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ABSTRACT

A study of heterogeneous detonations, which is motivated by the current belief that liquid rocket motor instability can result in a detonation or "detonation like" phenomenon, is made. First, the computer results of the jump conditions across Chapman-Jouguet detonation waves in dilute diethylcyclohexane (DECH)-gaseous oxygen are presented. The theoretical propagation velocities, Mach numbers, pressure ratios, density ratios and temperature ratios at equivalence ratio range of .1 - 3 are included. Second, three different types of experiments are described: a) detonations in a polydisperse spray, b) detonations in which the fuel is in the form of a thin film on the walls of the tube, and c) detonations in monodisperse sprays. DECH is used in all experiments because of its purity and low volatility, and the detonation is started by a transmitted shock. In type (a), for a spray with an estimated mean droplet diameter of 200 μ , and for a stoichiometric mixture, propagation velocities of 5300-5700 ft/sec and pressure ratios of ~ 30, which are

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much below the theoretical values, are observed. In type (b), steep fronted waves which are obviously maintained by the combustion process are also observed. Velocities of 3160 and 3750 ft/sec and corresponding pressure ratios of 10.3 and 17.0 are measured. For type (c), with a drop diameter of 940 μ and an equivalence ratio of . 5, velocities over 5000 ft/sec are measured. Schlieren photographs indicate a complicated structure behind the front, although the front is surprisingly planar.

NOMENCLATURE

- a = velocity of sound
- c = specific heat of liquid
- C_p = specific heat of gaseous phase at constant pressure
- h = specific enthalpy
- M = Mach number
- M' = Mach number based on the velocity of sound of oxygen

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- m = molecular weight
- n = stoichiometric fuel-oxidizer mole ratio
- P = pressure
- Q = heat release/unit mass of mixture
- R = universal gas constant
- T = temperature
- u = drop shedding velocity
- u_{0} = terminal drop velocity
- V = velocity

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NOMENCLATURE (cont.) = ratio of specific heats γ δ = ratio of specific heat of fuel to that of the oxidizer = liquid volume fraction € = fuel-oxidizer mass ratio η = density D φ = equivalence ratio Subscripts f = gaseous fuel g = gaseous oxidizer G = "all gaseous" case

- S = spray
- 1 = upstream of wave
- 2 = downstream of wave

INTRODUCTION

In recent years, large liquid propellant rocket motors have been beset with the serious problem of combustion instability. These instabilities have been experienced in all of the three possible modes; radial, longitudinal, and tangential. The latter mode is usually the most serious of the three with chamber pressure excursions of many hundreds of pounds per square inch being experienced. Considerably smaller pressure fluctuations than this are sufficient to cause mission failure in some cases. For the most part, the combustion instability problem has been treated as an acoustic problem. In recent years more attention has been given to the possibility of non-linear phenomena, i. e., shock waves coupled to the combustion process. The question then arises as to whether detonation or "detonation-like" waves play a role in rocket motor combustion instability. A significant experimental result on a relatively large (18 in. I. D.) liquid motor is provided by Clayton and Rogero⁽¹⁾. Utilizing a number of high frequency response pressure transducers located in the wall at varying axial positions, they determine that a strong, steep wave rotates around the motor at a very stable constant velocity. The pressure ratio across the wave tends to increase near the injector plate and to decrease in the direction of the nozzles. Their conclusion is that this must be a detonation or detonation-like wave.

On the premise that an understanding of the existence, properties and mechanism of heterogeneous detonations may shed light on the instability problem, the experimental work described herein is undertaken. Three types of experiments will be described—shock induced detonations in polydisperse sprays, shock induced combustion of a liquid fuel film, and detonation in monodisperse sprays. More detailed information on the first two cases may be found in Ref. 2.

The problem of heterogeneous detonation has received relatively little attention to date. Williams^(3, 4), in an analytical treatment of the problem, concludes that two phase detonations would be impossible because of the extended reaction zone arising from the leisurely droplet evaporation processes.

