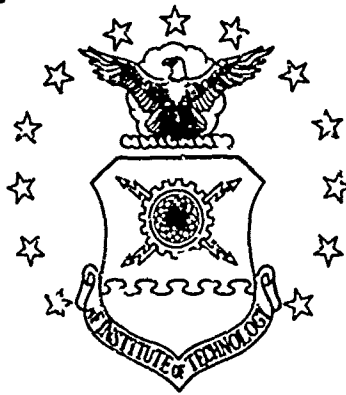
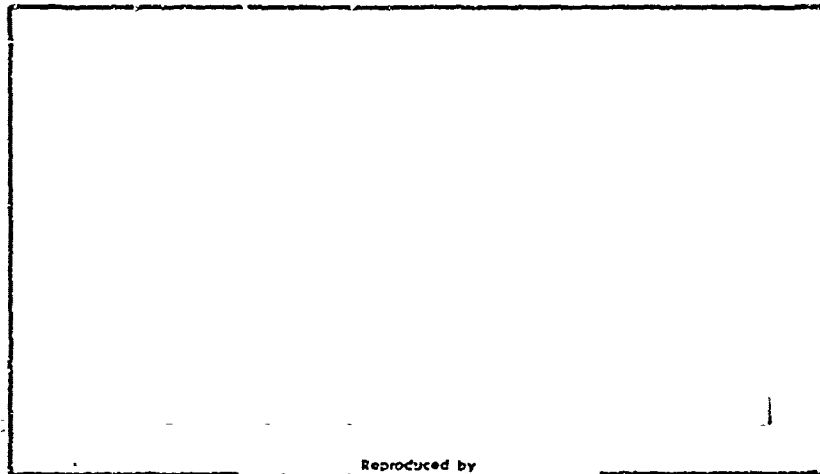


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A STUDY OF DETONATION PROPAGATION
IN NON-STOICHIOMETRIC HYDROGEN-OXYGEN-
DILUENT MIXTURES

THESIS

GAM/ME/72-4

William F. Balmano
Captain USAF

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Preface

This investigation is concerned with the initiation and propagation of detonation waves in non-stoichiometric hydrogen-oxygen-diluent mixtures. The results, which are all experimental in nature, are compared with some existing theories.

I wish to thank Maj. R. M. Jensen, of the Air Force Institute of Technology, for his valuable advice and encouragement during the course of this study, and Maj. R. G. Surette, of the Aero Propulsion Laboratory, for his assistance and continuing interest in the project. I would also like to thank Mr. John Parks and Mr. Richard Brown, both of the Air Force Institute of Technology, for their skillful and dedicated assistance in operating the equipment, and Dr. Tiernan and his staff at the Aerospace Research Laboratories for their assistance.

Finally, I wish to express my deepest gratitude to my wife, Susan, for her patience, understanding, and unselfish assistance during the many months required to complete this study.

W. F. BALMANNO

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Abstract

An experimental investigation of the initiation and propagation of shock induced detonation waves in non-stoichiometric hydrogen-oxygen-diluent mixtures was conducted. Equivalence ratios of 0.50, 0.75, 1.25, and 1.50 were used with 0, 50 percent, and 90 percent diluent. Diluents utilized consisted of either argon or helium. The data appeared to verify the finite reaction zone theories of Brinkley and Richardson as well as to give some information concerning critical Mach numbers and detonation onset distances. All test gases were initially at rest at 10 in. Hg. absolute pressure and approximately 293 degrees Kelvin. Compressed helium was used as the driver for the initiating shock.

A STUDY OF DETONATION PROPAGATION
IN NON-STOICHIOMETRIC HYDROGEN-
OXYGEN-DILUENT MIXTURES

I. Introduction

Background

The earliest reports of detonations in the literature can be traced to 1881. Twenty-five years later, reasonably accurate theories were established. Detonations have continued to be a subject of interest and excellent discussions of this phenomena as well as combustion in general can be found in Jost (Ref 4) or in Lewis (Ref 5).

The supersonic ramjet has, in recent years, become increasingly more feasible. However, supersonic combustion is still not adequately understood. Further work is needed to relate experimental results with kinetic calculations and to incorporate the results into possible hypersonic air-breathing combustion systems. Currently there are two types of supersonic combustion possible: (1) premixed and (2) diffusional burning. Premixed combustion occurs when an already mixed fuel and oxidizer are rapidly raised to combustion temperatures, and diffusional combustion occurs when the fuel is injected into a preheated oxidizer. Detonation is a special form of supersonic premixed combustion in which a strong shock is used to rapidly raise the temperature (and pressure)

of the combustibles (Ref 7:2778).

Many investigators have attempted to use shock induced detonations to study supersonic combustion and the associated chemical kinetics. However, detonation theory and associated models are still incomplete. Recently Maj. R. G. Surette, AFAPL, undertook a study of detonation. His study was to investigate theoretical models of detonation and, using computer techniques, to obtain a model which would adequately describe the initiation, onset, and stabilization of shock initiated detonation waves. Adequate data was not available in the literature so he assembled a shock initiated detonation tube and obtained data with stoichiometric hydrogen-oxygen-argon mixtures.* A good discussion of the basics of such models can be found in an article by Skinner, Mueller, Grimm and Scheller which describes "The steps involved in the initiation of detonation by incident shock waves . . ." (Ref 8:436). Such a model, predicting pressures, velocities, temperatures, compositions as well as other parameters would be of questionable value if it could not be verified with experimental data.

Purpose

Belles states that, ". . . if it is found that a well-defined shock strength is required to ignite a given mixture, it is reasonable that the detonability of the mixture should

*Based on personal conversations with Maj. R. G. Surette.

be related to that shock strength" (Ref 1:216). The critical Mach number for the purpose of this report is defined as that "well defined shock strength" required to ignite the mixture. Belles further stated in the same reference that ". . . it may also be speculated that the detonation limits may occur at concentrations such that these two Mach numbers (the critical Mach number and the Chapman-Jouguet Mach number) are equal." He concludes, in part, after investigation and experimentation, that ". . . The measured shock ignition temperatures do not agree at all well with those predicted from explosion limit studies in glass bulbs" and that "Detonation limits bear no evident relation to shock ignition data" (Ref 1:220).

A weak shock, at or above the critical Mach number, initiated into a combustible mixture will, after travelling for some distance, suddenly accelerate to detonation velocities. This transformation starts slowly, but quickly becomes very rapid and well defined. The point at which detonation velocities suddenly occur is called the point of onset of detonation by Edse (Ref 3:2).

It seemed that the critical Mach numbers, the detonation onset distance, and the detonability limits might all be related and with this in mind the investigation was undertaken. The purpose of this investigation was, therefore, to obtain data on such parameters as critical Mach numbers, detonation onset distances, detonability limits and the stability of

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detonation waves relative to Chapman-Jouguet predictions for non-stoichiometric hydrogen-oxygen-diluent mixtures.

Scope

Diluent gases have two desirable effects on detonations. First, they absorb energy thus reducing temperatures and reaction rates and, secondly, they alter the speed of sound thus making a wider range of initial Mach numbers available for shock induction. The diluents, helium and argon, were chosen because they were both monatomic, but varied greatly in molecular weights.

The scope of this investigation was to obtain time vs. distance and velocity vs. distance data for shock initiated detonations of hydrogen-oxygen mixes with helium and argon used as diluents. Equivalence ratios of 0.5, 0.75, 1.25, and 1.50 were used. The diluent gases (helium and argon) were each used at zero %, 50%, and 90% concentrations. The diaphragms and gas mixture ratios used are listed in Tables I and II respectively.

II. Equipment and Instrumentation

The apparatus used in this investigation consisted of a helium driven shock tube with the necessary controls and data acquisition system components. Each basic component will be discussed individually. An overall schematic diagram of the apparatus is shown in Figure 1 and photographs are shown in Figures 2 through 5.

The Shock Tube

The shock tube consisted of an eight-foot driver section and six separate two-foot sections of type 304 stainless steel tubing. The tubing has a 1.5-inch inside diameter with a 0.5-inch wall thickness. The six individual test sections were flanged and bolted together to make the overall test section of a nominal twelve-foot length. Care was taken to provide precise alignment of the sections at the flange joints during construction of the tube so that flow disturbances would be minimized. All flange joints were fitted with O-ring seals since all instrumentation and plumbing connections had to be sealed and leak checked prior to commencing a test. This procedure was mandatory because the tube had to first be evacuated and then filled with the desired gas at less than atmospheric pressure. A CENCO Hypervac 25 vacuum pump was used for evacuation.

