586	2.8928 $\times 10^{-3}$	1.096 $\times 10^{-3}$	0.982 $\times 10^{-3}$	1.069 $\times 10^{-3}$	1.7545 $\times 10^{-3}$	0.6033	1.25e5 $\times 10^{-3}$

TABLE 1

Gas properties for constant property theory

Appendix C3

The NASA Adiabatic Flame Temperature program was utilized to determine the flame temperature and equilibrium compositions for the ten test conditions. These correspond to the case numbers in the output and the following conditions:

CASE NO.	FUEL	ATMOSPHERE	PRESSURE (atm.)
1	heptane	20% O ₂ - 80% N ₂	1
2	heptane	20% O ₂ - 80% N ₂	1/2
3	heptane	25% O ₂ - 75% N ₂	1
4	heptane	30% O ₂ - 70% N ₂	1
5	heptane	50% O ₂ - 50% N ₂	1
6	decane	20% O ₂ - 80% N ₂	1
7	decane	25% O ₂ - 75% N ₂	1
8	decane	30% O ₂ - 70% N ₂	1
9	decane	50% O ₂ - 50% N ₂	1
10	decane	80% O ₂ - 20% N ₂	1

THEMODYNAMIC EQUILIBRIUM COMBUSTION PROPERTIES AT ASSIGNED

PRESSURE

7435770.

CHEMICAL FORMULA	MOLES	ENERGY CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC
FUEL: C ₂ H _{5.858888} H _{14.000000}	1.000000	-5329.000	1	300.00	-V
Oxidant: O ₂ 2.000000	11.000	0	G	300.00	-V
Oxidant: N ₂ 2.000000	50.263	0	G	300.00	-V

OX: 17.5000 PERCENT FUEL: 5.3829 EQUIVALENCE RATIO: 1.0000 PH: 1.0000 REACTANT DENSITY: 0

THEMODYNAMIC PROPERTIES

P, ATM	1.0000
T, DEG K	2110
H ₀ , BTU	1.5717E4
H, CAL/G	-2.9
S, CAL/(G)(K)	2.2063
P, MLC/RI	24.352
K ₂ , MLC/RI	-1.30173
K ₃ , MLC/RI	1.1542
C ₂ , CAL/(G)(K)	1.049
C ₃ , (S)	1.1002
S ₀ , BTU/(G)(K)	0.7110

MOLE FRACTIONS

CO	0.0760
CO ₂	0.0711
H	0.0019
H ₂	0.0010
H ₂ O	0.1524
N ₂	0.7159
N ₂	0.7004
O	0.0012
O ₂	0.1104
O ₂	0.0730

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-06 FOR ALL ASSIGNED CONDITIONS

C ₂ H ₄	C ₂ H ₆	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₄	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₂
C ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₂
C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂
H ₂ O(L)	-202	V	H ₂ O	LH	H ₂ O	LH	H ₂ O	LH	H ₂ O
N ₂	1206	1220	N ₂	O ₂	N ₂	O ₂	N ₂	O ₂	N ₂

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THERMODYNAMIC EQUILIBRIUM COMBUSTION PROPERTIES AT ASSIGNED

PRESSURES

CASE NO. 2

CHEMICAL FORMULA	MOLES	ENERGY CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC
FUEL: C ₂ H _{5.9250} H _{16.0000}	1.00000	-5319.000	1	300.00	-0
OXIDANT: O ₂	11.000	0	0	300.00	-0
OXIDANT: N ₂	50.240	0	0	300.00	-0

O/F = 17.5279 PERCENT FUEL = 5.5479 EQUIVALENCE RATIO = 1.0000 P/F = 1.0000 REACTANT DENSITY = 0

THERMODYNAMIC PROPERTIES

P, ATM	1.0000
T, DEG K	300.00
RHO, G/CC	1.85E-03
H, CAL/G	-8.5
S, CAL/(G*DEG K)	2.31E-01
W, MOLE WT	78.320
Q, W/WT	-1.0000
Q, CAL/WT	1.0000
Q, CAL/(G*DEG K)	1.0000
Q, CAL/G	1.0000
Q, CAL/(G*DEG K)	1.0000

MOLE FRACTIONS

O ₂	0.07602
CO ₂	0.07776
H ₂	0.07776
H ₂ O	0.07776
N ₂	0.07776
O	0.07776
CO	0.07776
CH ₄	0.07776
C ₂ H ₆	0.07776

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-06 FOR ALL ASSIGNED CONDITIONS

C(S)	C	CH	CH ₂	CH ₃	CH ₄	CH ₅	CH ₆	CH ₇	CH ₈
C ₂	C ₂ H	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₈	C ₂ H ₁₀	C ₂ H ₁₂	C ₂ H ₁₄	C ₂ H ₁₆
C ₃	C ₃ H	C ₃ H ₂	C ₃ H ₄	C ₃ H ₆	C ₃ H ₈	C ₃ H ₁₀	C ₃ H ₁₂	C ₃ H ₁₄	C ₃ H ₁₆
H ₂ O(L)	H ₂ O ₂	N	N ₂	N ₂ O	N ₂ O ₂	N ₂ O ₄	N ₂ O ₆	N ₂ O ₈	N ₂ O ₁₀
H ₂ O	H ₂ O ₂	N ₂ O	N ₂ O ₂	N ₂ O ₄	N ₂ O ₆	N ₂ O ₈	N ₂ O ₁₀	N ₂ O ₁₂	N ₂ O ₁₄

THEMODYNAMIC EQUILIBRIUM COMPUTATION PROPERTIES AT ASSIGNED

PRESSURES

CASE NO.

CHEMICAL FORMULA
FUEL: C₂H₄ 1.000000
OXIDANT: O₂ 2.000000
OXIDANT: N₂ 2.000000

MOLES ENERGY STATE TEMP DENSITY
CAL/MOL DEG K G/CC
1.000000 -2339.000 1 320.00 -0
11.000 0 0 300.00 -0
37.500 0 0 320.00 -0

C/P = 14.093 PERCENT FUEL = 6.6453 EQUIVALENCE RATIO = 1.0000 P/F = 1.0000 REACTANT DENSITY = 0

THEMODYNAMIC PROPERTIES

P, ATM .0920
T, DEG K 3200
RHO, G/CC 1.4140-4
H, CAL/G 13.6
S, CAL/(G)(K) 2.3063
H, KJ/MOL 25.172
H, KJ/MOL 1.55509
H, KJ/MOL 1.1423
C_p, CAL/(G)(K) 1.654
C_p, KJ/(G)(K) 1.1569
S₀, KJ/(G)(K) 216.9

MOLE FRACTIONS

C₂ .00150
C₂H₄ .15520
H₂ .00173
H₂O .17442
H₂O .00190
N₂ .77110
O₂ .00173
CO₂ .15520
CO₂ .00190

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-06 FOR ALL ASSIGNED CONDITIONS

C(S)	C	CH	CH ₂	CH ₃	CH ₄	CH ₅	CH ₆	CH ₇	CH ₈
C ₂	C ₂ H	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₈	C ₂ H ₁₀	C ₂ H ₁₂	C ₂ H ₁₄	C ₂ H ₁₆
C ₃	C ₃ H	C ₃ H ₂	C ₃ H ₄	C ₃ H ₆	C ₃ H ₈	C ₃ H ₁₀	C ₃ H ₁₂	C ₃ H ₁₄	C ₃ H ₁₆
H ₂ O(L)	H ₂ O	N	N ₂	N ₃	N ₄	N ₅	N ₆	N ₇	N ₈
N ₂ O	N ₂ O ₂	N ₂ O ₃	N ₂ O ₄	N ₂ O ₅	N ₂ O ₆	N ₂ O ₇	N ₂ O ₈	N ₂ O ₉	N ₂ O ₁₀

THEMODYNAMIC EQUILIBRIUM PROPERTIES AT ASSIGNED
PRESSURES

CASE NO.	OPTICAL FORMULA	MOLES	ENERGY CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC
FUEL C 7.15000	M 14.00000	1.000000	-9339.000	1	300.00	-0
Oxidant O 2.10000		11.000	0	G	300.00	-0
Oxidant N 2.10000		27.304	0	G	300.00	-0
O/F 11.7000	PERCENT FUEL 7.4710	EQUIVALENCE RATIO 1.0000	PWIF 1.0000	REACTANT DENSITY	0	

THEMODYNAMIC PROPERTIES

P, ATM	9.020
T, DEG K	2930
RHO, G/CC	1.31000
H, CAL/G	-14.2
S, CAL/(G)(K)	2.4000
M, MOL WT	27.379
U, KJ/KG	-11.2563
U, KJ/KG	1.2497
CP, CAL/(G)(K)	1.249
CP, KJ/(G)(K)	1.1412
SON, VEL, M/SEC	929.4

MOLE FRACTIONS

CO	.03091
CO2	.11436
H	.04279
H2	.00070
H2O	.15965
NO	.00065
N2	.00079
O	.00000
O2	.00000
O2	.00000

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-06 FOR ALL ASSIGNED CONDITIONS

C(S)	C	CH	CH2	CH2O	CH3	CH4	CH	C2H	C2H2
C2	C2H	C2H2	C2H4	C2H6	C2H	C2H2	C2H	C2H	C2H2
C4	C4	C4H	C4H2	C4H4	C4H6	C4H	C4H2	C4H	C4H2
H2O(L)	H2O2	N	N2O	NH	NH2	NH3	N2O	N2O	N2H4
H2O	N2O4	N2O2	N3	O3					

THERMODYNAMIC EQUILIBRIUM PROPERTIES AT ASSIGNED

PRESSURES

CASE NO. 5

CHEMICAL FORMULA	MOLES	ENERGY - CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC
FUEL: C ₂ H ₂ 7.22500 H ₂ 14.00000	1.000000	-5339.000	1	300.00	-0
Oxidant: O ₂ 2.00000	11.000		0 G	300.00	-0
Oxidant: N ₂ 2.00000	12.562		0 G	300.00	-0
C/F = 7.0445 PERCENT FUEL = 12.4619 EQUIVALENCE RATIO = 1.0000 P/F = 1.0000 REACTANT DENSITY = 0.					

THERMODYNAMIC PROPERTIES

P, ATM	1.9826
T, DEG K	2050
RHO, G/CC	1.1041-4
H, CAL/G	-10.7
S, CAL/(G)(K)	2.6266
M, MOL WT	26.086
(DLV/DLT) _T	-1.03099
(DLV/DLT) _P	1.6422
C _P , CAL/(G)(K)	1.5120
GAMMA (S)	1.1178
SP. HEAT, BTU/SEC	100.0

MOLE FRACTIONS

CO	.1726
CO2	.1723
H	.0162
H2	.0001
H2O	.0264
N	.2879
N2	.7165
O	.4009
O2	.0113
O3	.0077
O4	.0472

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.0000E-04 FOR ALL ASSIGNED CONDITIONS

C(S)	C	CH	CH2	CH2O	CH3	CH4	CH	C2N	CN2
C2	C2H	C2H2	C2H4	C2H6	C2H	C2H2	C2	C3	C3H2
C4	C5	HCO	HCO	HCO	HNO	HNO2	HN13	H2O(S)	H2O(L)
H2O2	N	N2	NH	NH2	NH3	NO2	NO3	N2H4	N2O
N2O4	N2O5	N3	O3						

THERMODYNAMIC EQUILIBRIUM COMBUSTION PROPERTIES AT ASSIGNED

PRESSURES

CASE NO.	CHEMICAL FORMULA	MOLES	ENERGY CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC
FUEL	C ₁₂ H ₂₂ O ₁₁ H ₂ O ₂	1.000000	-7172.000		300.00	-0
OXIDANT	O ₂	15.500		0 G	300.00	-0
OXIDANT	N ₂	70.000		0 G	300.00	-0
O/F = 17.457 PERCENT FUEL = 5.4272 EQUIVALENCE RATIO = 1.0000 P/F = 1.0000 REACTANT DENSITY = 0						

THERMODYNAMIC PROPERTIES

P, ATM	.9026
T, DEG K	2159
R, G/CC	1.5791E-4
H, CAL/G	71.7
S, CAL/(G DEG K)	2.7022
W, M/SEC	287405
U, VOL/SEC	1.1001E-3
V, VOL/SEC	1.1001E-3
C _p , CAL/(G DEG K)	1.5032
G, G/SEC	1.1992
S, VEL, M/SEC	270.6

MOLE FRACTIONS

CO	.00767
CO ₂	.10068
H	.00019
H ₂	.00179
H ₂ O	.11994
N ₂	.00159
N ₂	.78549
O	.00012
O ₂	.00161
O ₂	.00092

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-03 FOR ALL ASSIGNED CONDITIONS

CH ₄	C ₂ H ₆	C ₂ H ₄	C ₂ H ₂	CH ₃	CH ₃	CH	CN	CN ₂
C ₂	C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂
C ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂
C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂
C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂	C ₂ H ₂

THERMODYNAMIC EQUILIBRIUM : SECTION PROPERTIES AT ASSIGNED

CASE NO. 7

PRESSURES

CHEMICAL FORMULA		MOLES	ENERGY CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC
FUEL: C ₂ H ₂ 2.00000	H ₂ 2.00000					
OXIDANT: O ₂ 2.00000		1.00000	-71.12,000		300.00	-0
OXIDANT: N ₂ 2.00000		19.900		0 6	300.00	-0
		51.103		0 6	300.00	-0
O/F 13.9007 PERCENT FUEL 6.6931		EQUIVALENCE RATIO 1.0000		P/R 1.0000		REACTANT DENSITY 0

THERMODYNAMIC PROPERTIES

P, ATM	1.0000
T, DEG K	300.00
R, G/G	1.4101-4
H, CAL/G	-8.6
S, CAL/(G)(K)	2.0010
V, M/G	25.037
LOG VAPOR PRESS	-1.0000
LOG VOLTAGE	1.1420
CP, CAL/(G)(K)	1.0000
SMVA (S)	1.1000
SMV, VEL, M/SEC	200.0

MOLE FRACTIONS

CO	.07174
CO ₂	.11169
H ₂	.00102
H ₂ O	.00477
H ₂ O	.13024
NO	.00097
N ₂	.77637
O	.00079
O ₂	.10014
O ₂	.00070

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-06 FOR ALL ASSIGNED CONDITIONS

C ₂ H ₂	C ₂ H ₄	C ₂ H ₆	C ₂ H ₈	C ₂ H ₁₀	C ₂ H ₁₂	C ₂ H ₁₄	C ₂ H ₁₆	C ₂ H ₁₈	C ₂ H ₂₀
C ₂	C ₂ H	C ₂ H ₂	C ₂ H ₃	C ₂ H ₄	C ₂ H ₅	C ₂ H ₆	C ₂ H ₇	C ₂ H ₈	C ₂ H ₉
C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂	C ₂
H ₂ O (L)	H ₂ O	N	N ₂	N ₂	N ₂	N ₂	N ₂	N ₂	N ₂
N ₂	N ₂	N ₂	N ₂	N ₂	N ₂	N ₂	N ₂	N ₂	N ₂

PRESSURES

ॐ नमो भगवते वासुदेवाय

CRITICAL FORMULA			
FUEL	0.00000	W	0.00000
OXIDANT	0.00000		
OXIDANT	0.00000		

MOLES	ENERGY	STATE	TEMP	DENSITY
	CAL/MOL		DEG K	G/CC
1.000000	-2132.200	L	300.00	-0
10.0000	0	G	300.00	-0
41.292	0	G	300.00	-0

DATA: 1.1253 PERCENT FUEL 7.229 EQUIVALENCE RATIO 1.0000 PHIP 1.0000 REACTANT DENSITY 0

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P, 1TH	79325
T, 2ND	79338
W, 3RD	1,831,144
M, 4TH	74,0
S, 5TH	2,3947
T, 6TH	27,455
U, 7TH	1,87463
V, 8TH	1,75066
W, 9TH	1,8314
X, 10TH	1,1411
Y, 11TH	67,8

MOLE FRACTION IS

C2		.07939
CW2		.11730
H		.08775
M2		.07655
M2D		.12636
N3		.07605
P2		.03974
D		.07230
L4		.02276
OZ		.01715

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-06 FOR ALL ASSIGNED CONDITIONS