However, he tempers his conclusions with two observations; first, that drops below about 10 microns would produce essentially a gas phase detonation, and second, that drop shattering might circumvent the slow evaporation processes and thus support detonation. Webber⁽⁵⁾ and $\operatorname{Cramer}^{(6)}$ conducted some preliminary experiments by using a combustion driven shock tube to pass a shock wave through a diethylcyclohexane (DECH) spray in an oxygen atmosphere. These experiments resulted in a steep fronted high velocity wave which Cramer termed "detonation-like". An analytical treatment of the amplification of pressure waves in a two phase combustion system is advanced by Busch, Laderman, and Oppenheim⁽⁷⁾.

Before discussing the experiments, the idealized one-dimensional jump properties of liquid DECH-gaseous oxygen will be obtained. The general equations for spray detonations are formulated by Williams⁽³⁾ who also obtains the jump relations for the one-dimensional case in which the molecular weight and the ratio of specific heats are the same across the wave. Our aim here is to relax these conditions and to obtain an analytical expression for the comparison between the spray detonation properties and those of its "all gaseous" counterpart.

JUMP RELATIONS FOR ONE-DIMENSIONAL SPRAY DETONATIONS

Consider two systems, one of which consists of a spray of liquid fuel in a gaseous oxidizer and the other a uniform mixture of the same fuel in a gaseous form and the same gaseous oxidizer. Further, let the mass ratio η , of fuel to oxidizer be the same in both systems. The thermodynamic

TABLE I. COMPARISON BETWEEN SPRAY AND GASEOUS SYSTEMS	Gaseous Mixture		$\rho_{\rm G} = \rho_{\rm g}(1+\eta)$	$P_{G} = \frac{\rho_{G} RT}{m_{g} (1 + \eta)}$	$C_{p_{G}} = \frac{C_{p_{g}}}{\frac{1}{1+\eta}} \frac{(1+\eta\delta_{G})}{1+\eta}$	$\gamma_{G} = \frac{\gamma_{g} (1 + \eta \delta_{G})}{1 + \eta_{\gamma_{f}}^{\mathcal{B}} \delta_{G}}$	$h_{G} = \frac{C_{p_{g}}}{1+\eta} \frac{(1+\eta\delta_{G})}{1+\eta}$	$a_{G}^{2} = \frac{\gamma_{G} \operatorname{RT} \left(1 + \eta \frac{m_{g}}{m_{f}}\right)}{m_{g} \left(1 + \eta\right)}$	$m_{G} = \frac{m_{g}}{\frac{g}{1+\eta}} \frac{(1+\eta)}{m_{f}}$
	Spray	Approx. (dilute spray, $\epsilon \ll 1$)	$\rho_{\rm g}(1+\eta)$	$\frac{\rho_{\rm S}}{{\rm m}_{\rm g}^{\rm (1+\eta)}}$	+ Same	+ Same	+ Same	$\frac{\gamma_{\rm s} \rm RT}{m_{\rm g} (1+\eta)}$	$m_g (1 + \eta)$
TABLE I. COMPARISON BE		Exact	$\rho_{\mathbf{S}} = \rho_{\mathbf{g}}(1+\eta)(1-\epsilon)$	$\mathbf{p}_{\mathbf{S}} = \frac{\rho_{\mathbf{S}} RT}{m_{\mathbf{g}}^{2} (1+\eta)(1-\epsilon)}$	$C_{p_{s}} = \frac{C_{p_{g}}}{\frac{1}{1} + \eta} C_{s}$	$\gamma_{\rm S} = \frac{\gamma_{\rm g}}{1 + \eta \gamma_{\rm g}^{\rm o} \delta_{\rm s}}$	$h_{s} = \frac{C_{p_{g}}T(1+\eta\delta_{s})}{\frac{1}{1+\eta}}$	$a_{s}^{2} = \frac{\gamma_{s} RT}{m_{g} (1 + \eta) (1 - \epsilon)^{2}}$	m _S = (not defined)
	Property		Density	Equation of state	Specific heat at constant pressure	Ratio of specific heats	Enthalpy	Speed of sound	Molecular weight

properties of the spray system can be obtained from the work of Rudinger⁽⁸⁾. These properties are compared to the all gaseous system in Table I. It can be seen from this table that a dilute spray, i.e., $\epsilon \ll 1$, can effectively be treated as an ideal gas. The condition that $\epsilon \ll 1$ is well satisfied in our case since $\epsilon \cong 10^{-3}$ for the richest mixture that will be considered.