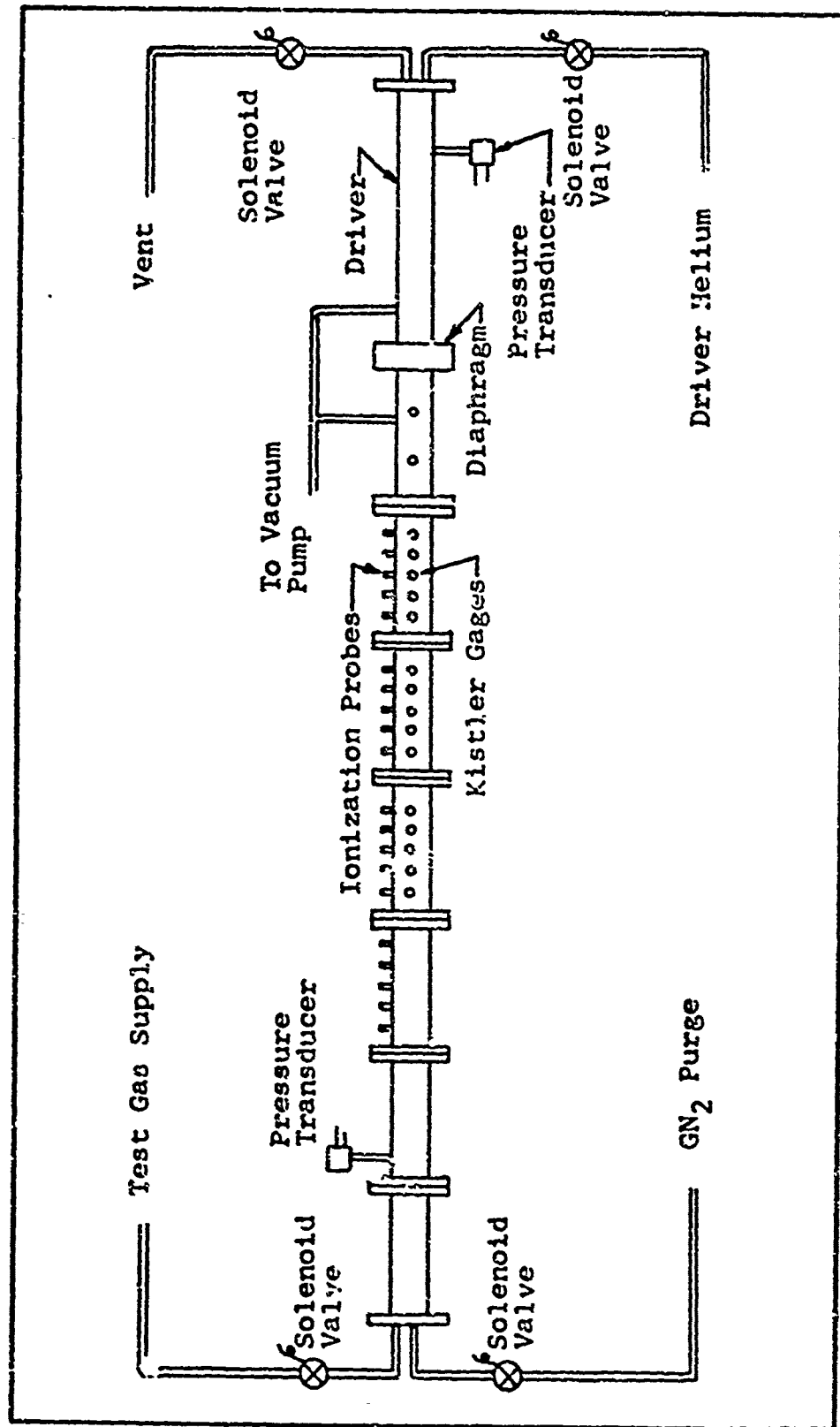


Figure 1. Schematic of Test Tube and Instrumentation.

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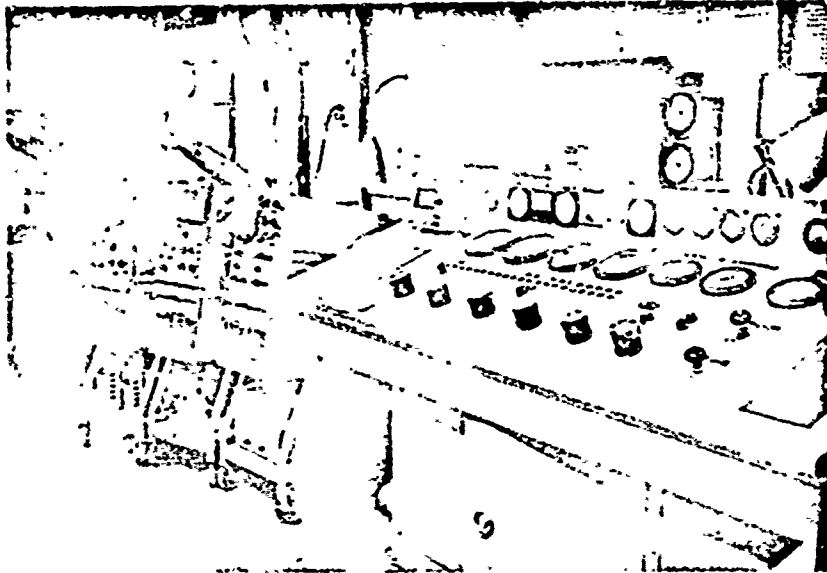


Figure 2. Oscilloscopes and controls.

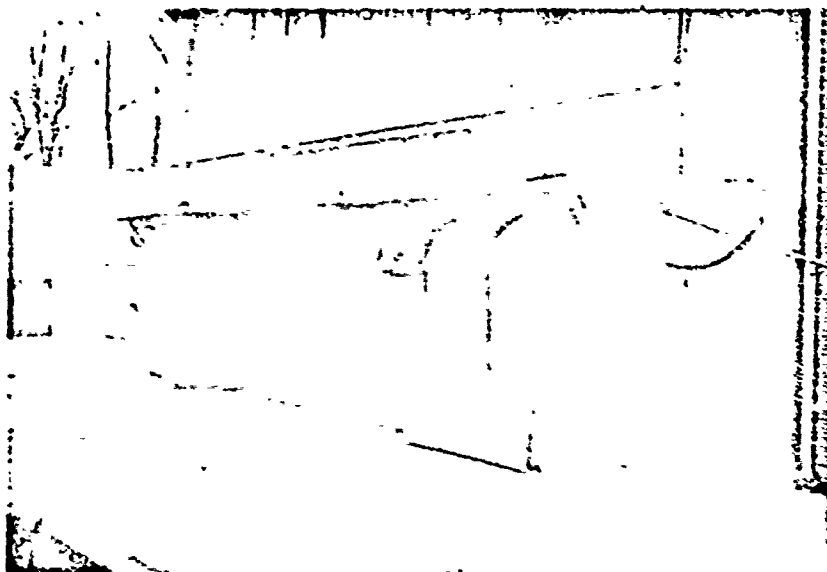


Figure 3. Mixing tanks and oxygen supply bottle.

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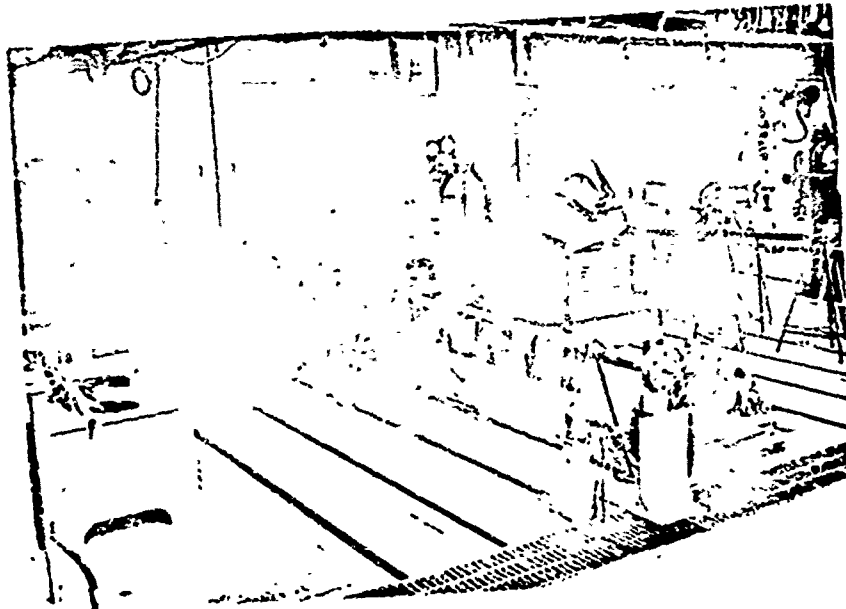


Figure 4. The shock tube.



Figure 5. Diaphragms before and after rupture
next to the diaphragm retainer of the shock tube.

The Diaphragm

The initial shock was created by bursting a scored aluminum diaphragm. Careful control of the score depth was necessary to obtain repeatable results since the diaphragm was not mechanically broken but burst when it could no longer withstand the differential pressure. Various thicknesses of aluminum, each scored to several different depths were used in an effort to obtain a reasonable range of Mach numbers. Table I lists the various diaphragms used and Figure 5 shows a typical diaphragm before and after rupture.

Table I
Diaphragms Used in This Investigation

Thickness (inches)	Score Depth (inches)
0.032	0.011
0.032	0.013
0.032	0.015
0.032	0.017
0.020	0.003
0.020	0.005
0.020	0.007
0.020	0.009

Gas Supplies

The shock tube driver was pressurized from a standard cylinder of industrial helium regulated to approximately 1200 psi. Pressures above 1000 psi were necessary to obtain repeatable diaphragm burst characteristics.

The hydrogen-oxygen-diluent gases were mixed in two tanks located outside the test cell. These tanks were

connected to the oxygen supply which consisted of a number of standard industrial oxygen cylinders, and to the hydrogen supply which was a high-pressure gas trailer manifolded to the system. Diluent cylinders were connected to the tanks when desired.

Nitrogen gas, also supplied from a high-pressure gas trailer, was used to purge the shock tube after each test to insure that residual reactive gas mixtures were not introduced into the vacuum pumps. The nitrogen purge also helped to remove moisture from the tube between experiments.

All gas flows were controlled remotely with solenoid valves.