C(S)	C	C4	C42	C420	C43	C44	C4	C4N	C42
C2	C27	C242	C214	C246	C2N	C2H2	C27	C3	C302
C4	C5	C54	C50	C504	C40	C402	C423	C42	C40151
H20(L)	H242	H	H00	H4	H42	H43	H02	N03	H249
H24	H246	H232	H3	C3					

THERMODYNAMIC EQUILIBRIUM COMBUSTION PROPERTIES AT ASSIGNED

PRESSURES

CASE NO.	CHEMICAL FORMULA	MOLES	ENERGY CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC
FUEL	$\text{C}_2\text{H}_5\text{OH}$	1.000000	-7122.000	1	300.00	-V
Oxidant	O_2	15.900	0	g	300.00	-V
Oxidant	N_2	12.701	0	g	310.00	-V

CASE 6.2/03 PERCENT FUEL 12.9459 EQUIVALENCE RATIO 1.0000 PH1 1.0000 REACTANT DENSITY 0

THERMODYNAMIC PROPERTIES

P, ATM	1.0000
T, DEG K	2000
RHO, G/CC	1.1100E-4
M, CAL/G	-6.5
S, CAL/(G*DEG K)	2.0000
W, MOLAL	2.0000
W, MOLAL	2.0000
W, MOLAL	2.0000
W, MOLAL	2.0000
W, MOLAL	2.0000
W, MOLAL	2.0000
W, MOLAL	2.0000
W, MOLAL	2.0000

MOLE FRACTIONS

CO	1.7691
CO2	1.7465
H	0.0001
H2	0.0001
H2O	0.0001
NO	0.0001
N2	0.0001
O	0.0001
O2	0.0001
O3	0.0001

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-03 FOR ALL ASSIGNED CONDITIONS

C(S)	C	CO	CH2	CH2O	CH3	CH4	CH	CNN	CN2
C2	C2H	C2H2	C2H4	C2H6	C2N	C2H2	C2H	C3	C3H2
C4	C5	H2	H2O	H2O2	H2O	H2O2	H2O	H2O2	H2O2
H2O2	N	N2	NH	NH2	NH3	NH2	NH3	N2H4	N2O
N2O4	N2O5	N3	O3						

THEMODYNAMIC EQUILIBRIUM COMBUSTION PROPERTIES AT ASSIGNED

PRESSURES

CASE NO.	10							
	CHEMICAL FORMULA	MOLES	ENERGY CAL/MOL	STATE	TEMP DEG K	DENSITY G/CC		
FUEL	C ₁₂ H ₂₂ O ₁₁ H ₂ O ₂	1.000000	-7122.000	1	300.00	-V		
OXIDANT O	O ₂	15.000	0	G	300.00	-V		
OXIDANT N	N ₂	4.525250	0	G	300.00	-V		
O/E= 4.3270 PERCENT FUEL= 18.6571 EQUIVALENCE RATIO= 1.0000 P=1.0300 REACTANT DENSITY= 0								

THEMODYNAMIC PROPERTIES

P, ATM	1.0300
T, DEG K	300.00
H ₂ O, G/CC	4.525250
H, CAL/G	-7122.000
S, CAL/(G*DEG K)	2.8540
M, MOL WT	24.394
ICLV/DEG K	-1.00108
ICLV/DEG K	2.7617
CP, CAL/(G*DEG K)	2.7617
GAMMA (S)	1.1121
SON VEL, M/SEC	1376.3

MOLE FRACTIONS

CO	1.4715
CO2	1.4786
H	3.4167
H2O	0.0702
H2	0.4752
N2	0.4145
O	0.0001
N	0.01375
N2	1.3674
O	0.0001
N	0.01375
O2	0.0001

ADDITIONAL PRODUCTS WHICH WERE CONSIDERED BUT WHOSE MOLE FRACTIONS WERE LESS THAN 5.00000E-06 FOR ALL ASSIGNED CONDITIONS

DEG	C	H	CH2	CH2O	CH3	CH4	CH	CNN	CH2
C2	C2H	C2H2	C2H4	C2H6	C2N	C2N2	C2O	C3	C3O2
C4	C4H	C4H2	C4H4	C4H6	C4N	C4N2	C4O	C4O2	C4O4
H2O2	H2O	H2	H2O2	H2O	H2O	H2O	H2O	H2O	H2O
N2O	N2	N2	N2	N2	N2	N2	N2	N2	N2

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APPENDIX D

SUMMARY OF SCIENTIFIC RESULTS

Idealizations common to theories of droplet burning include the approximations of spherical symmetry, quasisteady flow and chemical equilibrium. Since elimination of any of these assumptions makes it difficult to calculate theoretical results accurately, accurate comparison between theory and experiment may be facilitated by performing experiments under conditions that conform to the stated approximations. The most obvious source of departure from the assumptions for experiments with suspended droplets is buoyancy that produces nonspherical convection. Kumagai [1-3] pioneered the elimination of this effect by studying droplet combustion in a freely falling chamber. Results of experiments similar to those of Kumagai have been obtained.

In Kumagai's tests zero gravity was achieved for less than one second, so that burning to completion in spherical symmetry never occurred. It was found that unsteady conditions prevailed in the gas during the entire observation time. To see if quasisteady flow could be established under conditions of spherical symmetry, the present experiments were performed in a facility affording more than two seconds of zero gravity. An experimental apparatus was designed, and a series of experiments upon n-heptane and n-decane droplets burning in atmospheres consisting of 20, 25, 30, 50 and 80 percent oxygen diluted with

nitrogen at pressures of 1/2 and 1 atmosphere were performed at the NASA-Lewis Research Center's Zero-Gravity Facility. Data from the experiments were obtained from a 16 mm high speed motion picture camera which recorded the events that occurred in each test.

Typical data obtained through analysis of the film on a Vanguard Motion Picture Analyzer are shown in Figure 1. The experimental burning rate constant for n-heptane burning in air was determined by a linear regression analysis of the data to be $K = 1.775 \times 10^{-2} \text{ cm}^2/\text{sec}$. The theoretical value corrected for forced convective effects was $K = 1.083 \times 10^{-2} \text{ cm}^2/\text{sec}$ based upon the classical constant property theory. This heptane droplet was observed for 2.16 seconds in the absence of buoyancy and appeared to achieve quasisteady conditions but in the presence of forced convection due to droplet motion. Even after application of a correction for convection, the burning rate exceeded that measured by Kumagai [2]. The extent to which measured burning rates exceeded those calculated theoretically seemed to increase with increasing oxygen content of the atmosphere and to be greater for decane than for heptane.

A phenomenon termed "flash extinction" was observed in tests with decane. Minute satellite droplets were observed to burn to very small diameters at which point they seemed to explode. These tests were performed in atmospheres consisting of 50% O_2 - 50% N_2 by mass at one atmosphere pressure.

The most probable explanation for the flash extinction and the discrepancy between the theoretical and experimental values for the burning rate is the accumulation of hot carbon at or near the surface of the droplet through pyrolysis processes. The carbon production would be most severe in the tests with decane where the flash extinction occurred and where experimental-theoretical differences are the greatest. Ignition of the hot carbon at the droplet surface could cause the flash, and the increased heat transfer from the carbon to the droplet could increase the burning rate. Carbon buildup would be less severe in the experiments of Kumagai because of the shorter test time.

Carbon production can be seen then as possibly being a crucial factor in this type of combustion. The burning time, fuel used, and atmospheric content all are factors which affect the production of carbon, whose effect upon the burning rate has not yet been fully demonstrated. It may be inferred that even if conditions of quasisteady flow and spherical symmetry are achieved, effects of nonequilibrium chemistry seem likely to influence droplet burning.

References

1. Kumagai, S., Jet Propulsion 26, 786 (1956).
2. Kumagai, S., Sakai, T. and Okajima, S., Thirteenth Symposium (International) on Combustion, The Combustion Institute, Pittsburgh (1971) pp. 779-785.
3. Okajima, S. and Kumagai, S., Fifteenth Symposium (International) on Combustion, The Combustion Institute, Pittsburgh (1975) pp. 401-407.

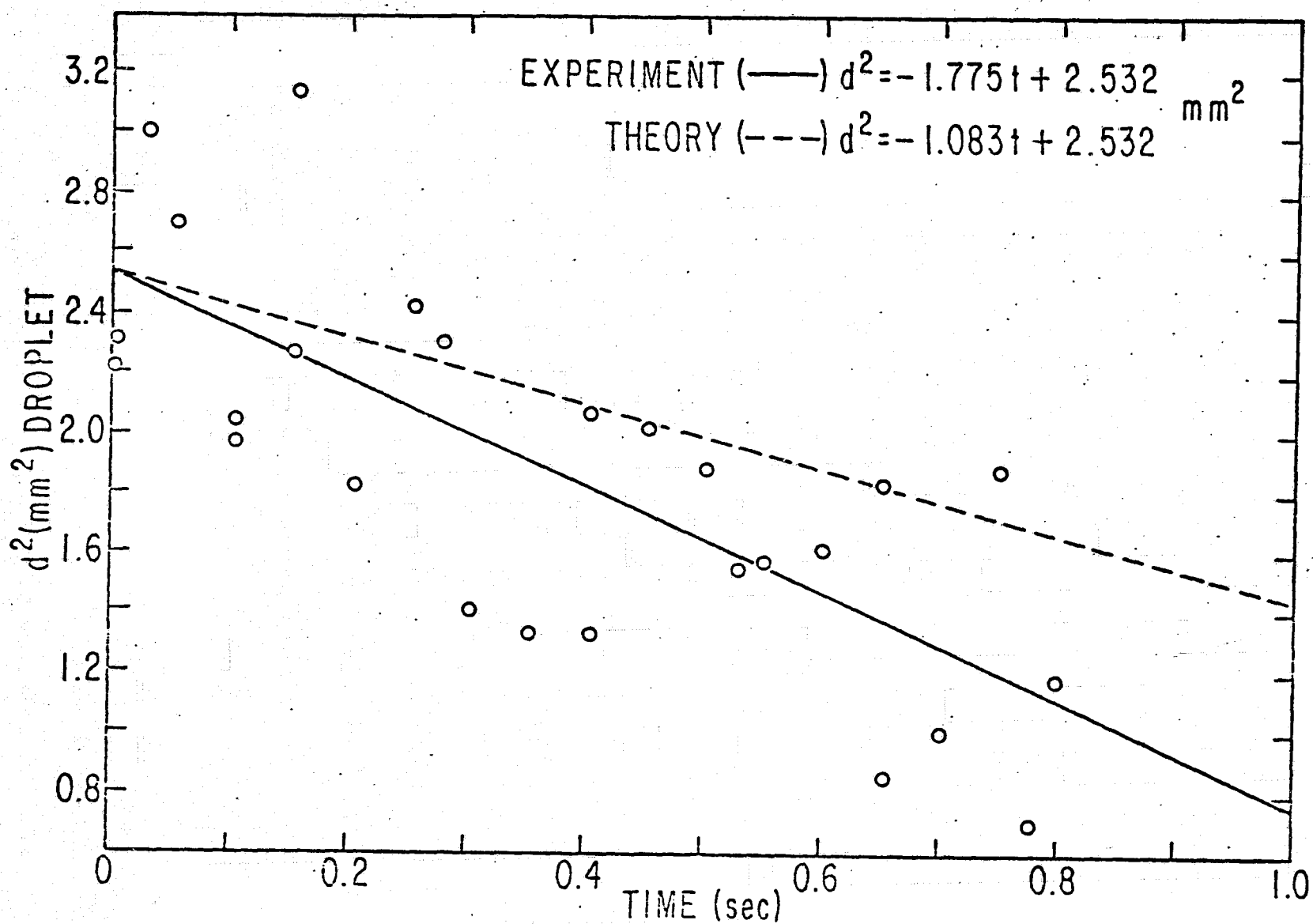


Figure 1. The square of the droplet diameter as a function of time for an n-heptane droplet burning in air at a pressure of one atmosphere and an ambient temperature of 20°C.

APPENDIX E

ELECTROSTATIC POSITIONING
OF DROPLETS1. Abstract

Electrostatic methods for positioning a droplet in a confined space were studied. The droplets were charged by induction, due to the presence of an electrostatic field between two parallel plates, one grounded and the other connected to a high positive voltage. A resulting attractive force between the charged droplet and the positive plate caused the droplet to move toward that plate. Experiments were performed in which the plates were made of aluminum and were 20 cm in diameter. The polar liquid, water, and the non-polar liquid, heptane, were investigated. The inductive charges attained by the droplets were measured to be on the order of 10^{-12} coulombs for millimeter-size droplets in electric fields on

the order of kilovolts per centimeter. The charges attained by water were approximately on order of magnitude greater than those attained by heptane. The results suggest that a six-electrode arrangement providing controllable electrostatic fields in three mutually perpendicular directions may position a droplet at a predetermined point in a confined space if influences of gravity are negligible.

2. Nomenclature

a	acceleration, Vq/md .
b	viscosity parameter, $6\pi\eta r/m$.
c	charge density.
d	separation between capacitor plates.
E	electrostatic field intensity, V/d .
g	acceleration of gravity.
j	current density.
k	droplet growth factor.
m	mass of droplet.
q	charge of droplet.
r	radius of droplet.
t	time.
V	voltage.

x	horizontal displacement of droplet.
y	vertical displacement of droplet.
ϵ	permittivity.
η	viscosity of the surrounding gas.
ρ	droplet density.
σ	electrical conductivity.
τ	time constant, ϵ/σ .

3. Introduction

The NASA Spacelab program affords the opportunity to perform experiments under conditions of negligible gravity, in various areas of basic research. One of those areas is combustion, and NASA has designed a combustion facility for pursuit of such experiments in Spacelab.¹ Buoyancy is a predominant force for removing hot combustion products from burning condensed materials, and the negligible acceleration of gravity in Spacelab effectively removes buoyancy, thereby potentially introducing significant modifications to burning mechanisms. A preliminary study has been performed by Knight² on the effect that elimination of gravitational forces exerts on the burning of liquid fuel droplets. As part of his study, experiments in a drop tower were performed, similar to those of Kumagai and Isoda.³

The results indicated that in earthbound experiments there are severe experimental difficulties in positioning a free, stationary droplet at a prescribed position and observing its entire burning history under conditions of negligible forced or free convection. Therefore droplet burning appears to be one promising area for study in Spacelab. Other droplet experiments, not involving combustion, also may be of interest. In connection with such experiments it is necessary to have a method for positioning a droplet, preferably remotely, at a selected location in a gas-filled chamber in the absence of gravity. The present paper addresses this problem of droplet positioning.

Three of the methods that may be considered for droplet positioning are mechanical,⁴ acoustical⁵ and electrical.^{6,7} Mechanical devices work best at reduced pressures but tend to introduce flow disturbances in atmospheres that may support combustion. Acoustical methods have been well developed recently,⁴ but they require placement of the droplet inside a relatively small chamber with a high degree of enclosure that may interfere somewhat with the observation of droplet combustion. Therefore electrical methods warrant study. Of the various conceivable electrical techniques, a straightforward use of electrostatics would appear to be simplest. Therefore an investigation was undertaken into electrostatic methods of droplet positioning.

The most direct concept for application of electrostatic methods begins with the assumption that the droplet is electrically neutral. Some

liquids have permanent dipole moments, and even those that do not are polarizable in an electric field. In either case, the net dipole moment $\vec{P}$ in the direction of an applied electric field $\vec{E}$ is $\vec{P} = \alpha \vec{E}$, where α is the polarizability, which is often expressed as being proportional to the permittivity through the electrical susceptibility.⁸ Since the force on a droplet in an electric field is $\vec{P} \cdot \nabla \vec{E} = \alpha \vec{E} \cdot \nabla \vec{E}$, a nonuniform field can serve to move an uncharged droplet. Although data on electrical susceptibilities are sparse, the permanent dipole moment of water, 6.1×10^{-30} coulomb-meters, can be used to estimate the fields needed for positioning a droplet in an apparatus having a size on the order of a few tens of centimeters, and it is found that fields on the order of a few hundred volts per centimeter may suffice. A suitable arrangement may be to place six spherical electrodes inside a grounded cage and to plan to attract the droplet toward any desired electrode by applying a voltage to that electrode.