The jump relations for C-J gaseous detonations, wherein the gases before and after the wave are considered as two distinct ideal gases with their appropriate γ 's and molecular weights, are known (9) and rewritten below:

$$\frac{\left(M_{1}^{2} - \frac{\gamma_{2}}{\gamma_{1}}\right)^{2}}{M_{1}^{2}} = 2\left[\frac{Q}{C_{p1}^{2}T_{1}} - \frac{\gamma_{1}^{2} - \gamma_{2}}{\gamma_{1}(\gamma_{2}^{2} - 1)}\right]\frac{\gamma_{2}^{2} - 1}{\gamma_{1}^{2} - 1}$$
(1)

$$\frac{P_2}{P_1} = 1 + \frac{\gamma_1}{\gamma_2 + 1} \left(M_1^2 - \frac{\gamma_2}{\gamma_1} \right)$$
(2)

$$\frac{\rho_1}{\rho_2} = 1 - \frac{1}{\gamma_2 + 1} \left(M_1^2 - \frac{\gamma_2}{\gamma_1} \right) \frac{1}{M_1^2}$$
(3)

$$\frac{\mathbf{T}_{2}}{\mathbf{T}_{1}} = \frac{\mathbf{m}_{2}}{\mathbf{m}_{1}} \left[1 - \frac{\left(\mathbf{M}_{1}^{2} - \frac{\gamma_{2}}{\gamma_{1}} \right)}{\mathbf{M}_{1}^{2}(\gamma_{2} + 1)} \right] \left[1 + \frac{\gamma_{1}}{\gamma_{2} + 1} \left(\mathbf{M}_{1}^{2} - \frac{\gamma_{2}}{\gamma_{1}} \right) \right]$$
(4)

For $Q/C_{p1}T_1 \gg 1$, which emplies $M_1^2 \gg 1$, the above equations can be greatly simplified. If the following assumptions are also made:

1. $Q_G = Q_S$; i. e., the heat release/unit mass for the spray case is the same as its gaseous counterpart. In actual systems they should differ approximately by the ratio of the heat of vaporization to the

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heating value of the fuel which is usually less than 1%,

- 2. $\delta_{S} = \delta_{G}$ i.e., the specific heat of the liquid fuel is the same as its C_{p} when it is in the gaseous phase,
- 3. γ_2 and m_2 in both cases are the same, a reasonable assumption since the initial constituents are the same,

the ratios of the spray detonation parameters to those of its all gaseous counterpart can be shown to be:

$$\frac{M_{1S}^{2}}{M_{1G}^{2}} = (1 + \phi n) \frac{(\gamma_{f1}^{2} - 1) + \phi n \gamma_{f1}^{2} (\gamma_{g1}^{2} - 1)}{(\gamma_{f1}^{2} - 1) + \phi n (\gamma_{g1}^{2} - 1)}$$
(5)

$$\frac{\mathbf{P}_{2}^{\prime \mathbf{P}_{1}}}{\mathbf{P}_{2}^{\prime \mathbf{P}_{1}}} = 1 + \phi \mathbf{n}$$
(6)

$$\frac{\left(\frac{\rho_{2}}{\rho_{1}}\right)_{S}}{\left(\frac{\rho_{2}}{\rho_{1}}\right)_{G}} = \frac{\left(\frac{T_{2}}{T_{1}}\right)_{S}}{\left(\frac{T_{2}}{T_{1}}\right)_{G}} = \frac{V_{1S}}{V_{1G}} = 1$$
(7)

It should be mentioned that despite the somewhat more generalized case used here, the predictions of Eqs. (5, 6, 7) are similar to those of Williams analysis.