Ionization Probes

The ionization probes were utilized in the test section of the tube to detect the passage of the flame front of the detonation wave. There were twenty probes placed along the test section at approximately four inch intervals except at the flanges. The exact location of all probes was known to the nearest thousandth inch. These probes are described in detail by McKenna (Ref 6:22-23). The probes were in a circuit with capacitors and resistors as shown in Figure 6. The capacitors were charged with a Hewlett Packard Model 712B Power Supply and the resulting voltage applied across the probe electrode and the tube wall. The passage of the detonation wave front was detected by the discharge of the capacitor which was displayed on one of the raster oscilloscopes

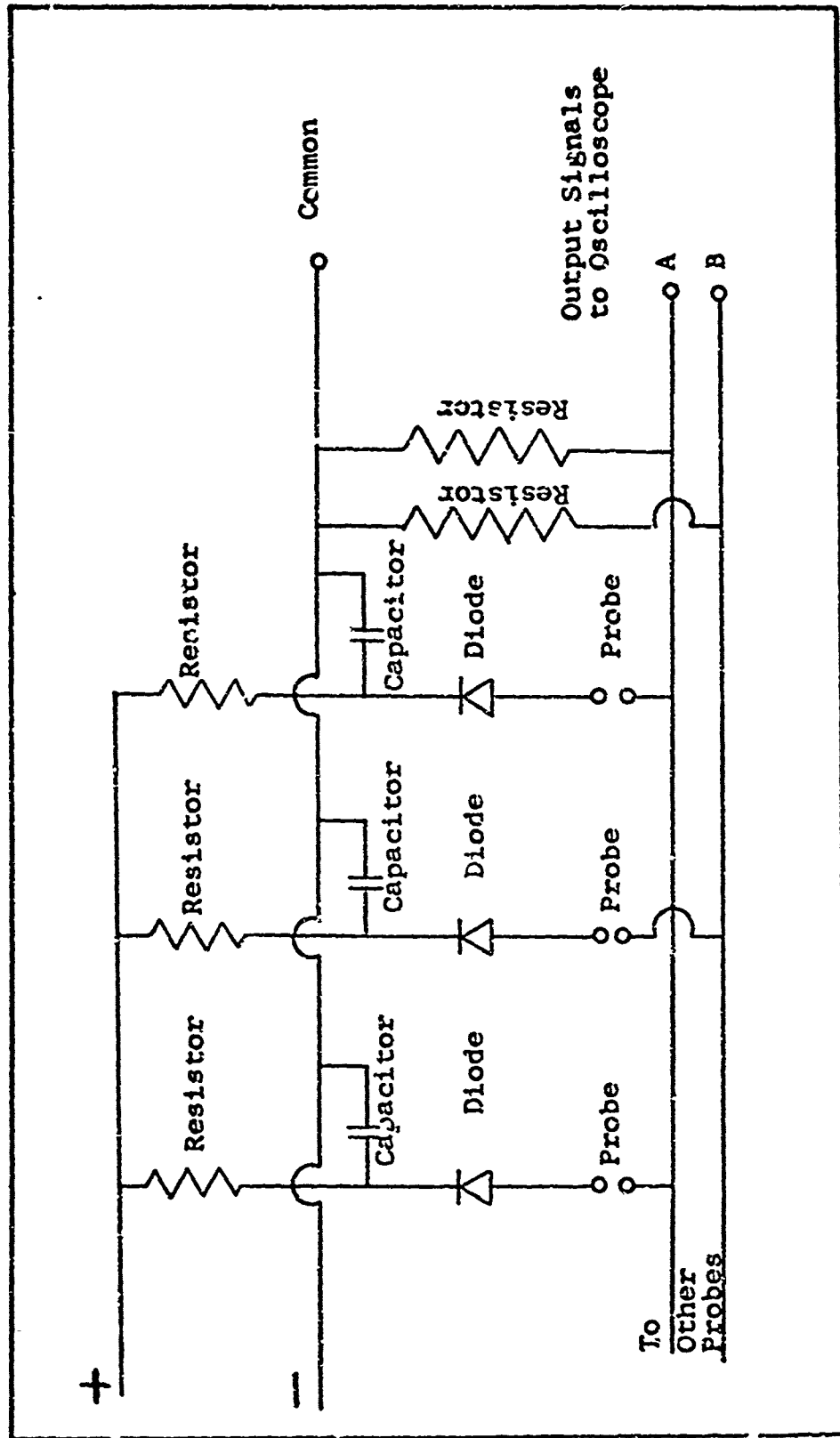


Figure 6. Schematic of Ionization Probe Circuit.

in a distinct positive-negative pattern which made it possible to determine the order in which the signals were received.

Kistler Pressure Transducers

There were seventeen Kistler miniature pressure transducers mounted in the test section of the tube. Two of the transducers were placed 7.756 inches apart in the middle of the first test section. The purpose of these transducers was to trigger the oscilloscopes and to determine the initial velocity of the shock. The remaining fifteen transducers were placed adjacent to each of the first fifteen ionization gages. The Kistler transducers were of two types: Model 601L and Model 603A. These two models were essentially alike except in response amplitude, which was of no concern to these experiments, and were used simultaneously solely because of their availability. The transducer signals were amplified with Kistler Model 566 Charge Amplifiers and displayed on the second raster oscilloscope in a manner similar to that described for the ionization gages.

Oscilloscopes

Two raster type Tektronix oscilloscopes and one dual-beam oscilloscope provided the means for recording and measuring the detonation velocity. The raster feature was necessary to extend the sweep time and still retain an acceptable sweep rate. A sweep rate of 0.1 milliseconds per centimeter

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was used with a raster rate of 50 microseconds per 10 centimeters. This enabled the time of passing of the shock to be determined to within \pm 0.25 microseconds. A type 181, Tektronix, Time Mark Generator was used to obtain accurate calibration of the sweep rate. The raster oscilloscopes were calibrated before and after each day's testing and the dual-beam oscilloscope was calibrated during each test. Calibration was accomplished by photographing 100 microsecond time marks on the oscilloscopes.

The ionization trace was displayed on a Tektronix Type 545B oscilloscope, with raster and Type D plug-in unit. The Kistler trace was displayed on a Tektronix Type 546 oscilloscope, with raster and Type G plug-in unit. The triggering oscilloscope was a Tektronix Type 551 Dual-beam with a Type CA plug-in unit. The plug-in units were the appropriate type necessary to display the positive-negative or the dual-beam inputs.

The oscilloscope traces were photographed using a polaroid camera and a Tektronix Type C-12 lens assembly. Typical oscilloscope trace photographs are shown in Figure 7.

Other Instrumentation

A Pace Transducer Indicator, Model CD25, was used to measure pressures in the test section of the shock tube and the mixing tanks. The transducers and indicators were calibrated against the atmosphere and a Welsh Scientific Company, Duo Seal Vacuum Pump prior to each day's use. Test with a

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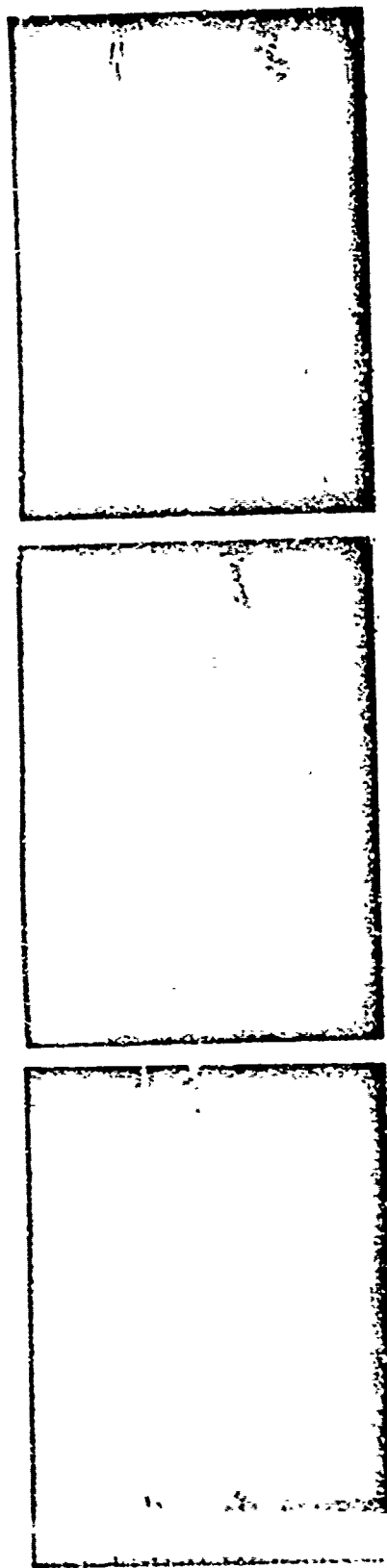


Figure 7. Typical Oscilloscope Trace Photographs. Dual-beam (left), Kistler (middle), and Ionization (right) Traces from one of the tests conducted.

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Mercury manometer indicated this pump provided a vacuum pressure of 0.36 inches Hg to the reference side of the transducers.

A Consolidated Electrodynamics Recording Oscillograph was used to record the driver pressures which verified diaphragm rupture so that pressurization could be terminated and venting initiated.

III. Experimental Techniques and Procedures

The experimental techniques and procedures used in this research are discussed below in an effort to clarify what was done, why it was done and most importantly, what effects these techniques had on the results. The two main procedures were mixing the test gases and operating the detonation tube.

The Test Gases

Mixing of the test gases to desired specifications was accomplished by utilizing the method of partial pressures. Care was taken to insure slow flow rates so that temperature effects would be kept to a minimum. The mixed gases, in all cases, were left in the tanks for several days before use to insure homogeneity. Each equivalence ratio gas was to be used with diluents in various amounts as can be seen from Table II. Therefore, the hydrogen and oxygen were mixed to the desired equivalence ratio, sampled and partially used. The diluents were then added to the remaining gas. This process was followed for each equivalence ratio specified. All gases were allowed to flow through supply lines for several minutes before sampling and subsequent use in the detonation tube. All test gas samples were analyzed on a mass spectrometer by the Aerospace Research Laboratories prior to use to insure validity of output data.