When an effort was made experimentally to test this concept, it was found that net charges that existed within droplets overbalanced the effect of the induced dipole moment. Therefore an alternative approach was studied, in which the charge on the droplet was used to move the droplet in a nearly uniform electric field. The present paper details a study that has been made on the motion of liquid droplets in a nearly uniform electrostatic field. The principal results are curves defining the extent to which the droplet can be charged.

4. Theoretical Considerations

The presence of a positive charge on one plate of a capacitor near a grounded droplet causes a redistribution of electrons within the droplet, giving it a negative surface charge, the creation of which is called charging by induction. After the droplet is released from the grounding element, the resulting attractive force between the droplet and the positive plate causes the droplet to move toward that plate.⁷

Under the influence of gravity acting in the downward (y) direction the equations of motion of a droplet exposed to an electric field in the horizontal (x) direction then are

$$m \ddot{x} = (V/d) q - 6\pi\eta r \dot{x} \quad (1)$$

and

$$m \ddot{y} = mg - 6\pi\eta r \dot{y}, \quad (2)$$

in which the last term is the Stokes formula for the droplet drag force. The geometrical arrangement in which Eqs. (1) and (2) apply is that which has been set up in the laboratory to test the concept of electrostatic positioning.

It is possible for the net charge q on the droplet to vary with time. An estimate of whether such a variation occurs may be obtained from a combination of the law of conservation of charge, $\nabla \cdot \vec{j} + \dot{c} = 0$, Ohm's law, $\vec{j} = \sigma \vec{E}$ and Maxwell's first equation, $\nabla \cdot \vec{E} = c/\epsilon$.

Here $\vec{j}$ is the current density and c is the charge per unit volume. These equations yield $c/\tau + \dot{c} = 0$, where $\tau = \epsilon/\sigma$ is the characteristic time over which the charge may change. Use may be made of the permittivity of free space⁸ ($\epsilon_0 = 8.85 \times 10^{-14}$ farad/cm) and of an approximate value for the conductivity of air⁹ ($\sigma = 0.25 \times 10^{-15}$ ohm⁻¹ cm⁻¹) to obtain the estimate $\tau \approx 354$ sec. Although measured variations in σ exceed an order of magnitude,⁹ characteristic charging times under the experimental conditions always are greater than a few tens of seconds, large compared with the time that a droplet remains between the condenser plates in the laboratory measurements (tenths of seconds). Therefore q may be expected to remain approximately constant in these tests. It may be noted that air ionization, which occurs above a critical field intensity¹⁰ of 30 KV/cm at atmospheric conditions, did not occur in the present experiments since the intensities did not exceed 12 KV/cm. It may also be inferred that collection of charge by the droplet during positioning may be expected to occur if the droplet is exposed to the field for times large compared with a few tens of seconds.

Strictly speaking, the reasoning just given refers to accumulation of charge in air. As an alternative model, assume a current density $j = \sigma E$ in the gas and a collection area of πr^2 for the droplet. Then $\dot{q} = \pi r^2 \sigma E$ provides a rough estimate of the rate of increase of charge of the droplet. For $r = 10^{-1}$ cm and $E = 10^3$ V/cm,

this gives $\dot{q} \approx 10^{-14}$ coulombs/sec, which again is found to result in negligible charge accumulation over the time scale of the present experiment but to produce significant charging over times on the order of tens of seconds.

With q taken as constant, Eqs. (1) and (2) may be written in the form

$$\ddot{x} + b\dot{x} = a, \quad \ddot{y} + b\dot{y} = g, \quad (3)$$

the constants a and b being defined in the nomenclature. The straightforward solution, subject to the initial conditions $x(0) = y(0) = 0$ and $\dot{x}(0) = \dot{y}(0) = 0$, clearly has $y = (g/a)x$. Therefore the trajectory of each droplet between the plates is a straight line, inclined to the horizontal, in these experiments. This prediction has been verified by observation and by photography of droplet trajectories.

By use of the known viscosity of air¹¹ it can be shown that the correction arising from the Stokes force is about 0.5% in these experiments, and so $b = 0$ is a good approximation for the present tests. With this approximation, if the droplets are released from a distance y_0 above the capacitor plates then when they reach the upper edge of the plates their velocity will be $\dot{y} = (2gy_0)^{\frac{1}{2}}$. To obtain an approximate solution for the trajectory, this value may be used as an initial condition in Eq. (3) which describes the motion of the droplet

between the plates. With $b = 0$ it is found that $x = at^2/2$ and $y = (2gy_0)^{\frac{1}{2}}t + gt^2/2$, which implies that

$$y = (g/a)x + 2(gy_0x/a)^{\frac{1}{2}} \quad (4)$$

Hence the trajectory of a droplet released above the capacitor is a curved line while the droplet is between the plates. This prediction also has been verified by experimental observation.

5. Experimental Apparatus and Procedure

About 150 droplet tests were conducted in the experimental program. The test set-up (Figs. 1 and 2) comprised the electrostatic field and the droplet release systems. In several tests an AE-1 Canon camera was used for photography of the droplet trajectory. The result shown in Fig. 3 illustrates the straight-line trajectory of droplets released between the plates. The increase in the spacing of the droplets as they move downward is indicative of their acceleration.

The electrostatic field system consisted of a parallel-plate capacitor (two aluminum plates, 20 cm in diameter, 0.5 cm width) and a power supply of 0 to 50 d.c. kilovolts (KV) — Integrated Laser Modulator, model ILM-70, by Electro-Optical Instruments, Inc. One plate was grounded (to the ground of the power supply), the other plate

being connected to the positive high voltage.

The droplet releasing mechanism consisted of a glass container with two narrow openings opposite each other, the lower one connected to a glass pipette (inner diameter varied from 5 to 0.5 mm), and the upper connected to vacuum (0.1 to 1 atm) through two continuous valves, one having an opening to ambient pressure. By adjusting first the vacuum valve and then the ambient valve, the pipette is caused to release droplets, one by one, as needed. To prevent possible secondary charging effects, the liquid in the pipette (and in the container) was grounded.

Two series of tests were completed. First the feasibility of the method was studied with water, and later tests were performed with heptane (hydrocarbon fuel). Droplets were released 1 cm apart from a position adjacent to the grounded plate, the voltage being adjusted until the droplets hit the bottom of the other plate. This technique was selected because it provided the most accurate means for determining the droplet trajectory. The distance between the plates was varied from 2 to 7 cm (6 positions), forcing the droplets to move horizontally 1 to 6 cm. In the vertical direction, the origin y_0 for the released droplet varied from 20 cm above the plates to the center of the plates, in increments of 10 cm (4 positions) with water, and from 10 cm above the plates to the center of the plates in increments of 5 cm (5 positions) with heptane. Each test was repeated about three times, the average

value of voltage of all runs being recorded. Typical variations in voltage were $\pm 5\%$, the differences being attributable to alignment, conductivity and polarizability variations.

6. Results and Discussion

The objective of these experiments was to obtain values for the inductively produced charge q of the droplets. This value must be available if a suitable positioning device, based on this electrostatic principle, is to be designed. With conditions adjusted so that the droplet just touches the lower edge of the positively charged plate, the values of x and y are known accurately at the end of the trajectory. By solving Eq. (4) for $\underline{a}$, it may be shown from the definition of $\underline{a}$ that

$$\frac{q}{m} = \frac{gx}{(V/d)y^2} \left[y + 2y_0 + 2\sqrt{y_0(y+y_0)} \right], \quad (5)$$

which provides the charge-to-mass ratio in terms of measured quantities. The time interval that the droplet spends within the electric field is also obtained by solving a quadratic, viz.,

$$t = \left(\sqrt{y+y_0} - \sqrt{y_0} \right) / \sqrt{g/2} \quad (6)$$

These results are to be used for $y_0 > 0$; if the droplet is released from between the plates then $q/m = g(x/y)/(V/d)$ and $t = \sqrt{g y/2}$, y now being measured from the point of injection rather than from the top of the plates. The time intervals that the released droplets spend within the electrostatic field are listed in Table 1.

Note from Eq. (5) that the charge-to-mass ratio may be obtained without measuring the mass of the droplet. The charge per unit surface area of the droplet also may be of interest, but obtaining this requires measuring the droplet radius. A quantity proportional to the charge per unit surface area is

$$\therefore \quad q/r^2 = (4/3) \pi r \rho(q/m) \quad (7)$$

It was observed that when the droplet was released from inside the electric field, its radius grew, depending on the voltage, before it became detached from the pipette. An approximate linear law was determined experimentally for the growth, namely,

$$r = r_0 (1 + kV) \quad (8)$$

Values of the growth factor k are given in Table 2. It is noted that k is smallest in the center of the field. This may indicate that a non-uniform field enhances the growth effect. Use of Eq. (8) enables q/r^2

to be calculated from Eqs. (5) and (7). Relevant physical properties of water and heptane, the first of which is used in this calculation, are listed in Table 3.

The simplest result to derive from the experimental observations is the dependence of the voltage between the plates on the horizontal distance of droplet travel, under the condition that the droplet impinges on the lower edge of the positive plate. Such results are shown in Figs. 4 and 5, for water and heptane respectively, for different elevations of droplet release. Positive values of y_0 correspond to release positions above the upper edge of the plates, and negative values between the plates. The near linearity of these plots suggests that if the electric field intensity were plotted instead of the voltage, then the values obtained would be nearly constant, for fixed values of y_0 . Figures 6 and 7 reveal that this is true only approximately. There is a tendency for the necessary field intensity to decrease as the distance of travel, or plate separation, decreases. It is understandable that higher field intensities are needed at larger values of y_0 because the velocity at which the droplet is traveling when it reaches the upper edge of the plates increases as y_0 increases.

The relationships shown in Figs. 4 through 7 enable the charge-to-mass ratios, calculated from Eq. (5), to be plotted as a function of the voltage difference between the plates, the electric field intensity or the horizontal distance of droplet travel, for various

values of the vertical position of droplet release. Such graphs are shown in Figs. 8, 9 and 10, respectively, for water and in Figs. 11, 12 and 13, respectively, for heptane. It is seen that typical charge-to-mass ratios for water droplets, achieved by induction in these experiments, are on the order of 10^{-5} coulombs/kg, while those for heptane are about an order of magnitude lower. The low polarizability of heptane in comparison to that of water is likely to be the cause of this difference.

Although the graphs as a function of the electric field intensity may be expected to have the greatest fundamental significance (Figs. 9 and 12), the curvatures in these plots make extrapolation of the data difficult. Extrapolation appears to be more straightforward if the voltage or the horizontal distance of travel is used as the horizontal scale, and the dashed lines in the figures illustrate such extrapolations. It is evident especially from Figs. 9 and 12 that as the elevation of the point of droplet release increases, the induced charge-to-mass ratio decreases. Stated differently, a higher electric field intensity between the plates is needed to produce the same charge-to-mass ratio at a larger value of y_0 . This is understandable for positive values of y_0 because the field intensity at the point of droplet release then decreases rapidly with increasing y_0 . However, it is somewhat surprising in Fig. 12 to find a measurable difference between the curves for $y_0 = -5\text{cm}$ and $y_0 = -10\text{cm}$. The variation in droplet mass, as

implied by Table 2, may be somewhat responsible for this result; the droplets at $y_0 = -5$ cm are somewhat larger than those at $y_0 = -10$ cm, and so if the charge does not increase in proportion to the mass, the charge-to-mass ratio will be smaller at $y_0 = -5$ cm.

The extrapolations in Figs. 8 and 11 suggest that even for zero potential difference a negative charge on the droplets may exist. Such charging may result from frictional effects operative during droplet injection and appear to lie in the range of 10^{-6} coulombs/kg for water and 10^{-7} coulombs/kg for heptane. These charges may be particularly important in droplet-positioning devices if the droplet is released in the absence of the electrostatic field, a procedure which appears desirable from the viewpoints of both convenience and safety. Therefore it is unfortunate that the present experiments have not been able to quantify this effect of residual charge more accurately.

Graphs of q/r^2 , corresponding to Figs. 8 through 13, may be obtained by use of Eq. (7). These results are plotted in Figs. 14 through 19. It is seen that the charge per unit surface area, induced on the water droplets, is on the order of 10^{-9} coulombs/cm², while that for heptane is about 10^{-10} coulombs/cm². The same trends that were evident in the q/m curves again are seen. Extrapolated values for zero potential difference are 10^{-10} coulombs/cm² for water and 10^{-11} coulombs/cm² for heptane.

Because of the very low electrical conductivity of heptane¹² (a factor of 3×10^6 less than that of water, see Table 3), it is to be anticipated that heptane will require a greater amount of time than water to acquire its induced charge. This expectation was borne out experimentally. About 10 seconds of exposure to the electrostatic field were required for the experimental results with heptane to be reproducible. The corresponding time delay for water was too short to be measured. Heptane and water also have different values of surface tension (Table 3). Partially because of this, the droplets formed by the two liquids may be of different sizes under otherwise identical conditions. Experimentally, droplet diameters were about 0.30 cm for water and 0.25 cm for heptane.

7. Concluding Remarks

There are obvious limitations in attempting to develop, through earthbound experiments, design of an apparatus for positioning of droplets electrostatically under conditions of negligible gravity. For example, residence times of droplets between the plates of the condenser in the present experiments were in the range of 0.1 to 0.2 sec., well below the residence times in gravity-free environments. Therefore relatively high fields, up to 11 KV/cm, were needed in the present experiments to measure droplet deflections. In the absence of gravity, weaker fields may be used, since times on the order of 10 sec. or more are expected to be available for adjusting the position of a droplet. These longer time

frames may have two effects. First, accumulation of charge during positioning, which was negligible in the present experiments, may be significant in the zero-g application; this phenomenon would enhance the effectiveness of the positioning device. Second, the field intensity required to effect positioning at zero g is likely to be appreciably less than that required to make the present laboratory measurements. For example, a heptane droplet with a charge of 10^{-6} coulombs/kg can be caused to move from rest a distance of 10 cm in 10 sec by means of a field as low as 20 V/cm, according to the design equation

$$x = (V/d)(q/m) t^2/2, \quad (9)$$

obtained through integration of Eq. (1) with drag neglected. Thus, the electrostatic method appears to be relatively promising for applications in which the acceleration of gravity is negligible.

The present experiments have served to determine the order of magnitude of the charge per unit mass on droplets released from a pipette in the vicinity of an electrostatic field. Although this information is helpful in developing a design of an electrostatic positioner, there remain uncertainties that should be considered during design. The magnitude of the electric field has a large influence on the droplet charge, and the charge will be small for a droplet released prior to initiation of

the field. Moreover, this initial charge may depend appreciably on the mechanism of droplet release. Although it may be possible to employ β -ray emitters to impart a charge to a droplet released with negligible charge, the present experiments suggest that provision for this is not likely to be necessary. Instead, it seems reasonable to provide a margin of safety in the positioner design by employing electrodes that can be charged to the kilovolt range, the potential being adjustable to 1% (e. g., 50 to 5000 V D. C.). With such a design, large enough field intensities will be available to cause the droplet to collect charge during its motion, if necessary, while at the same time fields as low as 10 V/cm can be controlled.

A schematic illustration of a positioner design, consistent with the dimensions of the combustion facility of Spacelab,¹ is shown in Fig. 20. This design is intended to provide flexibility, so that different modes of operation can be explored under zero-g conditions, and the most convenient mode can be selected. Note that in providing easy external access to the volume in which the droplet is to be confined, the design allows some divergence of lines of electrostatic force, thereby potentially making some use of the polarizability of the liquid as well. Although successful operation of the device requires zero-g testing for verification, there is every reason to believe, based on the experiments reported herein, that the device will perform its positioning function successfully.