The C-J detonation properties of DECH (hypothetically assumed to exist in the gaseous phase) with oxygen initially at P = 1 atm and $T = 298.15^{\circ}K$ are obtained from a computer program of Zeleznik and Gordon^{(10)*}. The heat of

The authors are grateful to Drs. Zeleznik and Gordon for making the computer program available to them.

formation for DECH and its C_p used are - 73(Kcal/mole) and .44(cal/gm⁰K) respectively, evaluated from Refs. (11, 12). The results are plotted in Fig. 1 wherein the properties of spray counterpart, calculated according to Eqs. 5, 6, 7 are also shown. It can be seen that like most hydrocarbons, DECH exhibits peak pressure ratio and detonation Mach number at richer than stoichiometric mixtures.

EXPERIMENTAL STUDIES

a. Detonations in Polydisperse Sprays

Experiments to initiate detonations in polydisperse sprays of fuel in gaseous oxygen were carried out in a vertical shock tube arrangement similar to that used by Webber $^{(5)}$ and Cramer $^{(6)}$ but with some important differences. The driver and the driven sections are 2 in. I.D. stainless steel tubes of 1/4 in. wall thickness with the driver mounted above the driven section. The length of the driven section is 8 ft whereas that of the driver is 4 ft. The four injectors (spaced $1 \frac{1}{2}$ ft apart) followed closely the design of Cramer; the liquid fuel was forced downward through a 0.022 in. diameter hole by a piston which was actuated by nitrogen at 150 psi. The first injector was located 1.5 ft away from the diaphragm thus allowing a buffer zone between the driver gas and the spray. DECH $(C_{10}H_{20})$ was chosen as fuel because of its low vapor pressure (less than 2 mm Hg at room temperature), because it is a pure compound with properties comparable to RP-1, and because it affords a direct comparison of our experiments with those of Cramer. The shock tube was fired 275 millisec after actuation of the injectors at which time all the liquid had been ejected and the spray cone

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extended at least three feet from each injector. While a large portion of the spray is contained in a 2 in. diameter, photographs of the unconfined spray indicated that the walls of the combus tion tube would also be heavily wetted. Cramer estimated that the mean drop diameter of the spray was 200μ .

The instrumentation consisted of pressure transducers to measure the velocity and pressure of the waves in the combustion tube, and ionization probes to sense the detonation wave in the driver. Two Kistler 603 and two Kistler 601A pressure transducers were flush-mounted in the combustion tube. The output from the transducers was amplified by Kistler 566 electrostatic charge amplifiers and then displayed on a Tektronix 555 dual beam oscilloscope. Before using the pressure transducers their response to a hydrogen-oxygen detonation wave in a $1/2 \times 3/8$ in. tube was checked and found to agree with the manufacturer's static calibration. Their rise time was $1-3 \mu$ sec. Despite their oscillatory response, it was decided not to damp the signal electronically or mechanically in order to preserve the short rise time of the pickups. Ionization probes in the driver were used to check the performance of the driver and to trigger the sweep on the oscilloscope for the pressure recordings.

The driver of the shock tube was operated in the detonation mode with hydrogen and oxygen. For a one atmosphere $2H_2 + O_2$ detonation into one atmosphere of N₂, theory⁽¹³⁾ predicts a transmitted shock at Mach number of 4.5. This shock is expected to decay rapidly and in fact measurements indicate that the shock Mach number at 1.5 ft from the diaphragm is about 4.1. The theoretical velocity of the interface between driver and test section

gases is 3400 ft/sec, so that a uniform flow behind the shock exists for 120 microseconds at a distance of 1.5 ft where the fuel is first available. The one-dimensional shock relations for a Mach 4.1 shock predict a temperature of 2189⁰R and a pressure of 293 psia.