Table II
Gas Mixtures Tested

-
- (1) $H_2 + O_2$
 - (2) $H_2 + O_2 + 2Ar$
 - (3) $H_2 + O_2 + 2He$
 - (4) $H_2 + O_2 + 18Ar$
 - (5) $H_2 + O_2 + 18He$
 - (6) $1.5 H_2 + O_2$
 - (7) $1.5 H_2 + O_2 + 2.5Ar$
 - (8) $1.5 H_2 + O_2 + 2.5He$
 - (9) $1.5 H_2 + O_2 + 22.5Ar$
 - (10) $1.5 H_2 + O_2 + 22.5He$
 - (11) $2.5 H_2 + O_2$
 - (12) $2.5 H_2 + O_2 + 3.5Ar$
 - (13) $2.5 H_2 + O_2 + 3.5He$
 - (14) $2.5 H_2 + O_2 + 31.5Ar$
 - (15) $2.5 H_2 + O_2 + 31.5He$
 - (16) $3H_2 + O_2$
 - (17) $3H_2 + O_2 + 4Ar$
 - (18) $3H_2 + O_2 + 4He$
 - (19) $3H_2 + O_2 + 36Ar$
 - (20) $3H_2 + O_2 + 36He$
-

Operation of the Shock Tube

Operation of the shock tube required that all instrumentation and equipment be properly prepared in a specific sequence. Checklist procedures were prepared to insure uniform, proper operation of the equipment.

Data Reduction and Handling

The data obtained from each test consisted of three polaroid photographs of oscilloscope traces. These traces were accurately measured with a comparator micrometer and further data reduction was accomplished on a digital computer. The computer program, which was written by Surette, converted the basic measurements into time and distance data to which a polynomial curve was fitted using a least squares fit technique. The derivative of this curve yields the velocity. The time vs. distance and velocity vs. distance data points were plotted using the on-line plotter. Each test required approximately one-half hour to conduct and one hour to measure the photographs and put all data into the computer which then required less than one second of computation time to reduce the data. The program was converted to machine language to eliminate compilation time and reduce core memory requirements resulting in extremely rapid turnaround times and more economical use of the computer. The plots and computer printouts represent the meaningful data from the tests.

Calibrations

Considerable increase in accuracy of oscilloscope sweep

rates was obtained by photographing time mark generator pulses displayed on each oscilloscope. The sweep rates used in this research are considered an "uncalibrated rate" by the manufacturer and variations of up to twenty percent were observed between scopes. The sweep rate of each individual scope as calibrated by the time mark generator was observed to vary less than one percent over the entire duration of the research. These calibrations were performed before and after each day's testing on both raster oscilloscopes and during each test throughout the day on the dual-beam oscilloscope.

IV. Discussion of Results

The immediate results of the 140 tests conducted were 72 graphs and computer printouts. The remaining 68 tests, as expected, either failed to detonate, or detonated too late to interpret for further processing. Some typical graphs are shown in Figures 8 through 12. Examinations of the data reveal several significant facts. The polynomial curve fits were quite good, giving errors of less than one percent in almost all cases. Some of the experimental data indicates that the flame front (Detonation) sometimes arrives at a station slightly before the shock front (Wave). This was, of course, not actually the case but reflects the fact that these two waves are quite closely coupled and that there are differences in the instrumentation response times of the Kistler transducers, ionization probes, and oscilloscopes. The apparent cross-over was more prominent with gas mixes containing no diluent, where the induction times and, therefore, coupling distances were smallest. Indeed, many of the cases with diluent gases showed no cross-over at all. No attempt was made to measure or further evaluate the differences in response times since it was felt this had little effect on the shapes of the curves or on the resulting velocities.

Wave Stability and Nature

Jouquet pointed out that rarefaction waves, which must

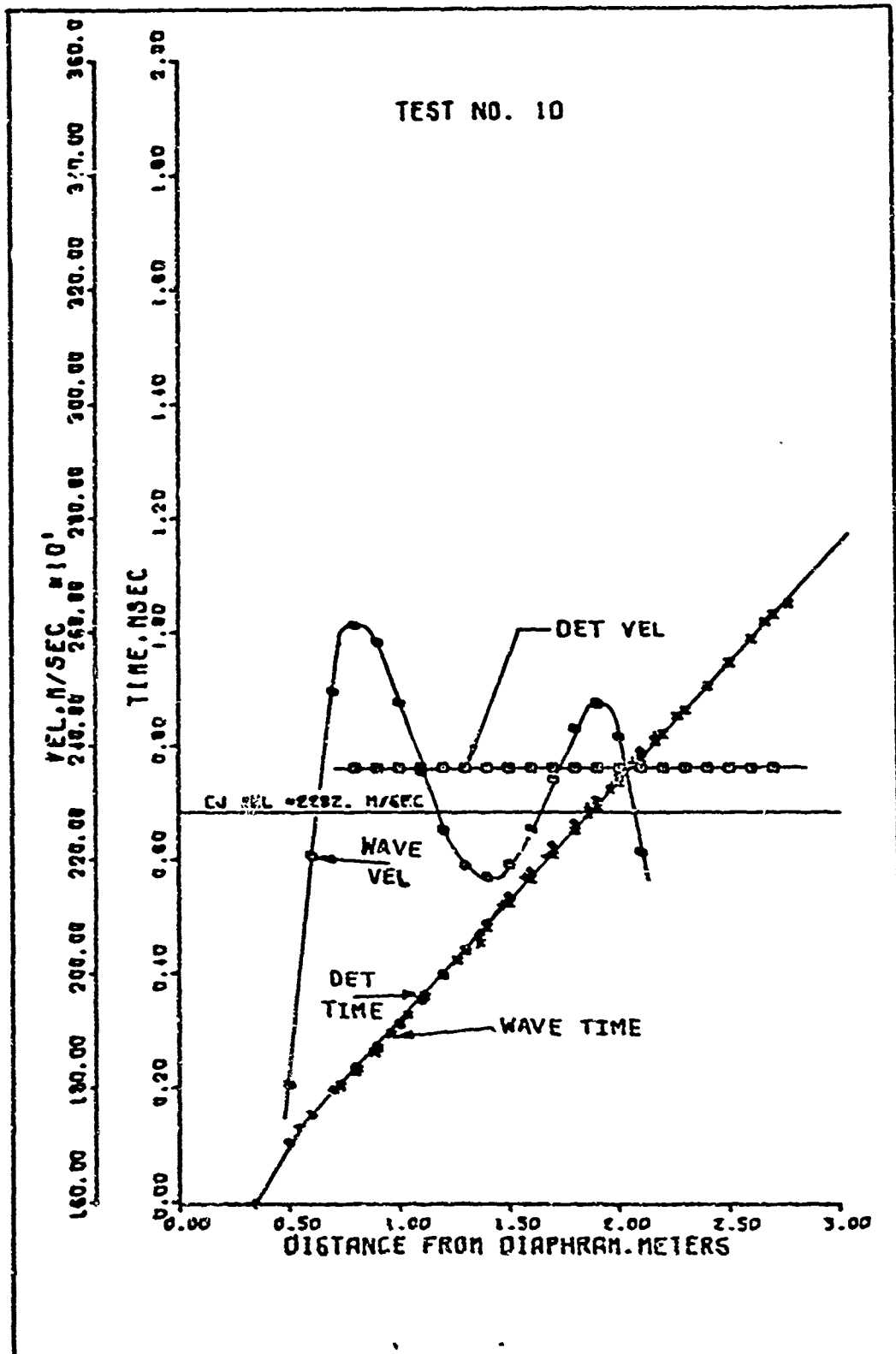


Figure 8. Plot of the Data Obtained From Test 10, 49.90% Hydrogen, 50.10% Oxygen.

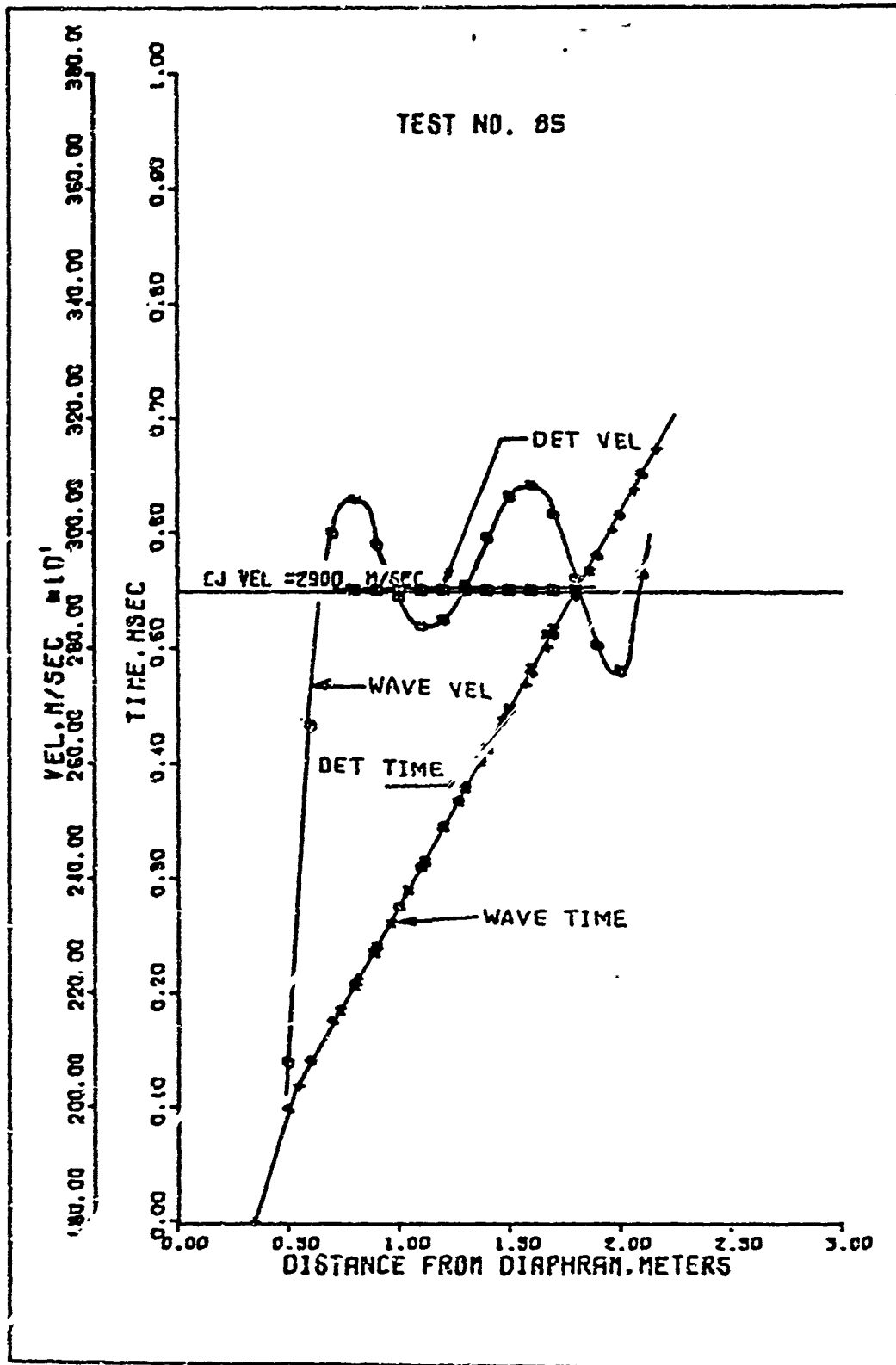


Figure 9. Plot of Data Obtained From Test 85, 69.67% Hydrogen and 30.33% Oxygen.