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Table 1. Time Intervals in the Electrostatic Field

$y_0(\text{cm})$	20	10	5	0	-5	-10
$t(\text{sec})$	0.0836	0.1045	0.125	0.202	0.175	0.143

Table 2. Values of the Droplet Growth Factor k (%/KV)

$y_0(\text{cm})$	0	-5	-10
water	3	-	1.5
heptane	2	1.5	1

Table 3. Properties of Water and Heptane

property	density ¹¹ (g/cm ³)	electrical conductivity ¹² ohm ⁻¹ cm ⁻¹	dipole moment ¹¹ (10 ⁻¹⁸ e. s. u.)	surface tension ¹¹ in air at 20 °C (dynes/cm)
water	1	10 ⁻⁶	1.87	73
heptane	0.684	$\sim 0.33 \times 10^{-12}$	~ 0	~ 40

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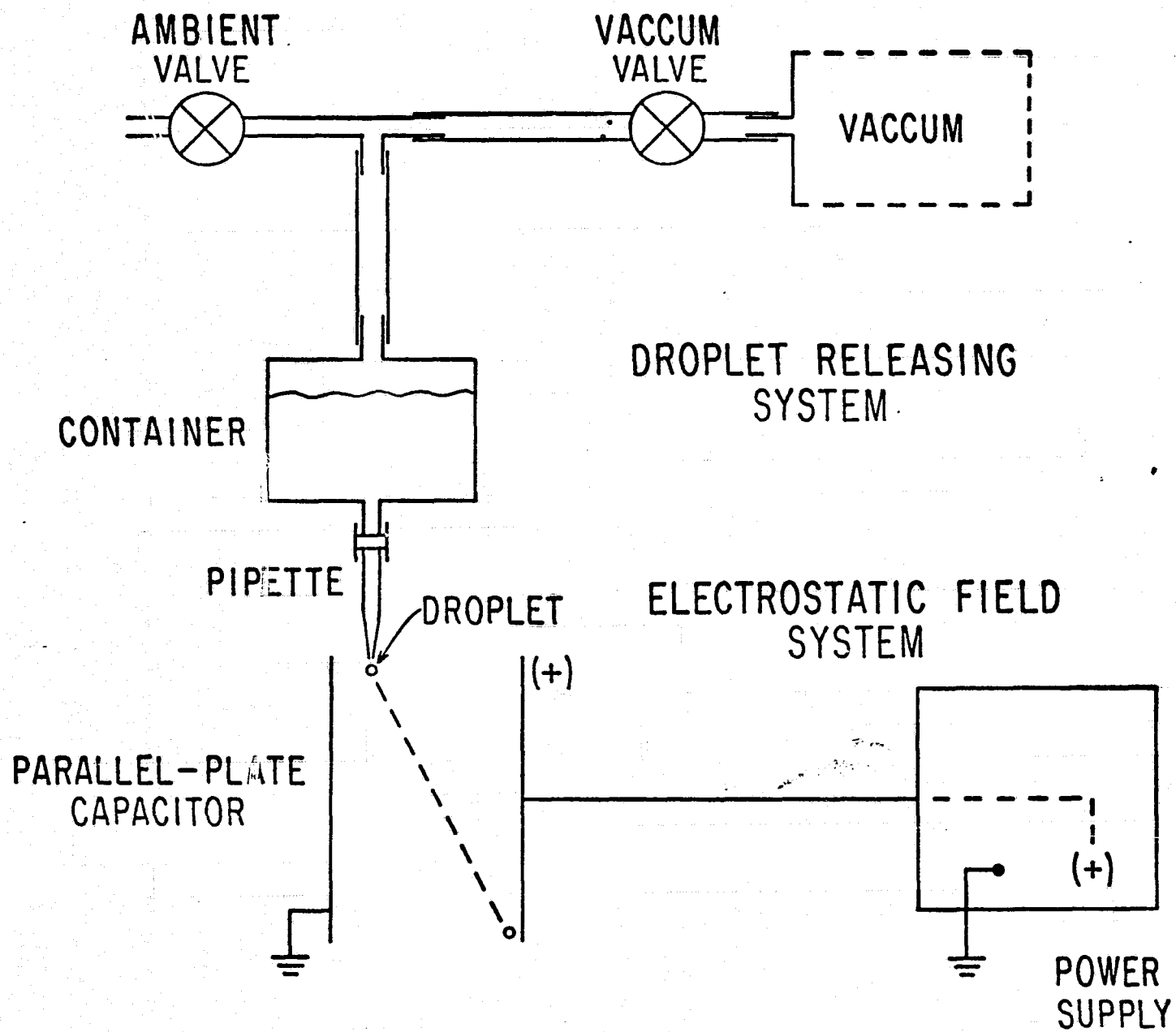


Fig. 1

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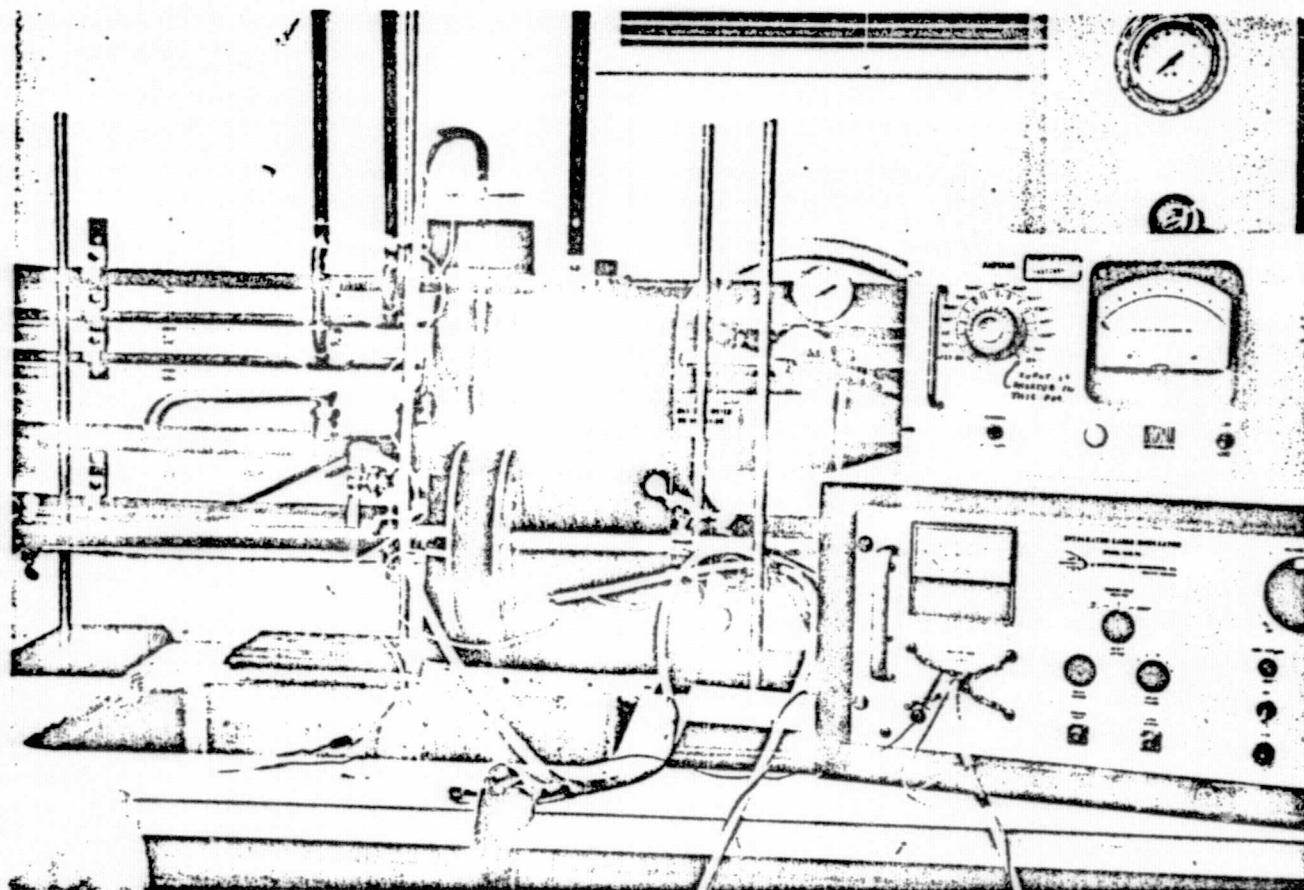


Fig. 2

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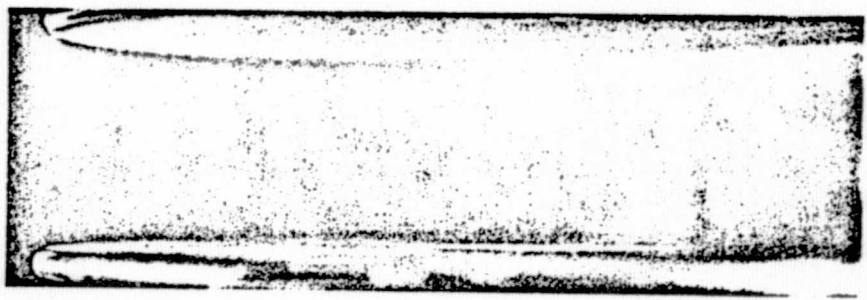


Fig. 3

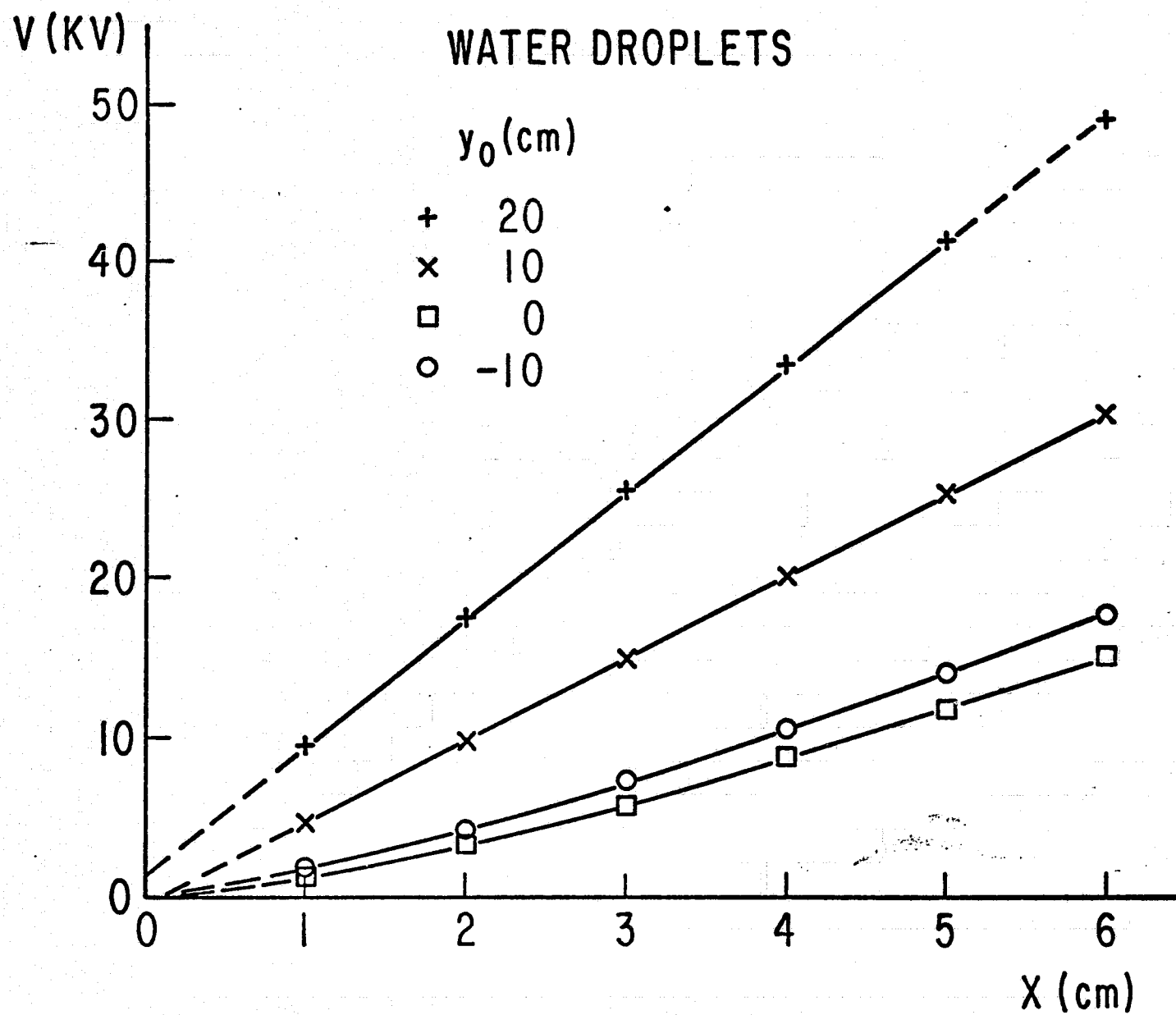


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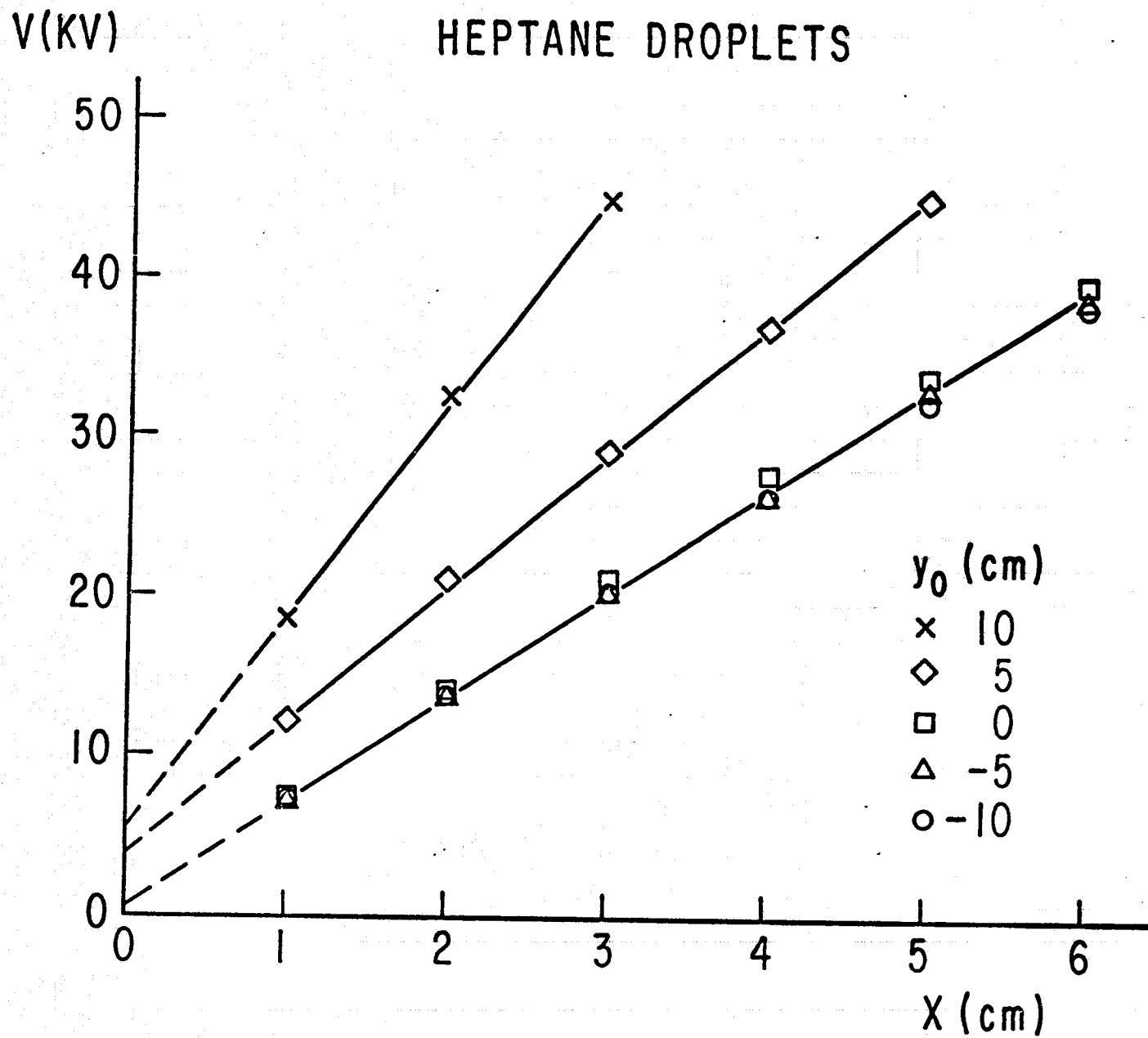