When nitrogen is used in the test section the initial shock Mach number is reduced rather rapidly as it progresses down the test section. When DECH spray (2.3 ml) is added to N_2 in the test section further reduction in the Mach number of the wave as it progresses down the tube, indicative of the retarding effect of the drops, is observed. On the other hand when 2.3 mf (~ 1.2 stoichiometric) of the fuel are sprayed into the test section containing O_2 the shock transmitted by the driver rapidly accelerates to a velocity of 5300 to 5700 ft/sec which is much lower than the theoretical velocity of 7850 ft/sec shown in Fig. 1. The pressure jump across the wave was also increased to approximately 30 to 1. The results from several runs, are plotted in Fig. 2 which includes the results for N_2 also. (The Mach number plotted in this figure is based on the velocity of sound of oxygen.) Now, if the average experimental velocity of propagation is used together with the appropriate spray sound velocity one finds from Eq. 2 that the pressure ratio should be about 23 for $\gamma_2 = 1.13$ which is obtained from the computer calculations. On the other hand the measured pressure ratio corresponds closely to a Mach 5 shock in oxygen. One possible explanation for the difference is that the shock front is followed by a reaction zone of considerable extent and the detonation follows essentially the ZND model.

Since the detonation velocity is considerably lower than the expected 11

gaseous detonation velocity for a stoichiometric mixture it is possible that only a fraction of the fuel is entering into the initial reaction. Experiments in which a larger quantity of fuel was used (up to an equivalence ratio of 7) showed no significant change in the results. In this respect our results appear to be different from those of Cramer⁽⁵⁾. While he finds that an equivalence ratio of 3 or more is necessary to induce detonation, consistent initiation was possible in our experiments at nearly stoichiometric mixtures. One explanation could be the technique of initiation as afforded by the two types of driver sections.

In our experiments initiation was also possible when the O_2 -H₂ mixture in the driver was at 1/2 atmosphere which resulted in a Mach 3.7 shock at the location of the first spray injector. Little change in the strength of the developed wave was observed.

b. Shock Induced Combustion of a Liquid Fuel Film

As indicated, wall wetting was unavoidable when the injectors were used. In some experiments it was observed that the fuel clinging to the walls in the absence of any spray was sufficient to support a steep fronted wave. For this reason it was decided to investigate the case where fuel existed solely in the form of a liquid film on the walls (no spray) and the case where there was a spray with no wall wetting.

For the former case, the driven section was replaced by a 1.64 in. square tube terminated by a 1 ft long section for optical observation. After filling the test section with oxygen, a thin layer of DECH was applied to the

walls up to within 1.5 ft of the diaphragm by means of a felt swab. In some of the tests only one wall was coated-the solid wall containing the pressure transducers; in other tests the two opposite solid walls were coated. For the case when DECH was swabbed on two walls the measured velocity was 3750 ft/sec and the pressure ratio was 17 to 1. When DECH was swabbed on one wall only the measured velocity was 3160 ft/sec and the pressure ratio was 10.5 to 1. In both cases the strength of the propagated wave is lower than the initial strength (M' = 4.1) of the transmitted shock. However, at the end of the test section the propagated wave is much stronger than the transmitted shock into nitrogen at a corresponding location, which indicates that chemical reaction is driving the shock front. Framing camera photographs of these tests show that the reaction originates at the wall and spreads inward from the wall until about 250 μ sec after passage of the initial front at which time the entire width of the channel is filled with luminosity. Spark schlieren photographs of the case where DECH is applied to two walls with oxygen in the test section are shown in Fig. 3. In one run the shock front is slightly convex, in another slightly concave. The dense reaction zone extending from the wall starts closely behind the shock. The turbulence in the center of the channel directly behind the shock front may be due to the complexity of the shock front rather than chemical reaction. Spark schlieren photographs of the case where DECH is applied to one wall are shown in Fig. 4. Here the reaction zone trails the leading shock front by 2/3 of a channel width or 30 μ sec. Note the dark zone along the wall which is interpreted to be the primary combustion zone.