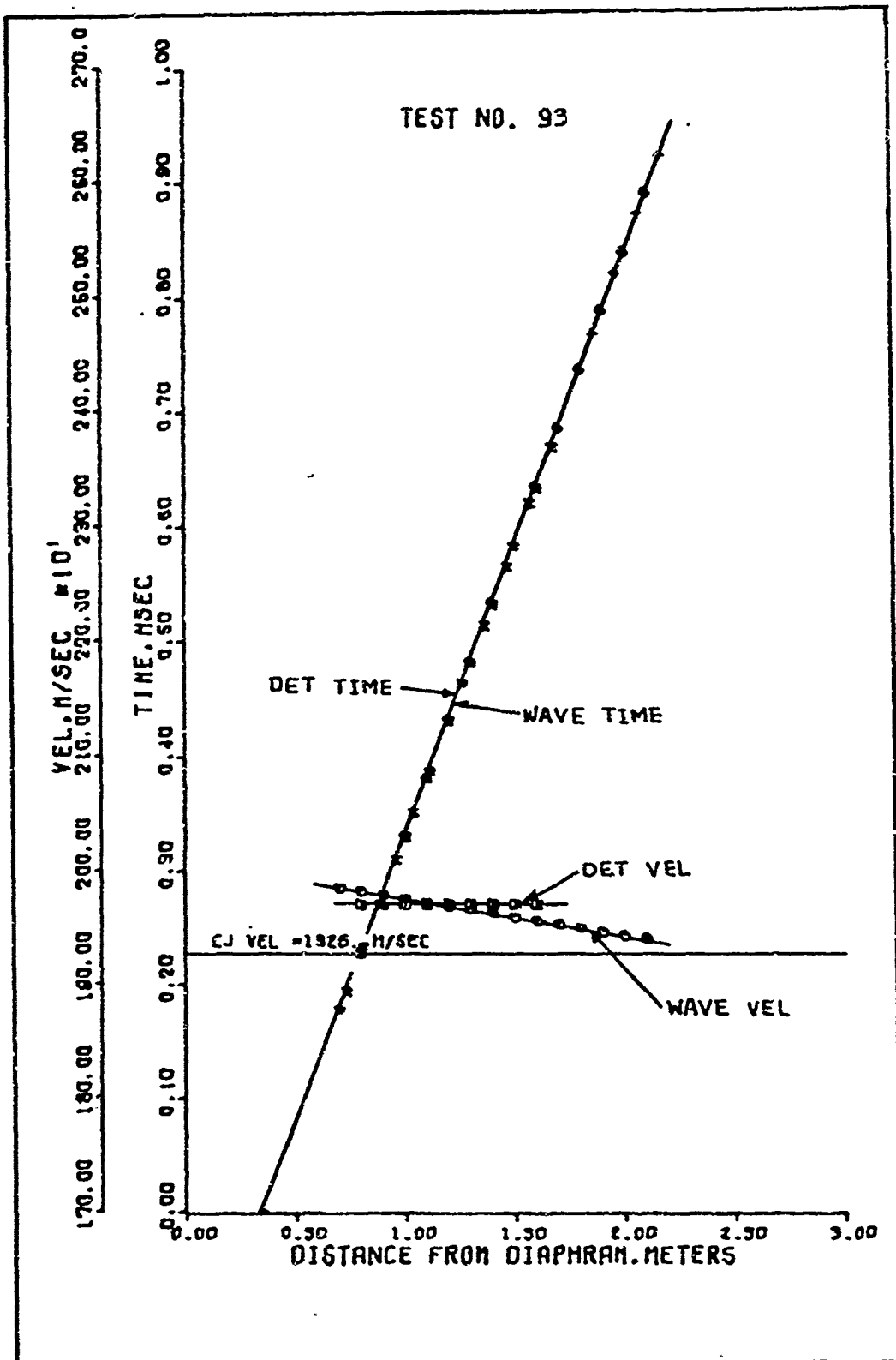


Figure 10. Plot of Data Obtained From Test 93, 36.84% Hydrogen, 13.87% Oxygen, and 49.29% Argon.

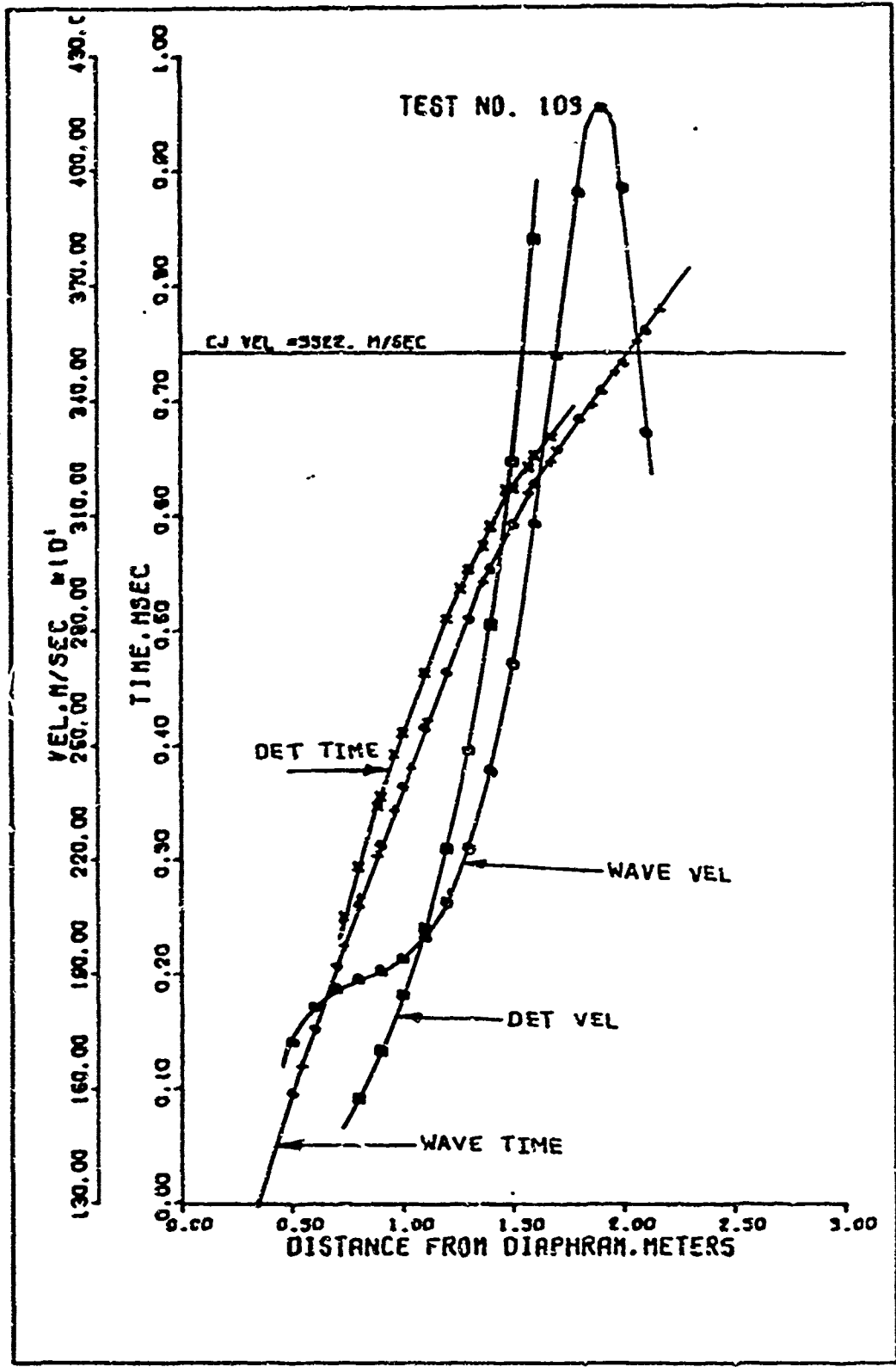


Figure 11. Plot of Data Obtained From Test 103, 35.40% Hydrogen, 14.94% Oxygen, and 49.66% Helium.