Fig. 5

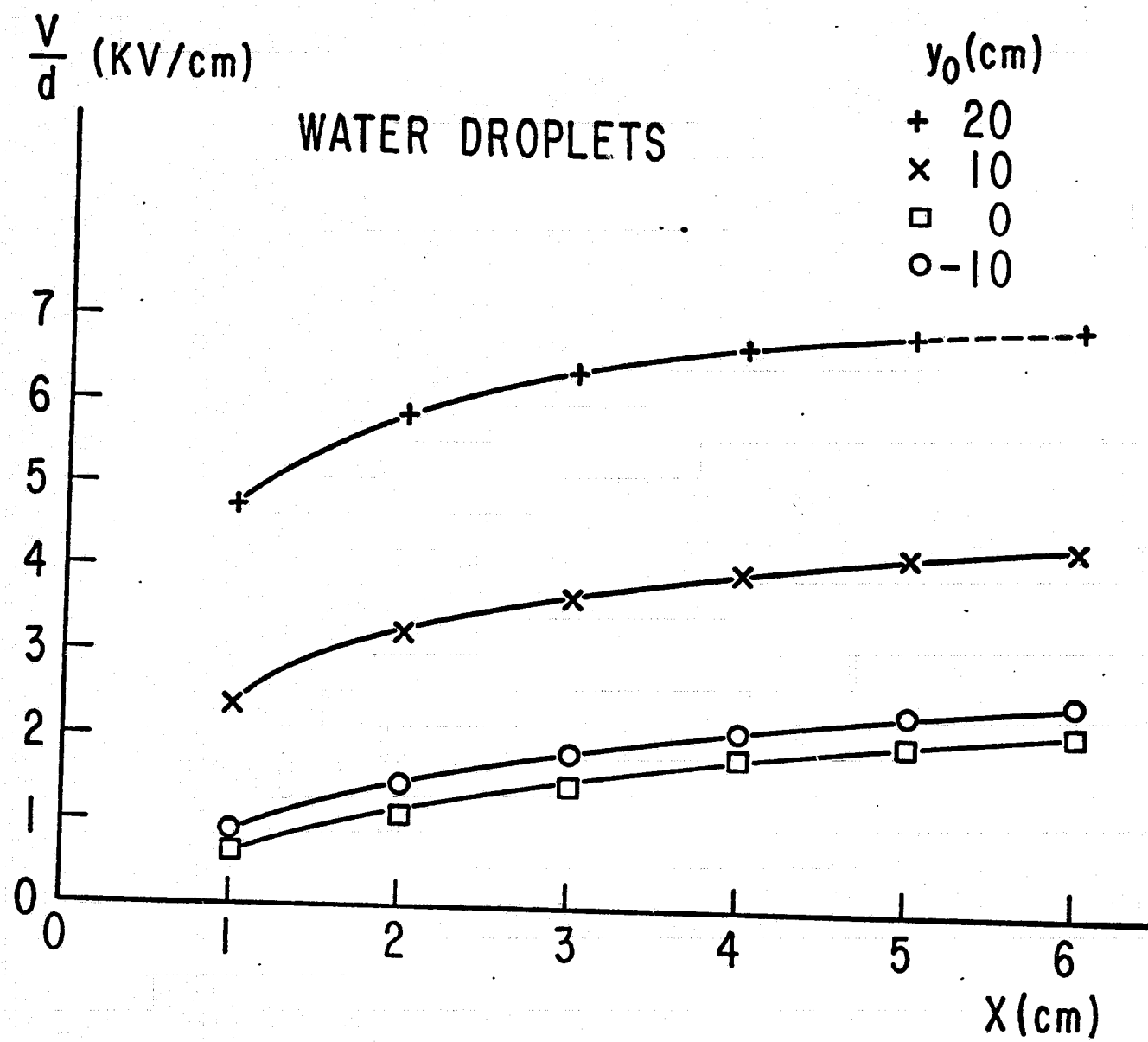


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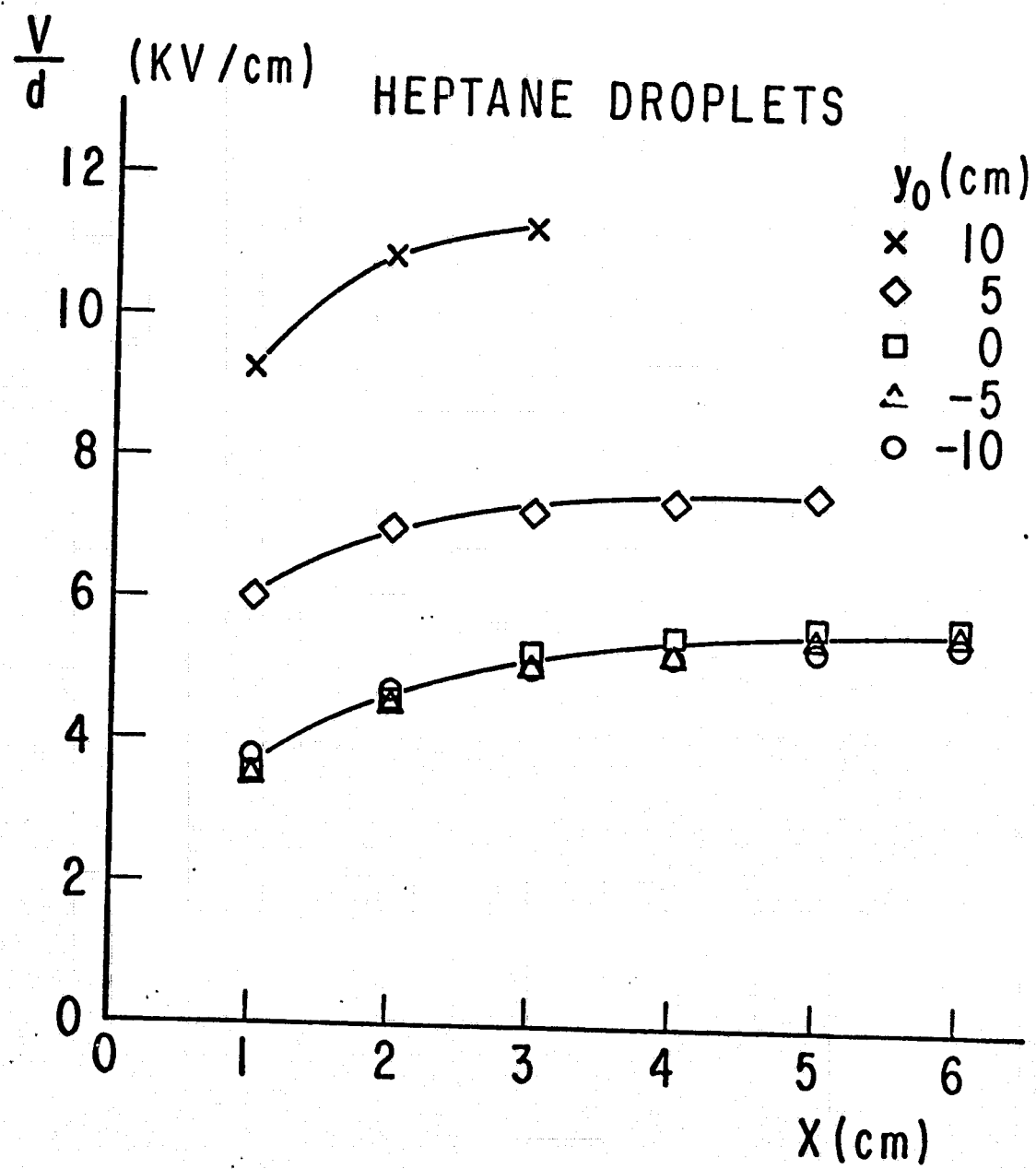


Fig. 7

$\frac{q}{m} \left(\frac{10^{-5} \text{ coul.}}{\text{kg}} \right)$

WATER DROPLETS

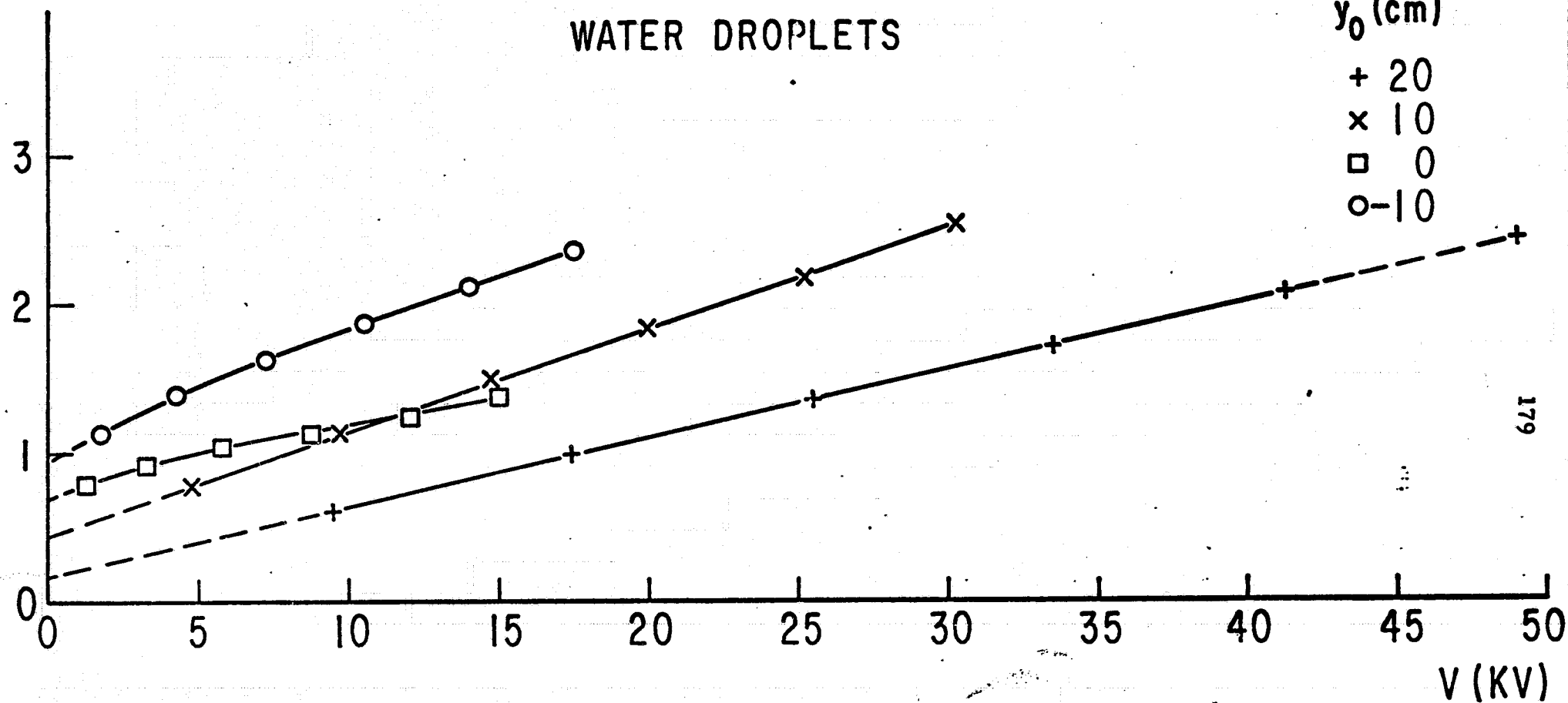
y_0 (cm)