To our knowledge this type of phenomena was first reported by Loison⁽¹⁴⁾ in 1952 and additional experiments have been recently conducted by Gordeev et al. $^{(15)}$ In Loison's experiment a steady state detonation of a thin film of lubricating oil propagating at 3600 ft/sec was observed in a 300 ft long tube. Combustion of the film was initiated by a pulse of hot gas of acoustic strength. In the experiments of Gordeev ignition of a thin film of libricating oil or grease on the walls of a 5 ft long tube was accomplished by a methaneoxygen detonation, an exploding wire, or a charge of lead azide. For all of these ignition sources Gordeev reports that an accelerating combustion develops and turns into a detonation with velocities as high as 5100 ft/sec.

c. Detonations in Monodisperse Sprays

Because of the apparent ease with which film combustion can occur, a study of sprays which avoid wall wetting is in order. Also, to study the effect of drop size, monodisperse sprays would be ideal. A technique which makes use of a number of vibrating capillary liquid jets $(2, 1^{6})$ can be easily used to produce monodisperse sprays with drop size above a level dictated by the physical properties of the capillary jets and the aerodynamics of the drops formed therefrom. Figure 5 shows the variation of the initial minimum possible velocity of liquid jets (and hence the velocity of the drops in oxygen are shown. It can be seen that for DECH the cross over point is $380 \,\mu$. For sizes greater than this, the drops will be accelerating and for lower sizes they will be decelerating. Since the distance between the centers of the drops is about 2.5 drop-diameters initially, it becomes apparent that

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collision and coalescence of the drops can take place when the drops are decelerating. In practice, it is found that $750 \,\mu \,drops$ —as can be produced by .016 in. diameter needles—or larger are necessary to avoid coalescence. Figure 6 shows nine streams of $750 \,\mu \,drops$ in a 3 x 3 array at the shedding location and at 2 ft below the drop generator. Although all streams are not in the same plane, the drops appear in good focus because collimated light was used. Their uniformity as well as the accelerating effect are apparent.

Experiments in the square tube mentioned in the previous section are conducted with monodisperse sprays of 940 μ drops. The configuration is shown schematically in Fig. 7. Because of the drop generator it was necessary to introduce the transmitted shock at an angle from a driver section with an area much smaller than that of the detonation tube proper. Since the velocity of the drops increases as they fall down the tube variable mixture ratio is expected along the tube. The variation is from $\phi = 2$ near the generator to a constant $\phi = .5$ at distances beyond 4.5 ft from generator. Velocities and pressure ratios measured at the lower portion of the tube for over 30 runs averaged 4550 ft/sec and 25 respectively. The spread was 3940-5090 ft/sec for the velocity and 16-32 for the pressure ratio. Again, the average velocity is 30% lower than the theoretical and the pressure ratio can be explained by a ZND model for the detonation. The experimental velocities can perhaps be explained by the fact that immediately behind the shock the mixture is leaner than that in the quiscent case because of the inertia of the fuel drops. Schlieren photographs typified by those in Fig. 8 show in some cases shocks around individual drops behind the leading shock.

Other shock patterns are also apparent, but in most cases the front appears surprisingly planar. Figure 9 shows a combined shadow and direct light photograph. It reveals that combustion starts approximately .6" (~ 11 μ sec) behind the front. It is interesting to note that this time delay corresponds very closely to drop break-up times based on extrapolation of the data of Wolfe and Anderson⁽¹⁷⁾. On this basis, it appears that the mechanism of heterogeneous detonation is controlled by the mechanical break-up of the drops. Further experiments, are being conducted to gain a complete understanding of the phenomenon.