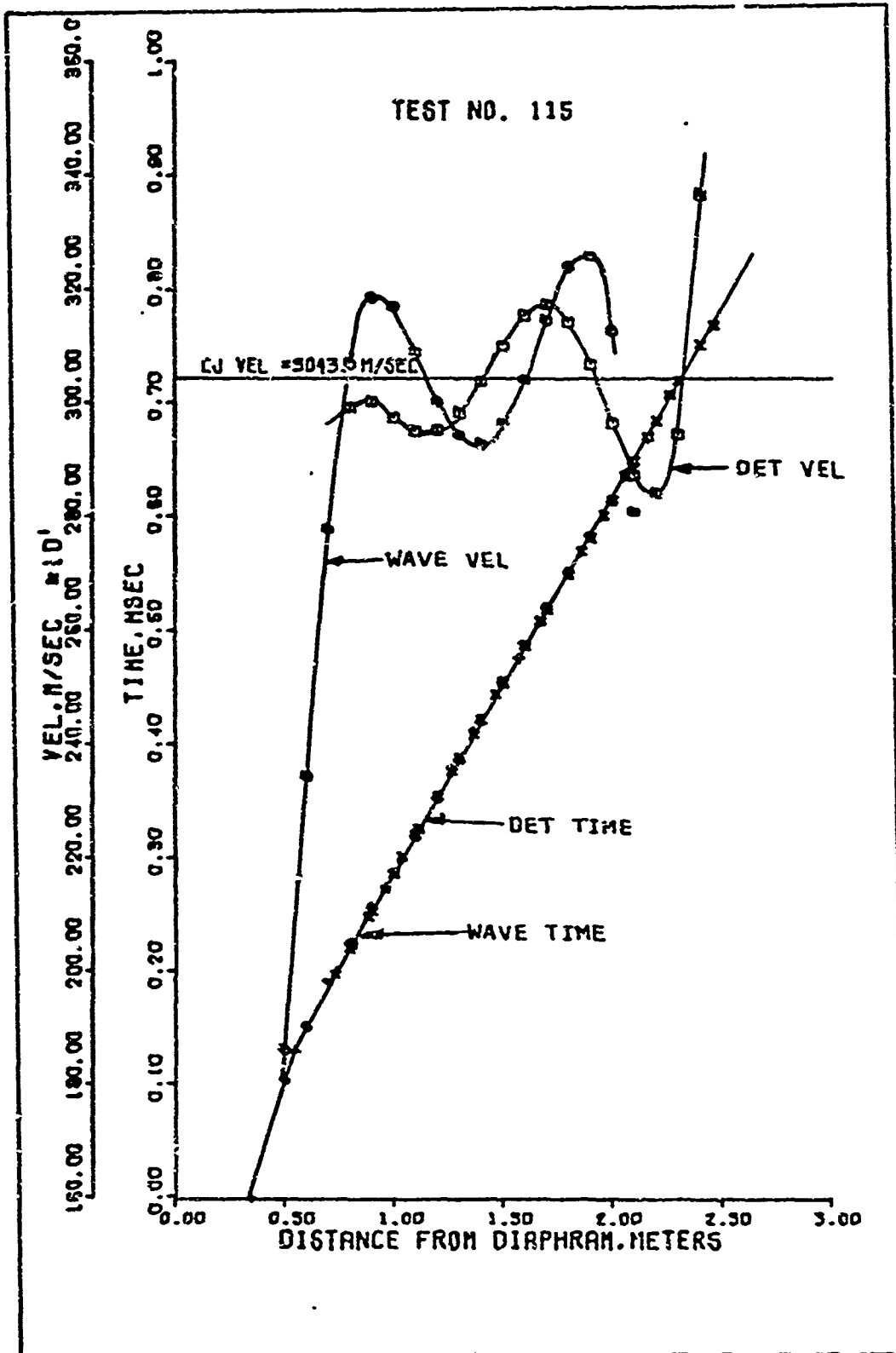


Figure 12. Plot of Data Obtained From Test 115, 73.17% Hydrogen and 26.83% Oxygen.

be formed somewhere behind a detonation, would travel at a speed equal to the local sonic velocity plus the local particle velocity. Brinkley and Richardson (Ref 2) have extended previous detonation theories to consider a finite reaction zone. They claimed that as a rarefaction wave proceeds forward into a reaction zone, toward the detonation shock front, it will find the temperature and local speed of sound dropping as it enters the relatively unreacted gases. Somewhere within the reaction zone, rarefaction waves will stabilize relative to the shock and the detonation will continue to propagate, but at a lower velocity. Compression waves, with their tendency to coalesce, would, of necessity, be formed somewhere behind the stabilized rarefaction waves and travel at speeds greater than rarefaction waves. Such compression waves would, therefore, overtake the rarefaction waves and proceed to interact with and strengthen the detonation front. This may lead to an instability comprising alternate periods of acceleration and deceleration. The acceleration occurring when the compression waves move forward and the deceleration occurring when the rarefaction waves move into the reaction zone. It could also happen that the compressions and rarefactions may merge and a stable detonation result (Ref 2:456-457). The pulsating nature observed for both the flame (see Fig. 12) and the shock fronts of the detonations shown apparently verify this theory. A related observation is that the flame velocities tend to be more

stable than those of the shock fronts. A few of the detonations appeared to be quite stable (see Fig. 10) as predicted.

Chapman-Jouguet Calculations

The calculated Chapman-Jouguet velocities, shown on the graphs, were obtained using a computer program written by Surette. The program uses the standard continuity, momentum, and energy equations as well as those for chemical equilibrium. Products assumed present include the following: H_2 , O_2 , H_2O , OH , H , O , HO_2 , H_2O_2 , and Ar or He. In all cases, the initial pressure of the test gas was ten inches of mercury. The initial temperature, however, could not be controlled or measured and varied some with climatic conditions. Theoretical computations were based on a temperature of 293 degrees Kelvin which introduced some small differences between theory and data observed. The calculations were considered adequate for comparative purposes.

Critical Mach Number

The critical Mach number is probably not a well defined value but rather a region. The number of tests conducted on each gas mix was sufficient to obtain approximate value for the critical Mach numbers. The approximate values obtained are presented in Table III.

Data for 90 percent mixes could not be presented because, as expected, very few tests detonated. It was not possible, with existing equipment, to obtain sufficiently high Mach

Table III
 Chapman-Jouguet Mach Number and Approximate
 Critical Mach Numbers for 0% and 50% Mixes

% H ₂	% O ₂	% Ar	% He	C-J Mach no.	Critical Mach (approximately)
49.90	50.10	0	0	5.07	2.5
62.42	37.58	0	0	5.17	2.1 to 2.3
69.67	30.33	0	0	5.14	2.2 to 2.9
73.17	26.83	0	0	5.17	2.1 to 2.5
27.68	25.94	46.38	0	4.92	not available
33.36	18.47	48.17	0	5.02	2.0 to 2.3
36.84	13.87	49.29	0	5.00	2.0 to 2.9
36.20	13.17	50.63	0	4.98	2.0 to 2.6
26.57	27.52	0	45.91	4.89	2.2 to 2.4
31.61	19.69	0	48.70	5.00	2.1 to 2.6
35.40	14.94	0	49.66	5.01	1.95
36.61	13.12	0	50.27	4.98	1.7 to 2.0

numbers to get detonation with 90 percent helium. It was also difficult to get initial Mach numbers below the predicted Chapman-Jouguet value with 90 percent argon. Therefore, the 90 percent mixes will be ignored in this discussion.

Table III shows the observed critical Mach numbers to be about one-half of the Chapman-Jouguet value for the cases tested. Determination of whether this is coincidence or not and exactly what the critical Mach numbers are, requires further testing. These values were significantly below those reported by Belles (Ref 1). The difference could be due to a number of factors, the most obvious of which is the pressure differences of the initial test gases. A relationship between critical Mach number and detonability could not be established.

Detonation Onset Distances

The distance required for onset of detonation is defined for this report as that distance of travel down the tube after which the initial shock achieves Chapman-Jouguet velocities. This distance is a function of many variables including the geometry and smoothness of the shock tube. However, it seemed reasonable that the distance would approach zero as the initial Mach number approached the Chapman-Jouguet value, and infinity as the initial Mach number approached the critical value. Figures 12 through 19

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show the detonation onset distances vs. initial Mach numbers obtained. This data is incomplete. However, it does show a hyperbolic trend which should be verified through more experimentation.

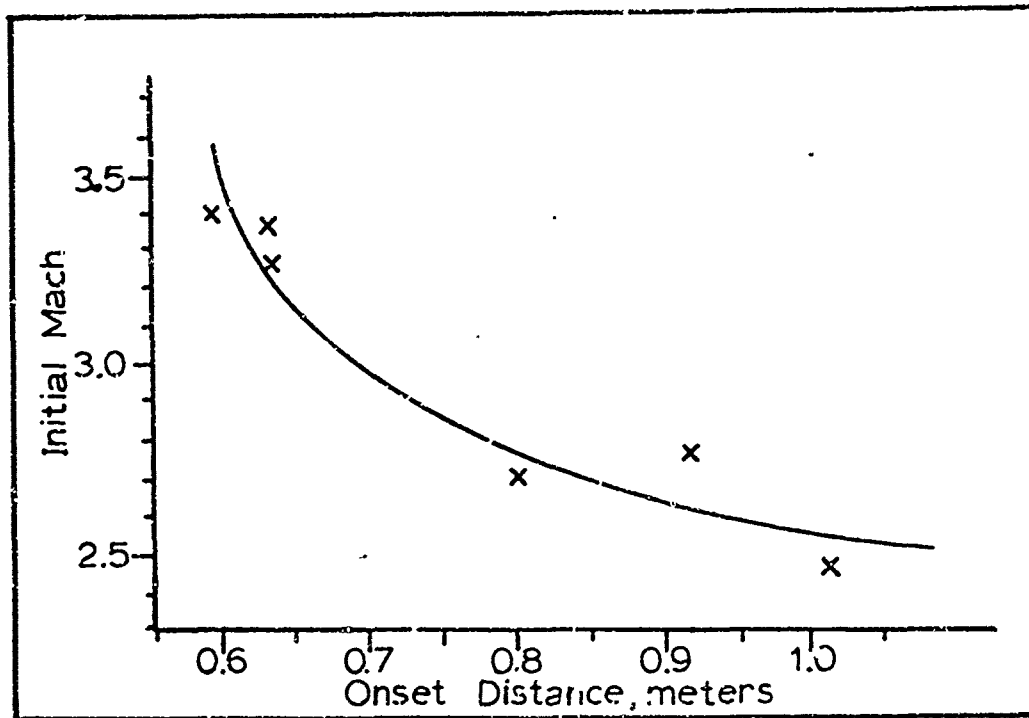


Figure 13. Detonation Onset Distance vs. Initial Mach Number for 49.90% H₂ and 50.10% O₂.