+ 20

x 10

□ 0

○ -10



179

Fig. 8

$$\frac{q}{m} \left(\frac{10^{-5} \text{ coul.}}{\text{kg}} \right)$$

WATER DROPLETS

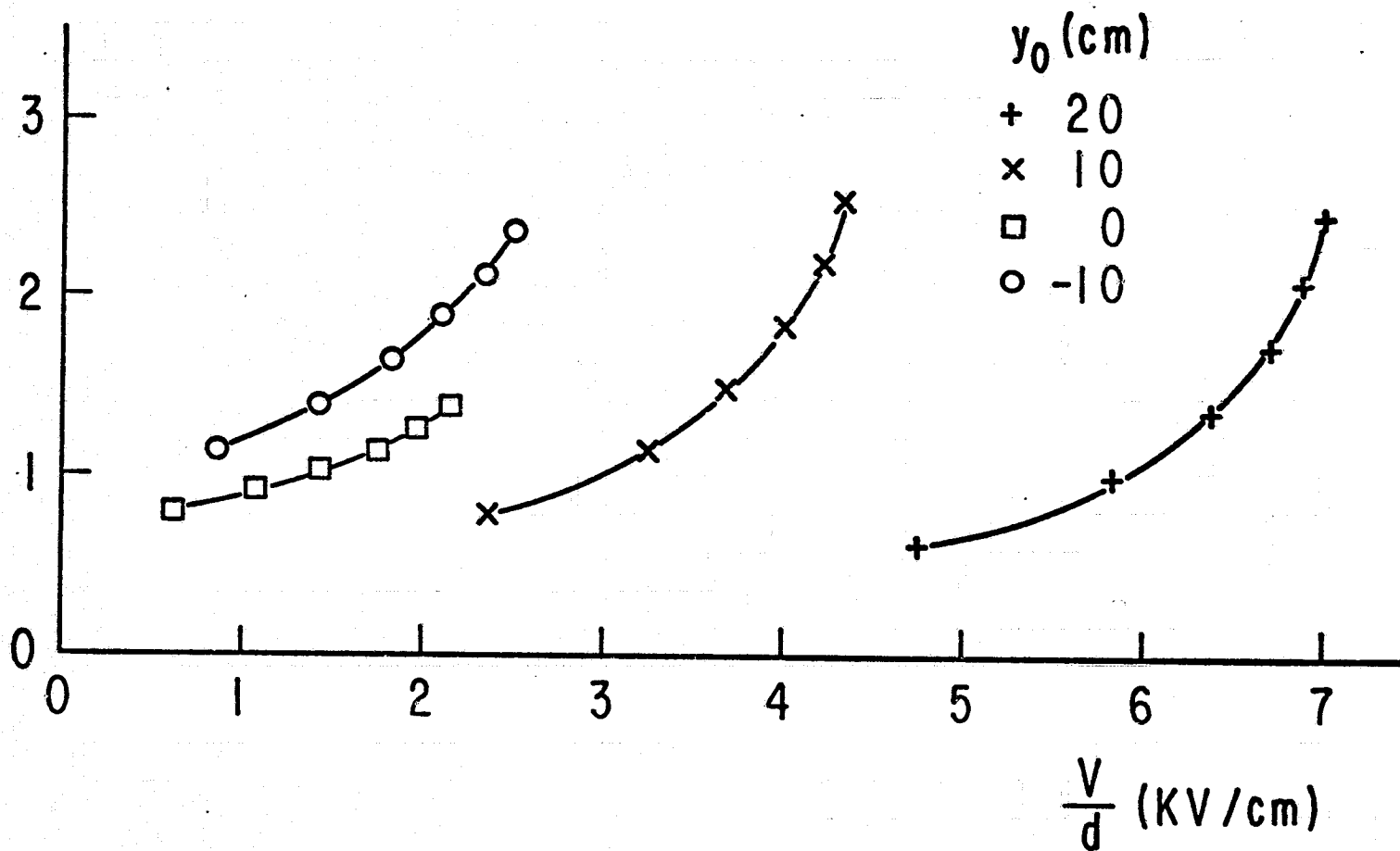


Fig. 9

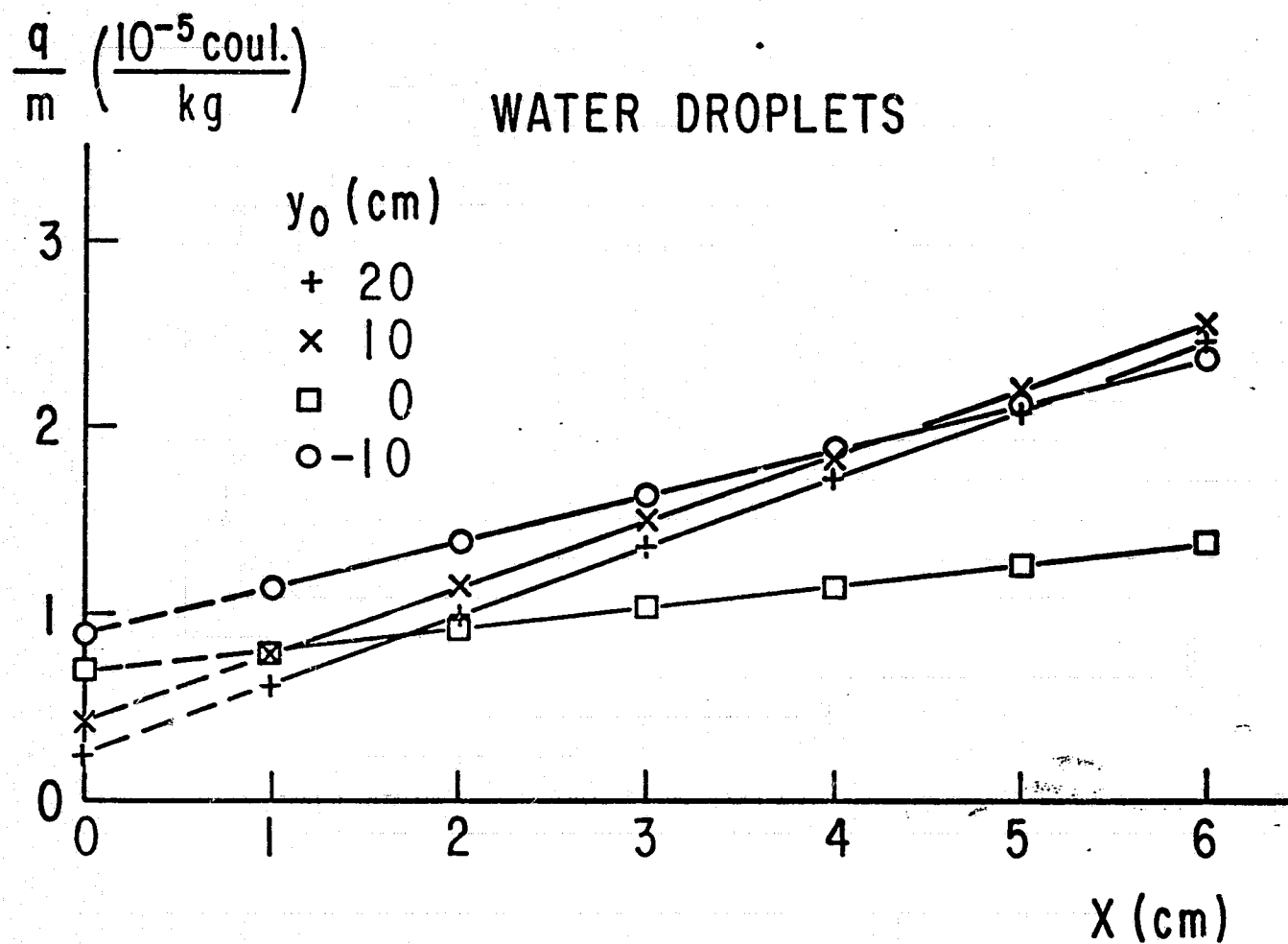


Fig. 10

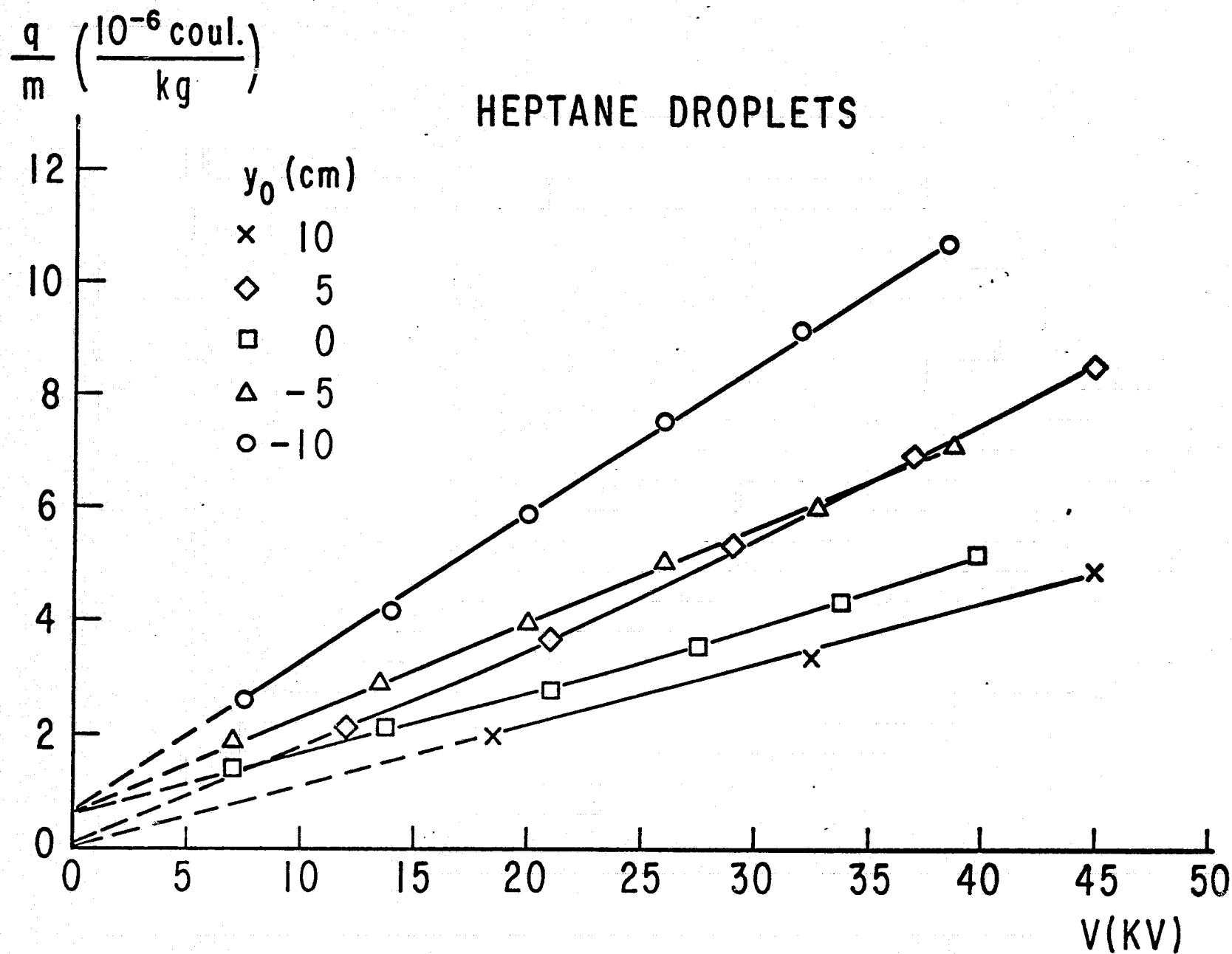


Fig. 11

$$\frac{q}{m} \left(\frac{10^{-6} \text{ coul.}}{\text{kg}} \right)$$

HEPTANE DROPLETS