CONCLUSIONS

It is apparent that steady-state high pressure-high velocity waves supported by combustion do exist in liquid sprays and that their gasdynamic properties can be roughly predicted by appropriate one-dimensional jump relations. The relative ease with which combustion and subsequent detonation of monodisperse sprays with 940 μ drops occur indicates that high pressure-high velocity waves may be possible over a wide range of drop sizes. Limited data indicate that in such sprays break-up of the drops is the controlling mechanism. High pressure-high velocity waves are also found to be supported by combus tion of a thin liquid film on the walls of a tube. Further studies are necessary for the understanding of heterogeneous detonations in sprays and liquid films.

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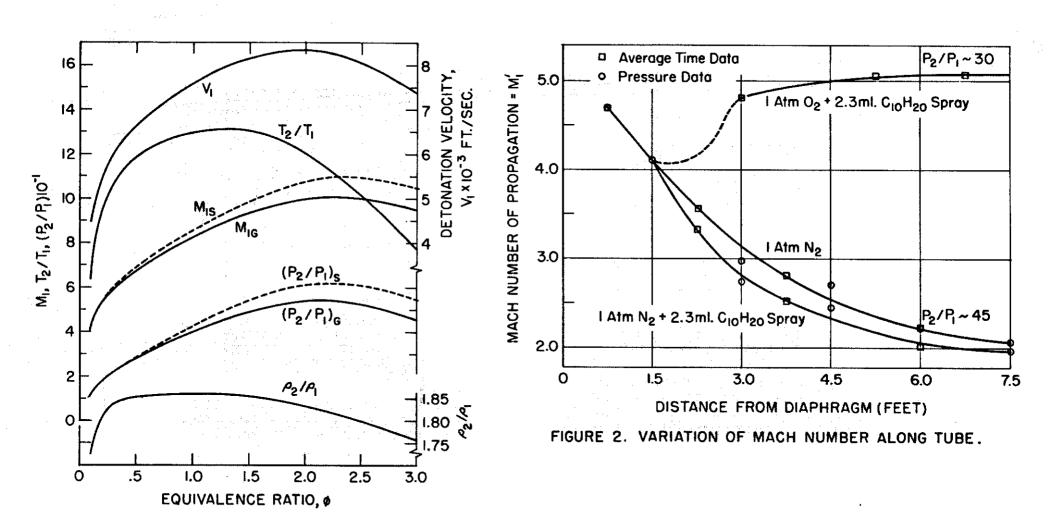


FIGURE I. PROPERTIES OF DECH-O2 DETONATIONS.

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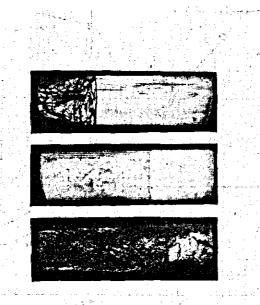
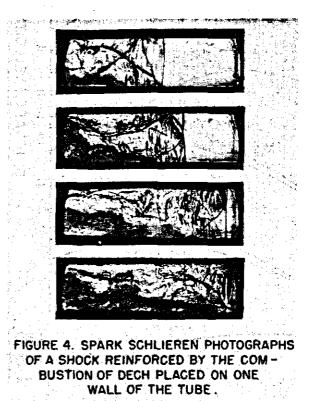
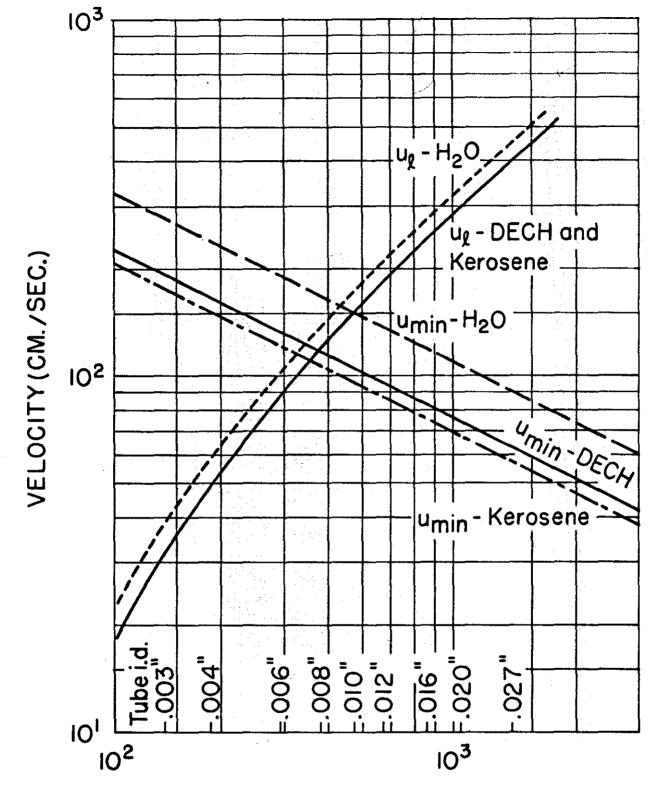