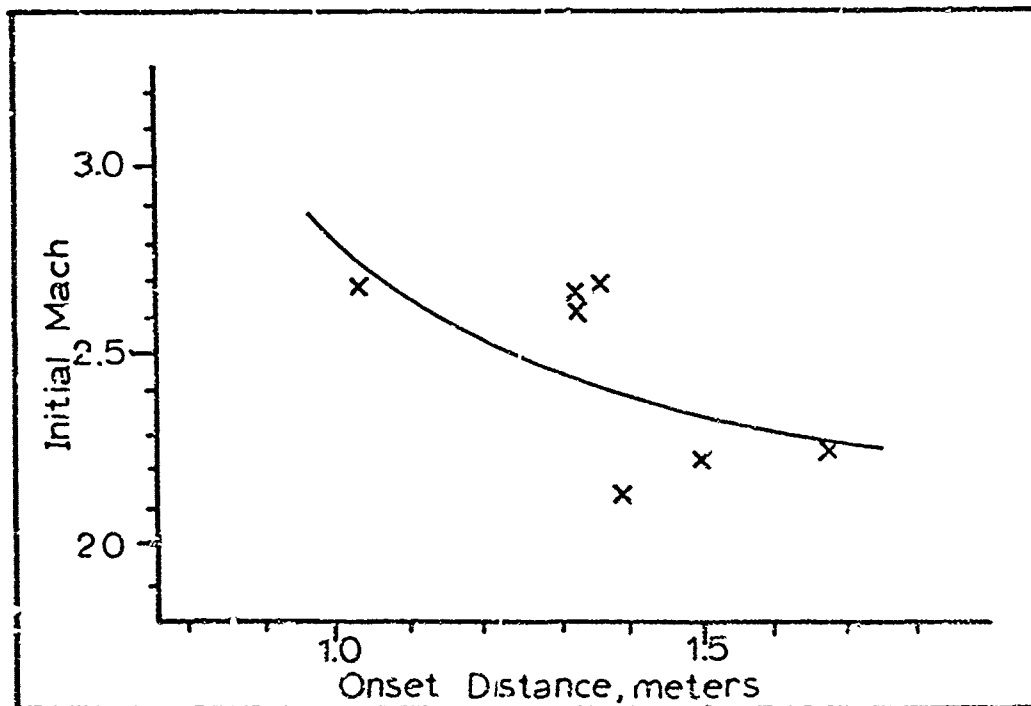


Figure 14. Detonation Onset Distance vs. Initial Mach Number for 26.57% H₂, 27.52% O₂, and 45.91% He.

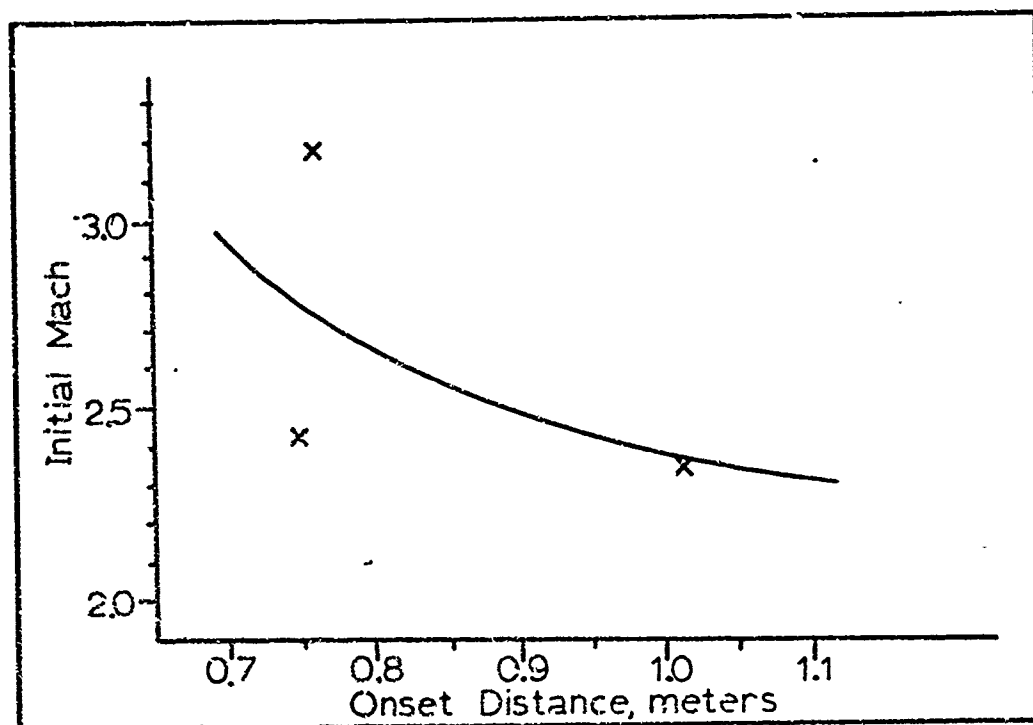


Figure 15. Detonation Onset Distance vs. Initial Mach Number for 62.42% H_2 and 37.58% O_2 .

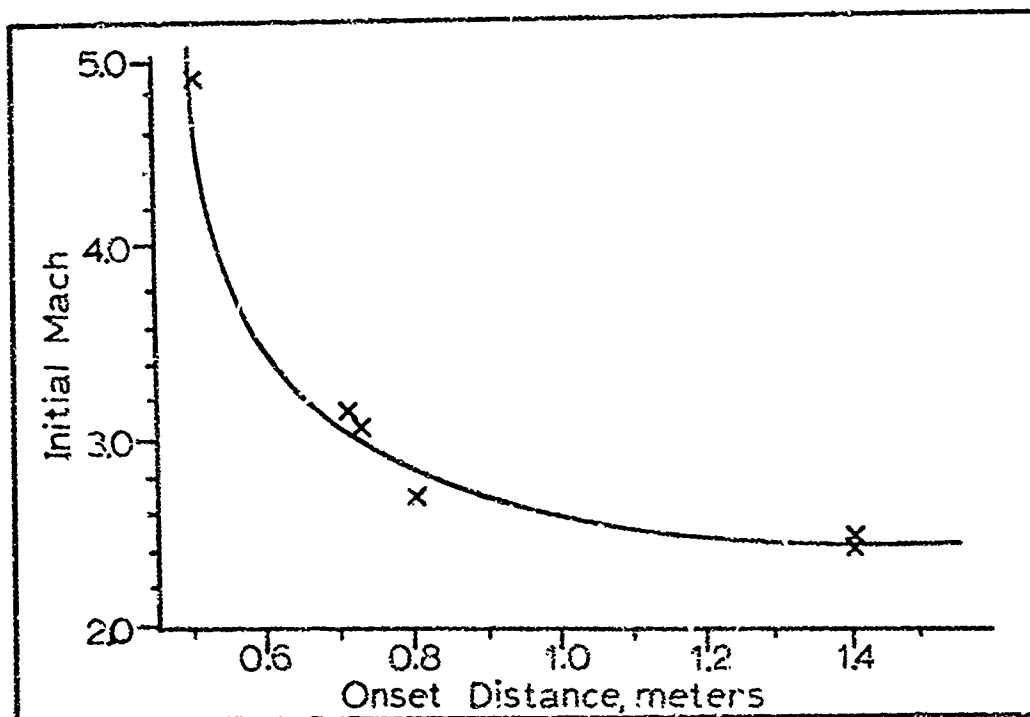


Figure 16. Detonation Onset Distance vs. Initial Mach Number for 33.36% H_2 , 18.47% O_2 , and 48.17% Ar .

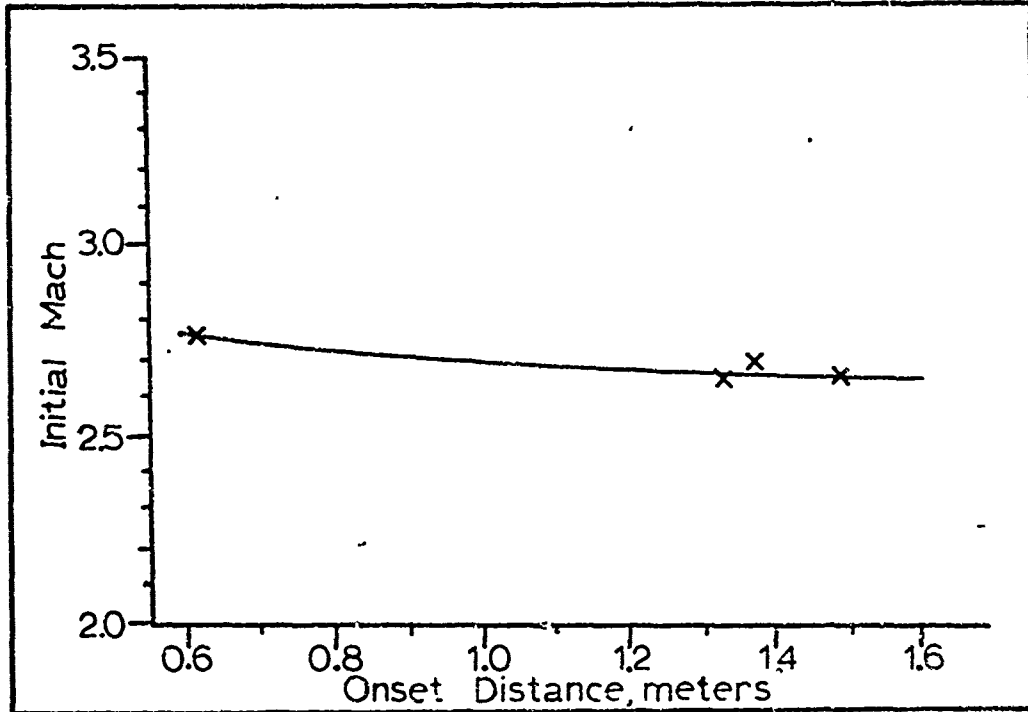


Figure 17. Detonation Onset Distance vs. Initial Mach Number for 31.61% H₂, 19.69% O₂, and 48.70% He.