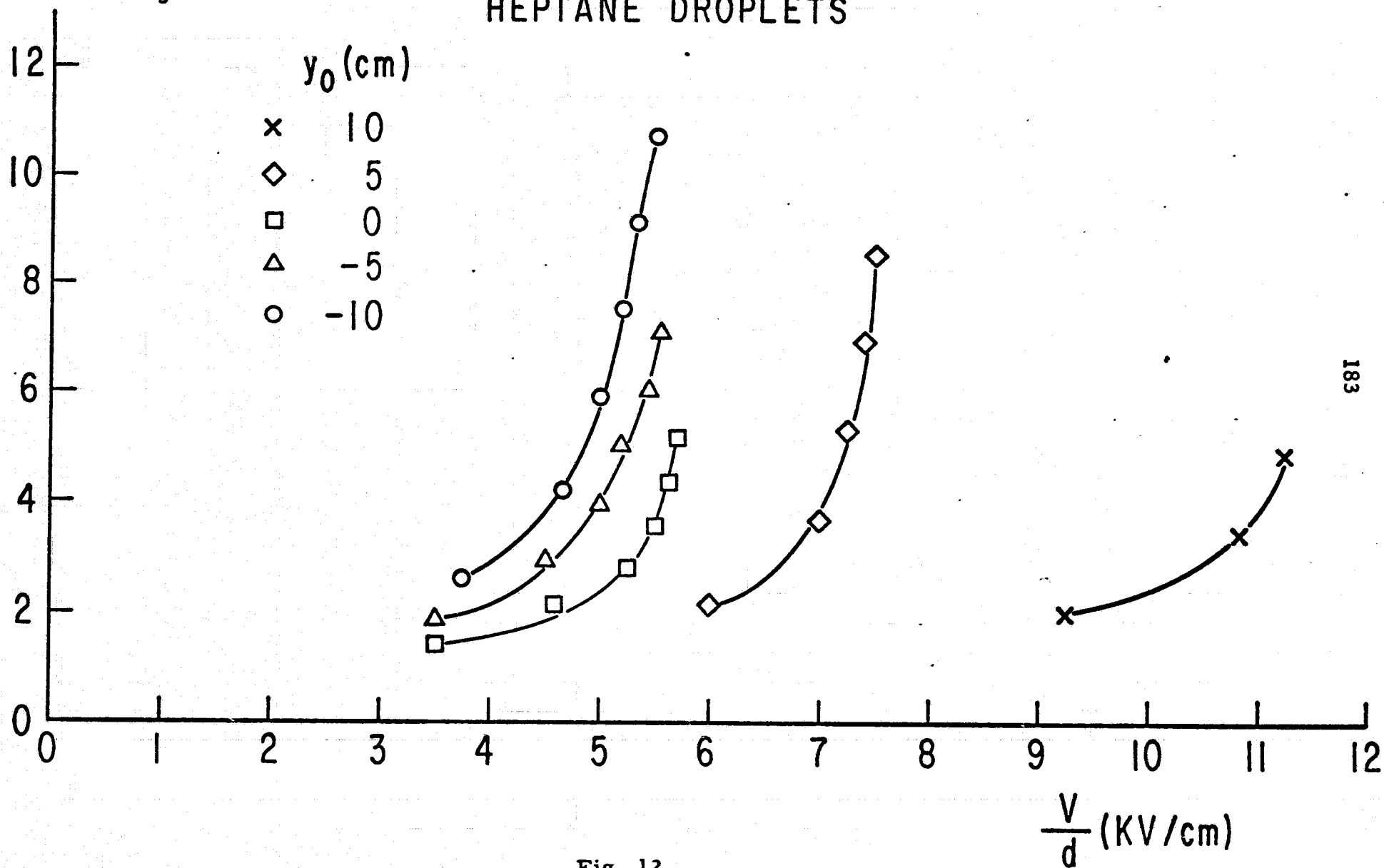


Fig. 12

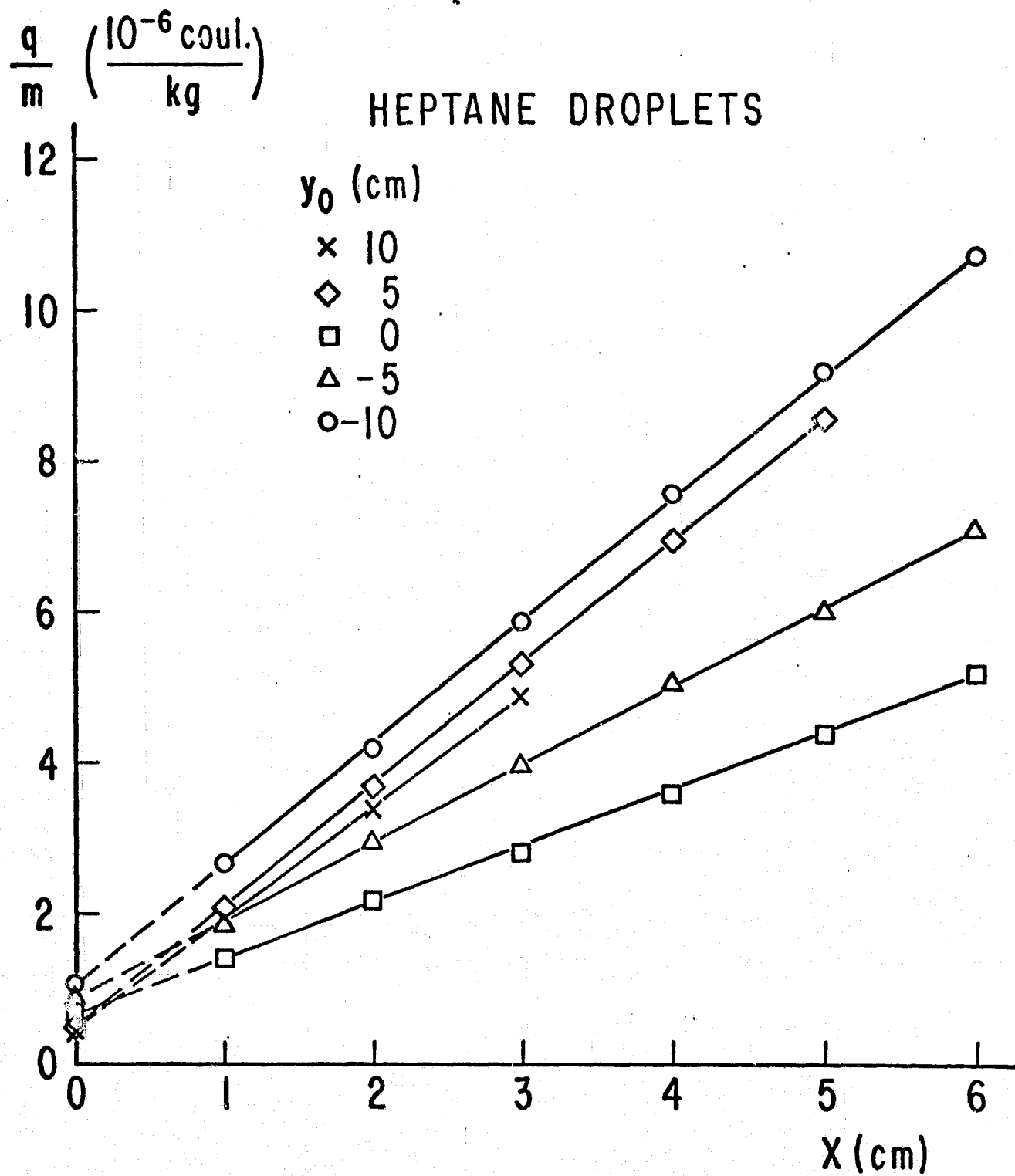


Fig. 13

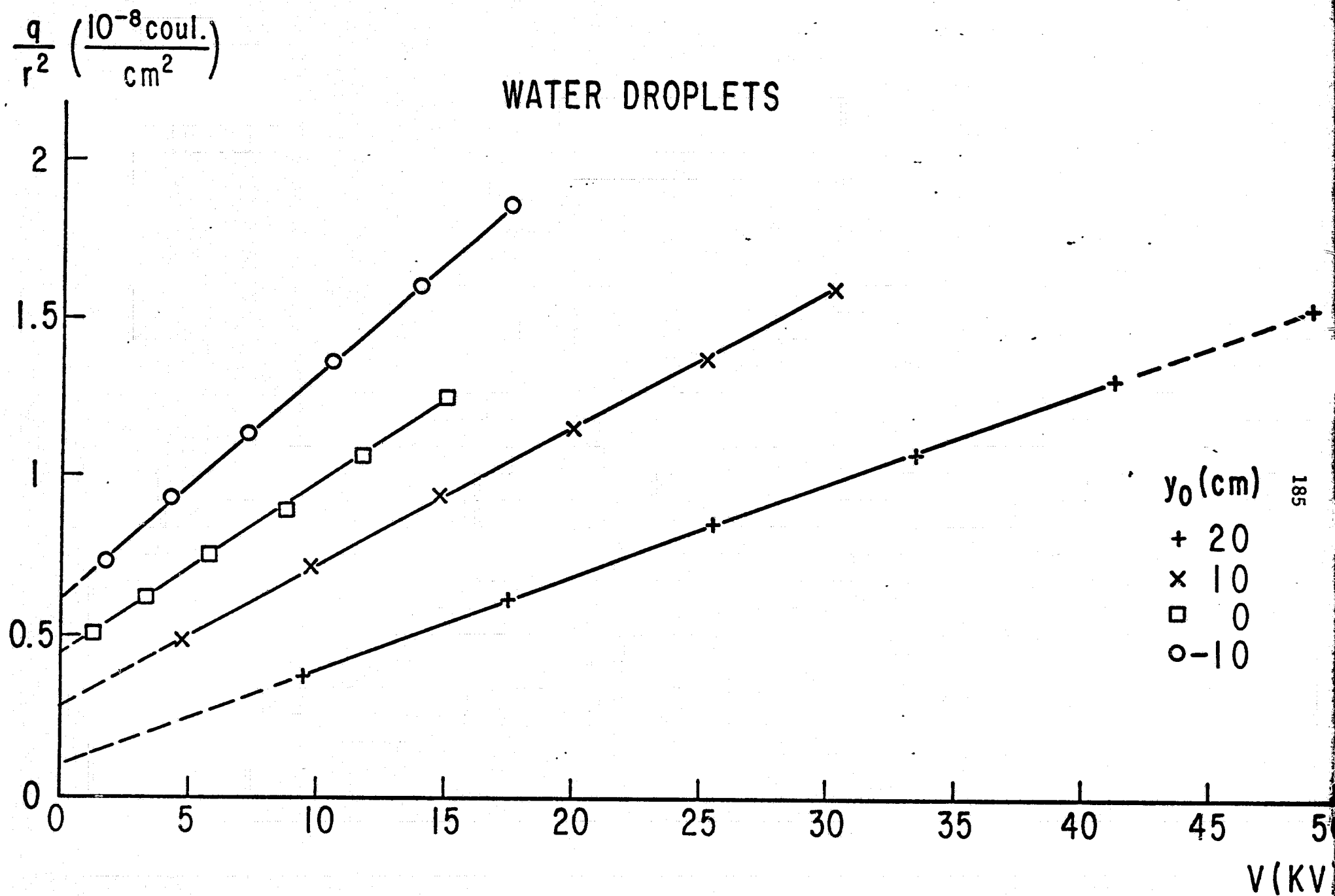


Fig. 14

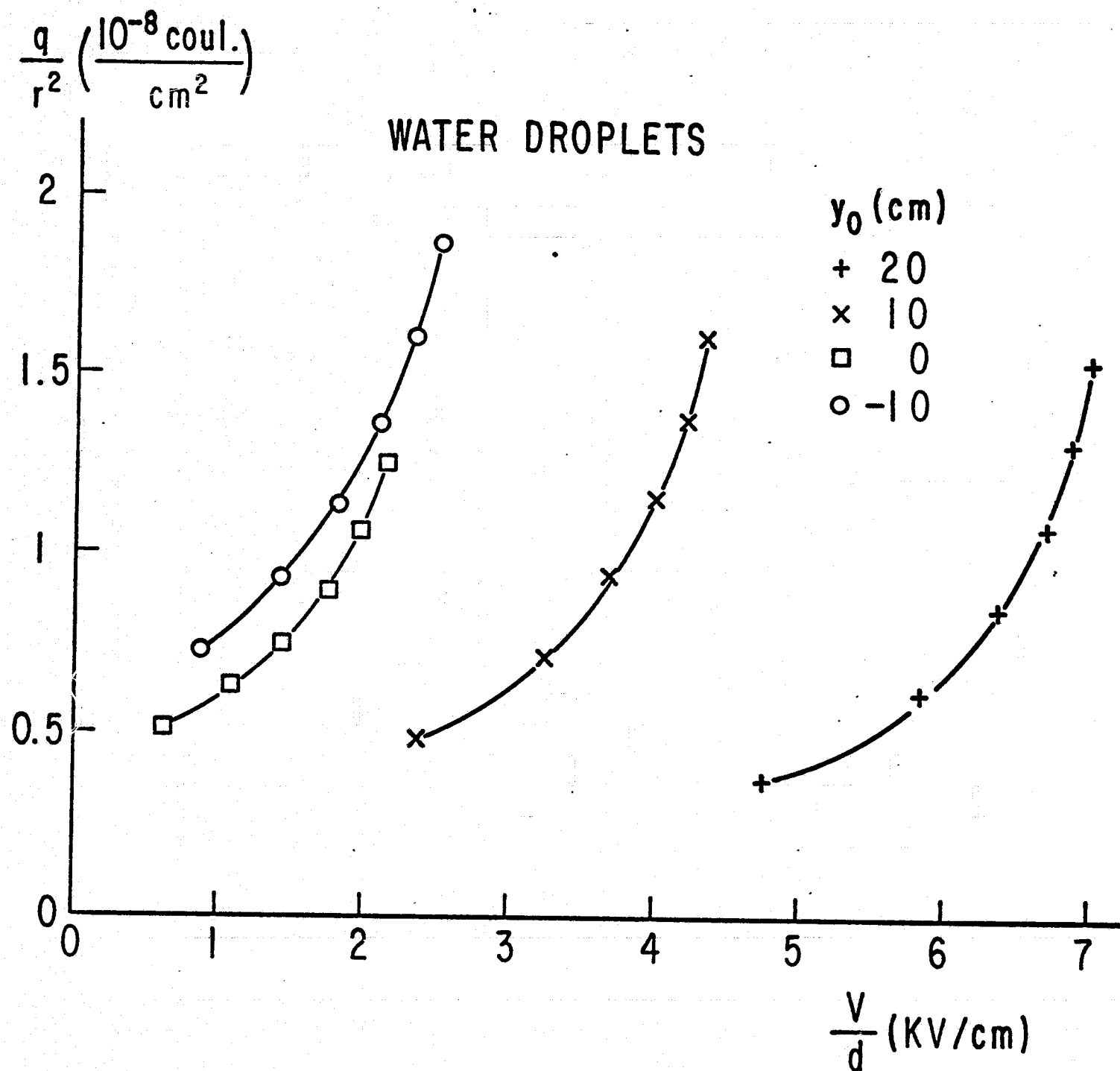


Fig. 15

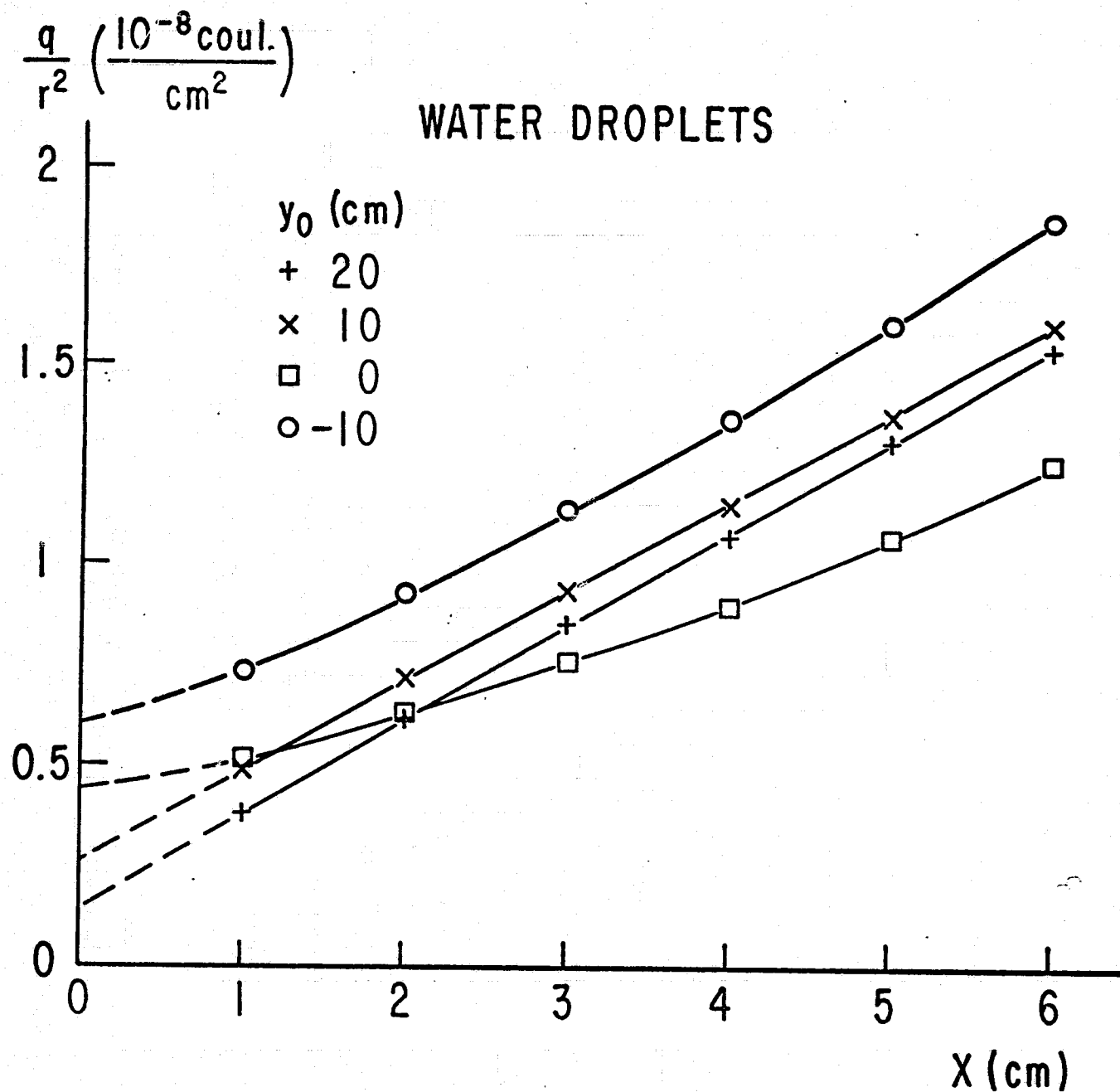


Fig. 16

HEPTANE DROPLETS

$$\frac{q}{r^2} \left(\frac{10^{-9} \text{ coul.}}{\text{cm}^2} \right)$$

y_0 (cm)