FIGURE 3. SPARK SCHLIEREN PHOTOGRAPHS OF A SHOCK REINFORCED BY THE COM-BUSTION OF DECH PLACED ON TWO WALLS OF THE TUBE.

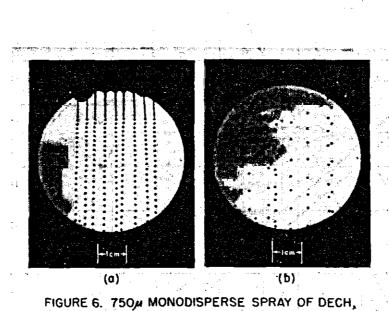


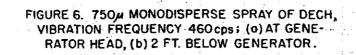
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DROP DIAMETER (μ)

FIGURE 5. TERMINAL AND MINIMUM VELOCI-TIES OF DROPS IN O₂ ATMOSPHERE FOR WATER, DECH AND KEROSENE.





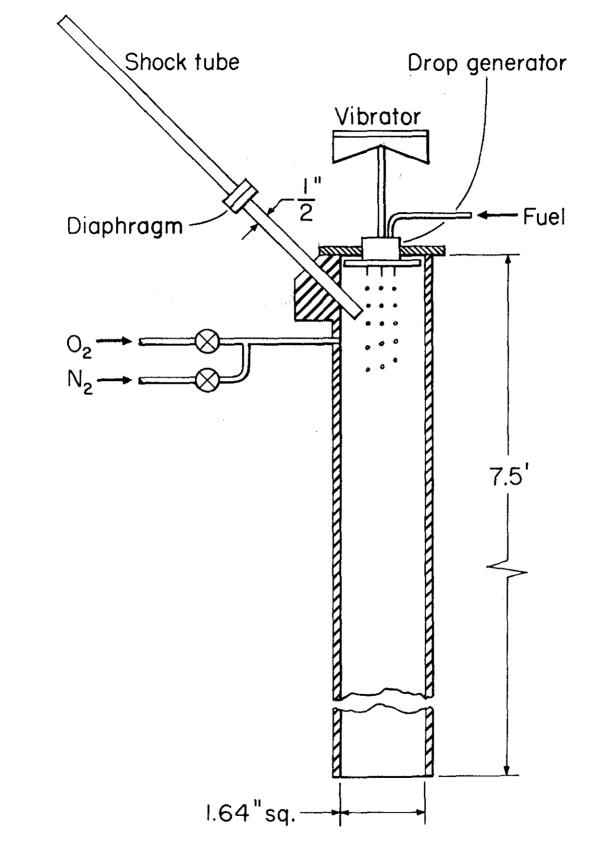


FIGURE 7. SCHEMATIC OF THE COMBUS - TION TUBE FOR MONODISPERSE SPRAY.



FIGURE 8. TYPICAL SCHLIEREN PHOTO-GRAPHS IN MONODISPERSE SPRAY DETONATIONS.



FIGURE 9. COMBINED SHADOW AND DI-RECT LIGHT PHOTOGRAPH OF DETO-NATION IN MONODISPERSE SPRAY.