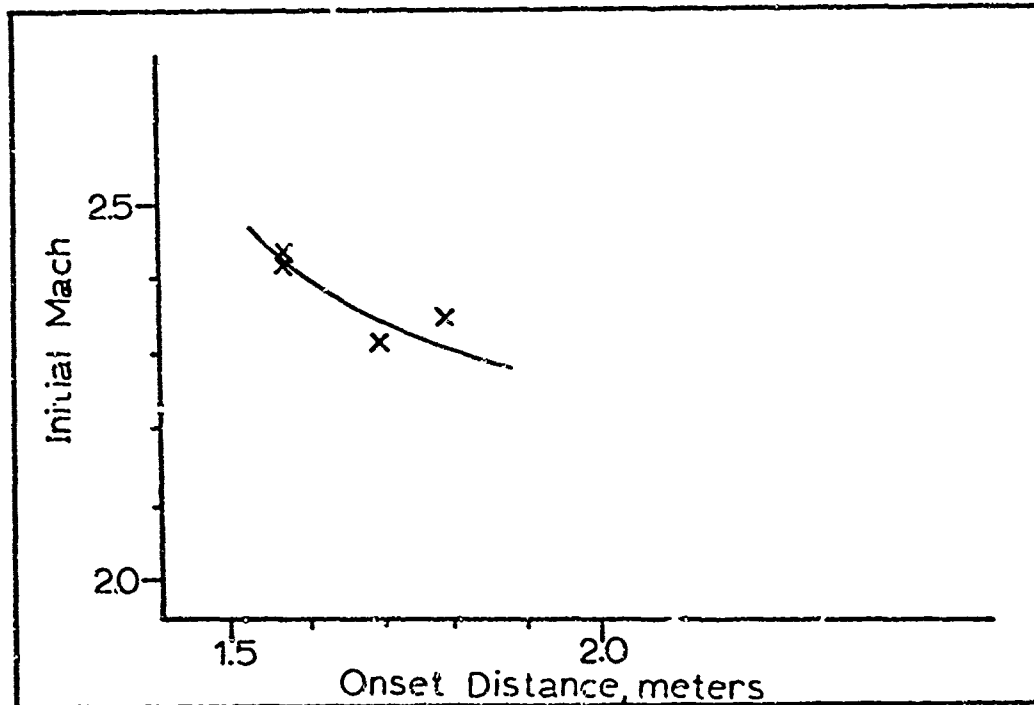


Figure 18. Detonation Onset Distance vs. Initial Mach Number for 35.40% H₂, 14.94% O₂, and 49.66% He.

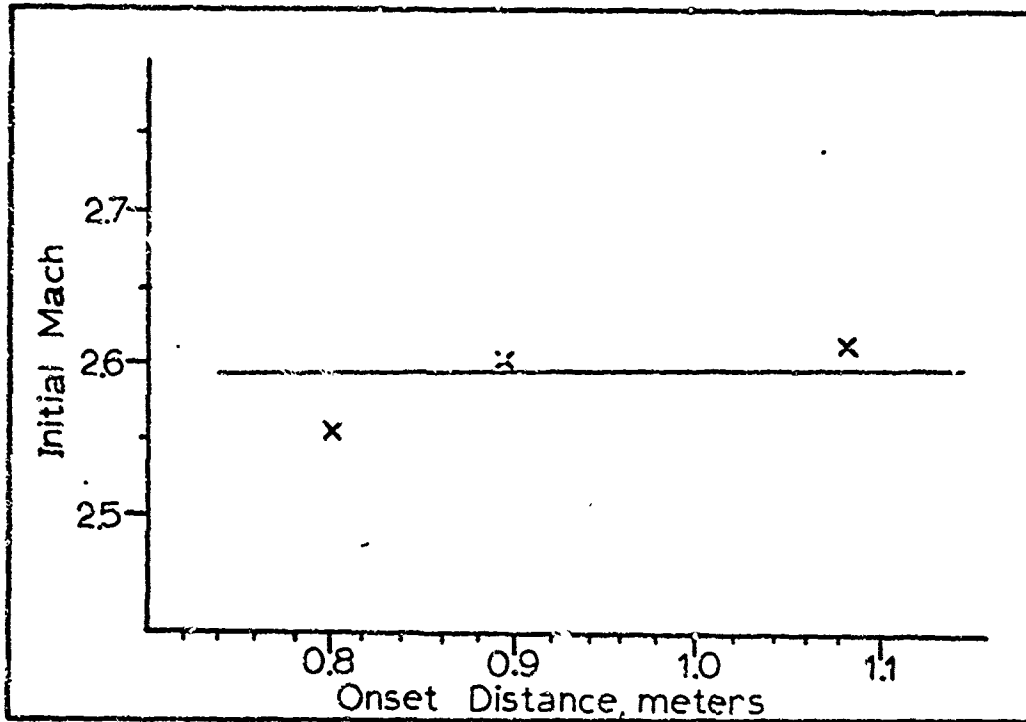


Figure 19. Detonation Onset Distance vs. Initial Mach Number for 73.17% H₂ and 26.8% O₂.

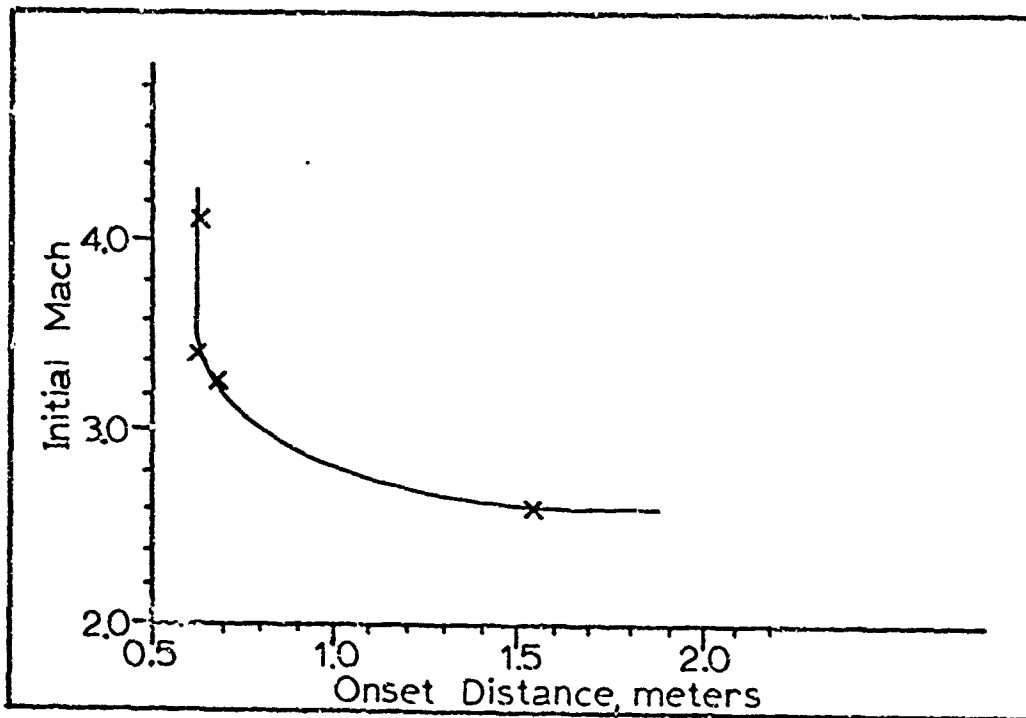


Figure 20. Detonation Onset Distance vs. Initial Mach Number for 36.20 H₂, 13.17% O₂, and 50.63% Ar.

V. Conclusions and Recommendations

Conclusions

The following conclusions were made from this investigation.

1. The observed pulsating velocities of the detonation waves tend to verify the theoretical structure of plane detonation waves proposed by Brinkley and Richardson.

2. Critical Mach numbers were approximately determined, for all gases tested the values and were found to be between 1.7 and 2.5. Furthermore, stoichiometry seemed to have a stronger influence on critical Mach numbers than did the diluent concentration.

3. The shock tube used was not adequate for test of 90 percent diluent mixtures.

4. Detonation onset distances are a function of initial Mach number and do tend to get long for near critical initial Mach numbers.

Recommendations

1. Further investigations of critical Mach numbers should be undertaken. However, more stoichiometric data, with and without diluents, should be obtained before proceeding to non-stoichiometric tests. These tests should be conducted for various initial pressures so that a shock initiated detonation limit curve can be obtained.

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Vita

William Francis Balmanno was born on 15 April 1943 in Beloit, Wisconsin, the son of William Crosby Balmanno and Virginia Anita Balmanno. Graduating from Saint Francis High School in Mountain View, California in 1961, he subsequently attended San Jose State College where he received a degree of Bachelor of Science in Mechanical Engineering in 1966, and a commission as a Second Lieutenant in the Air Force. He served as an Engineering manager at Vandenberg AFB for four years which entailed the launching of Atlas missiles down the Western Test Range. Captain Balmanno completed thirty of thirty-six semester hours with the University of Southern California towards a Masters degree in Aerospace Operations Management during this tour of duty. He entered the Air Force Institute of Technology in May, 1970.

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