x 10

◇ 5

□ 0

△ -5

○ -10

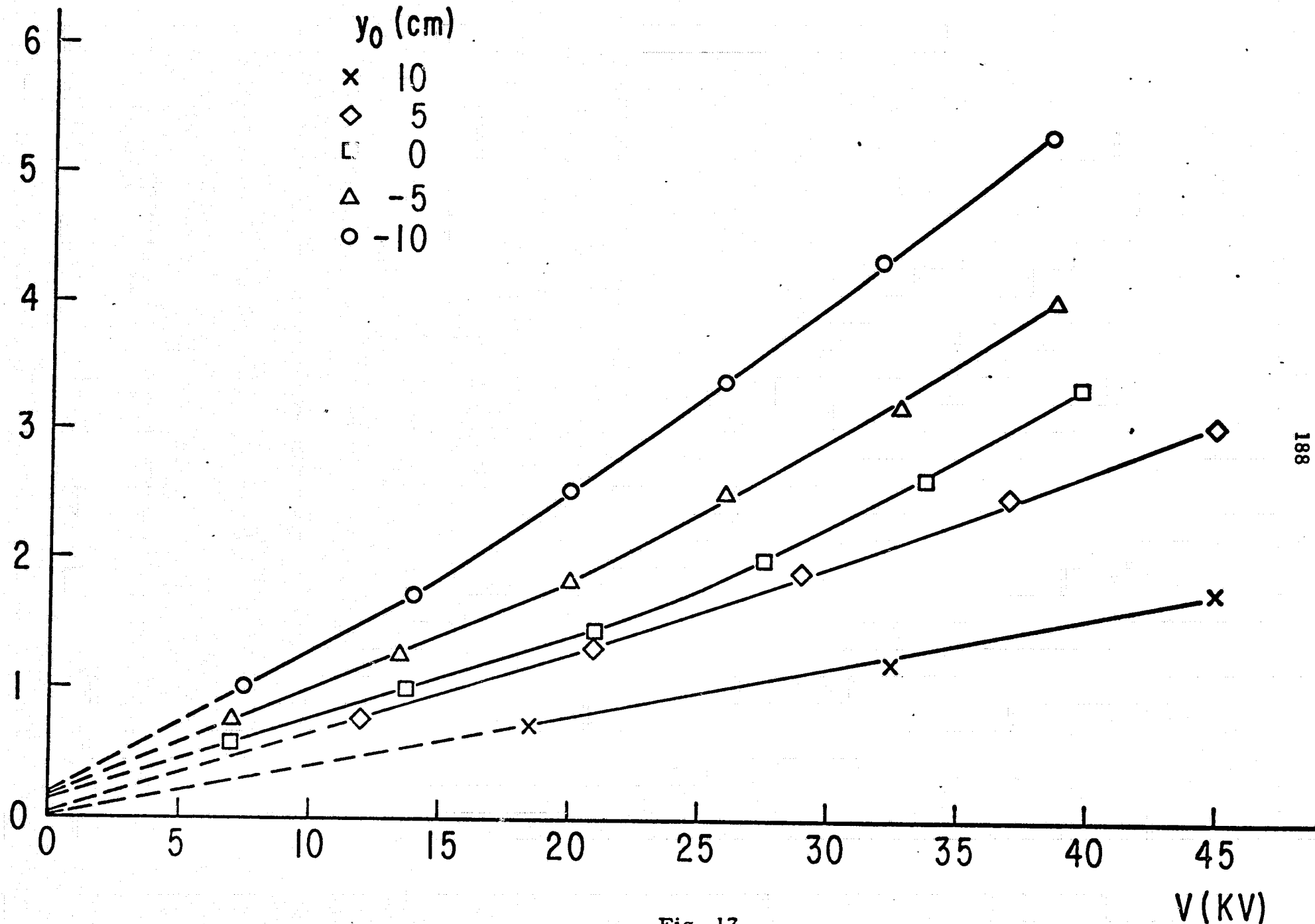


Fig. 17

$$\frac{q}{r^2} \left(\frac{10^{-9} \text{ coul.}}{\text{cm}^2} \right)$$

HEPTANE DROPLETS

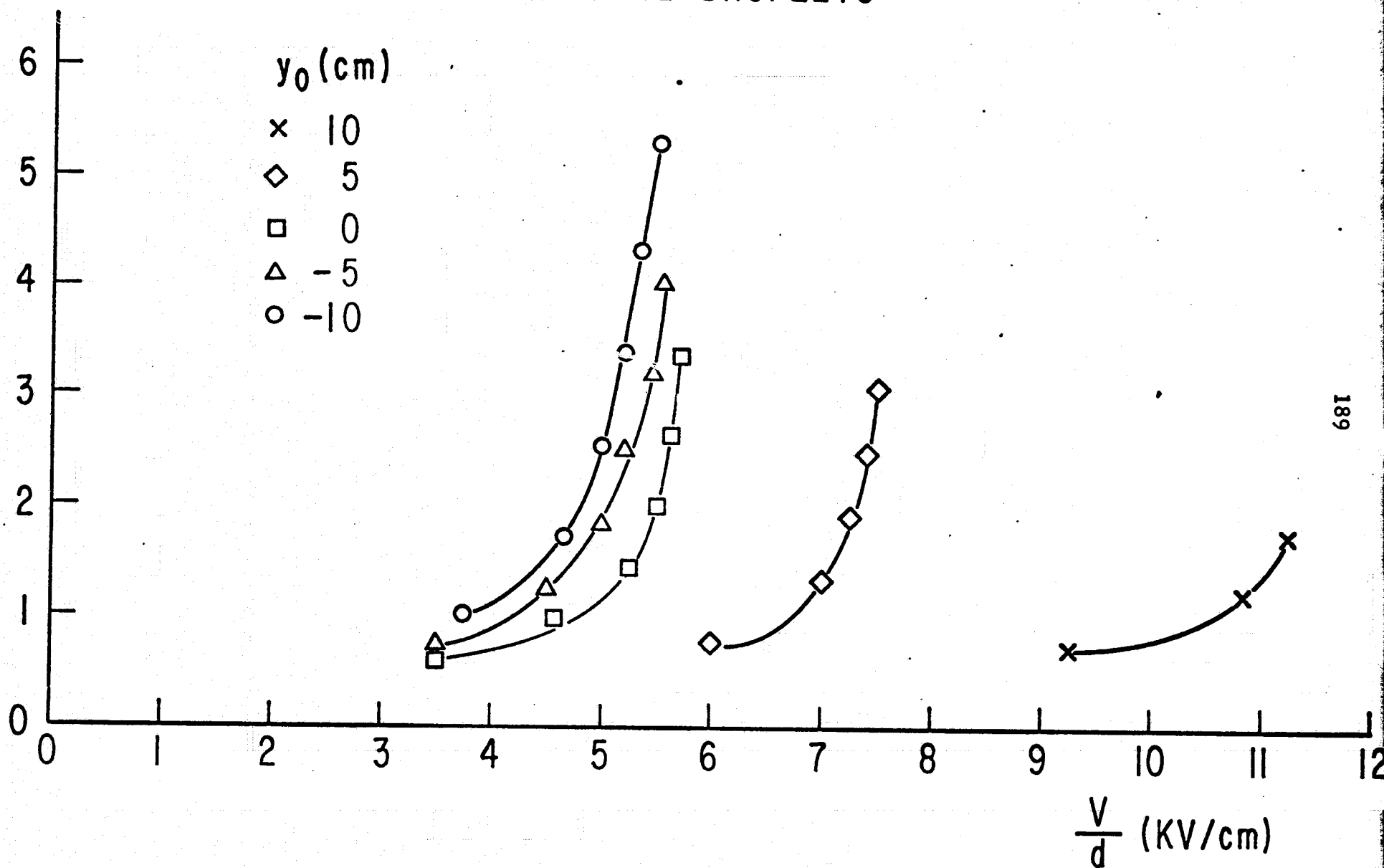


Fig. 18

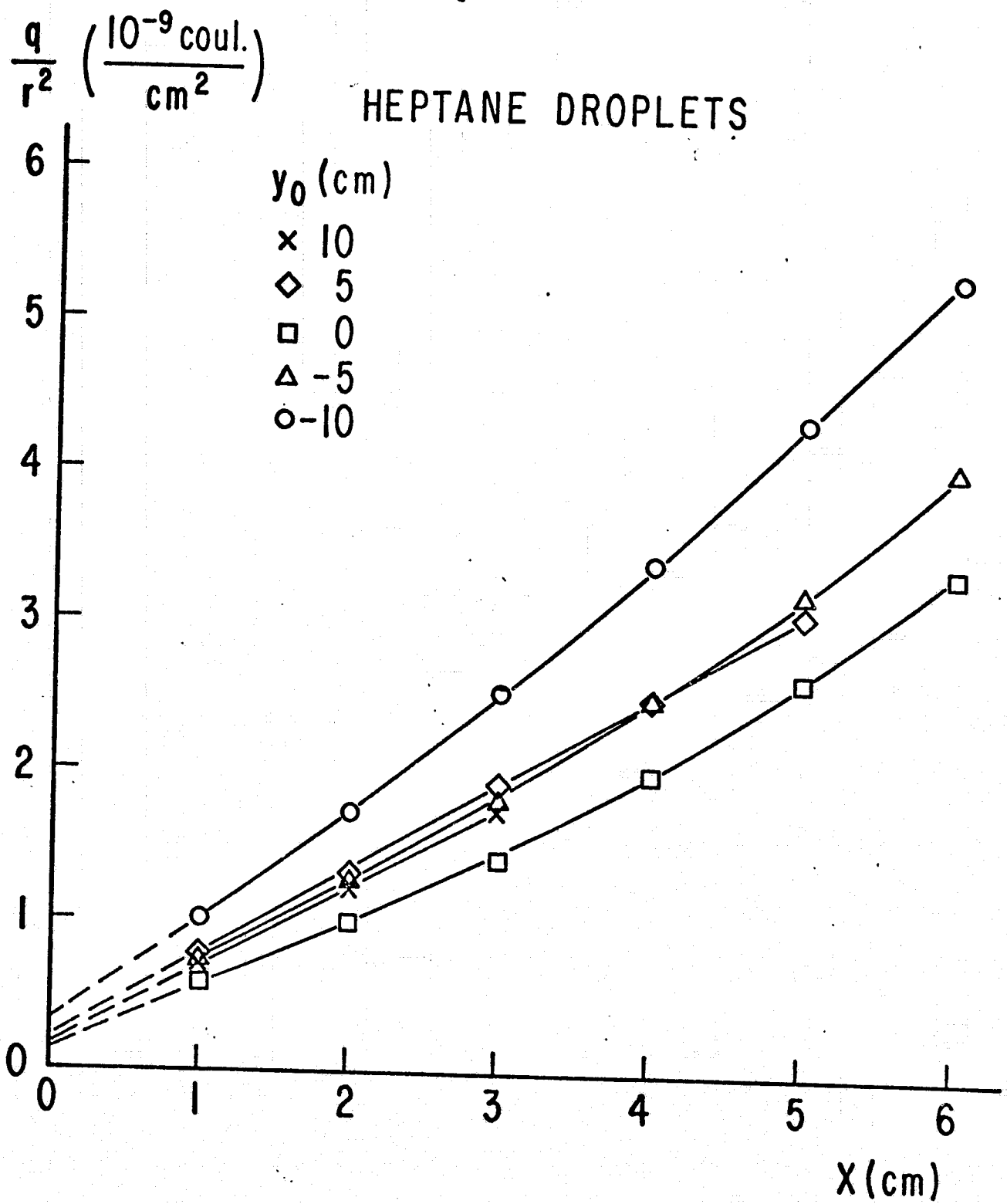
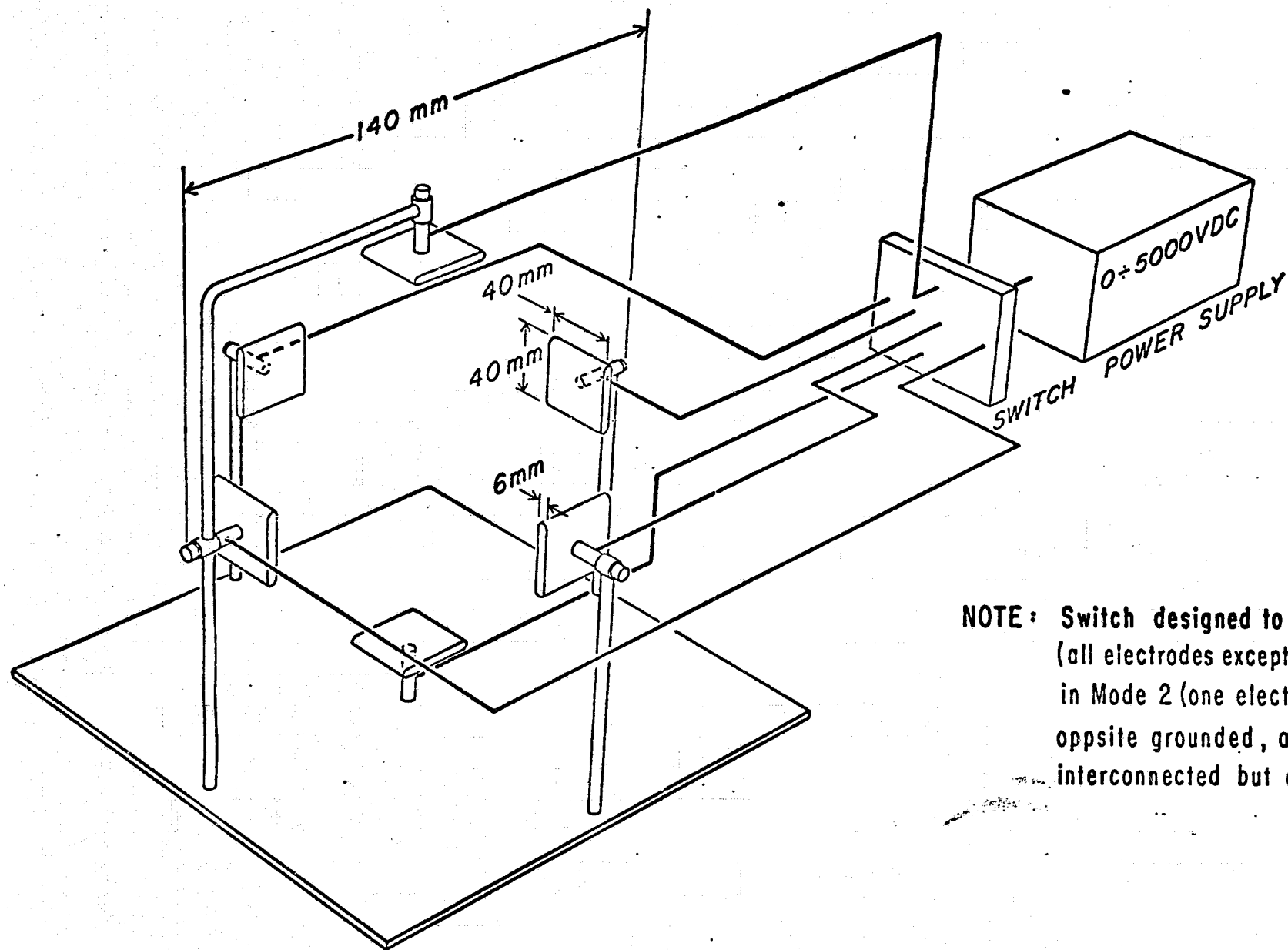


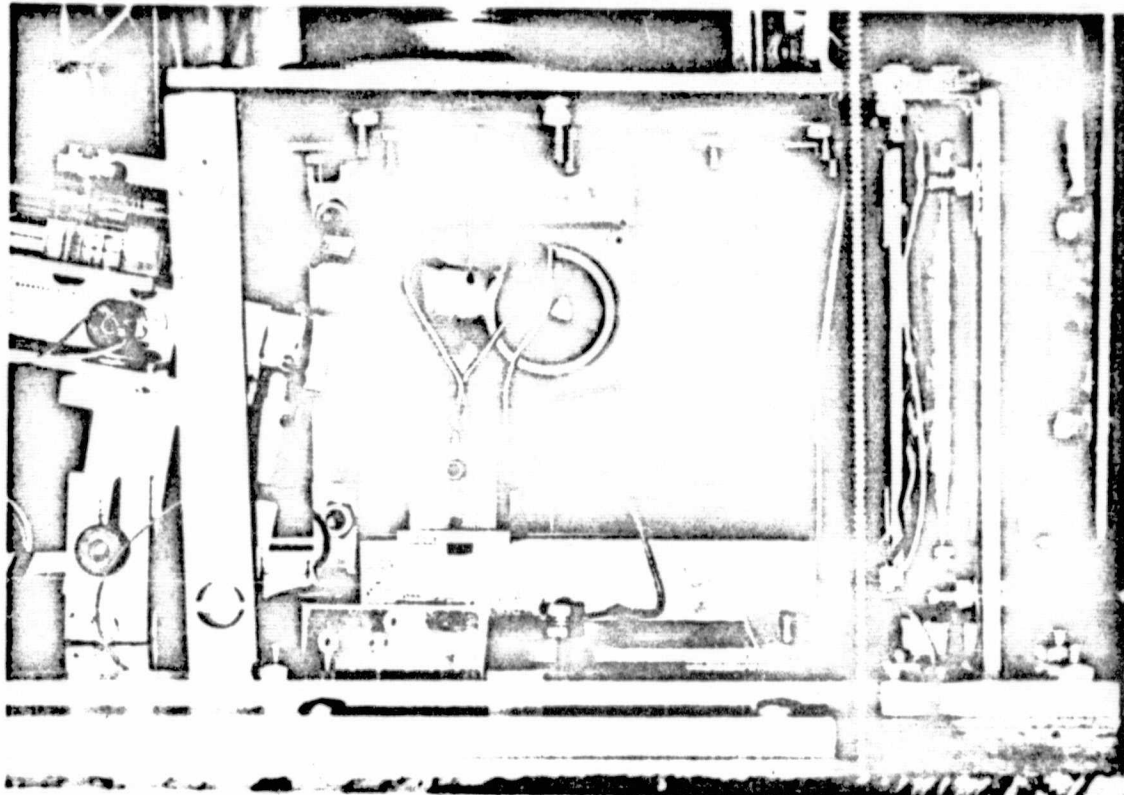
Fig. 19



NOTE : Switch designed to operate in Mode 1 (all electrodes except one grounded) and in Mode 2 (one electrode charged, the opposite grounded, and all others interconnected but electrically isolated.)

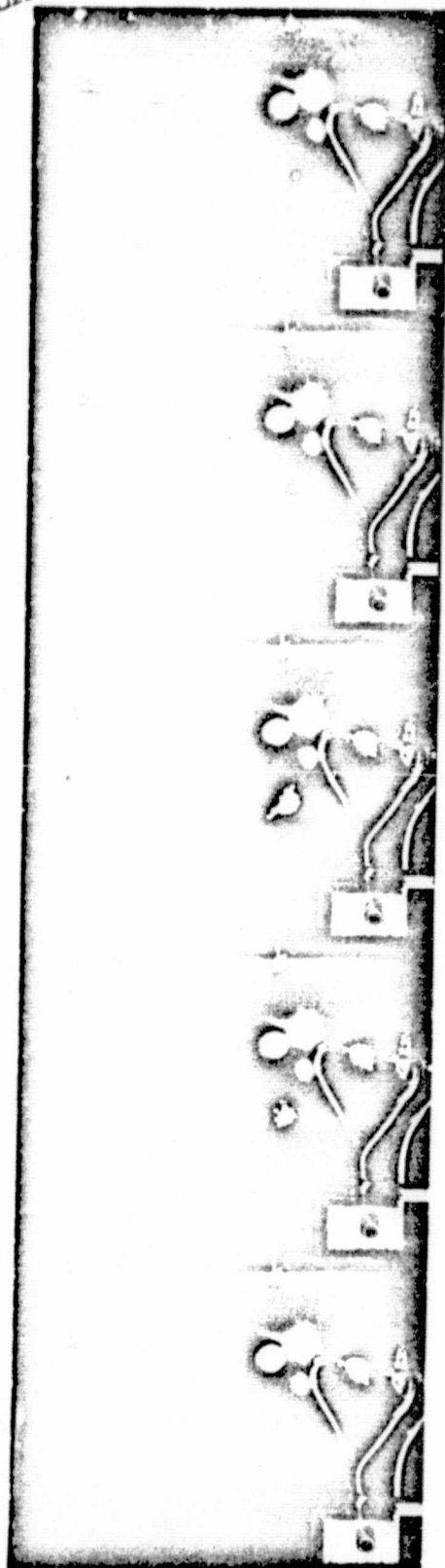
Fig. 20

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VIEW INSIDE COMBUSTION CHAMBER LOOKING TOWARD
CAMERA SHOWING DROPLET MOUNTED ON QUARTZ FIBER
PRIOR TO TEST. DROPLET MOUNTING MECHANISM
AND IGNITION ELECTRODES ARE ALSO SHOWN.

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DECANE SATELLITE
EXPERIENCING

"FLASH EXTINCTION"

$$Y_{O_{2\infty}} = 0.5$$

$$Y_{N_{2\infty}} = 0.5$$

$$T_{\infty} = 23.5^{\circ}\text{C}$$

$$P = 1 \text{ Atm.}$$

$$d_f = 1.5 \text{ mm}$$

400 FRAMES/SEC.