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CONDENSATION OF H2O AND D2O IN ARGON IN THE CENTERED EXPANSION --ETC(U)

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January 1977

CONDENSATION OF H₂O AND D₂O IN ARGON
IN THE CENTERED EXPANSION WAVE IN A SHOCK TUBE

by

C.F. Lee

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Submitted by Peter P. Wegener, Principal Investigator.
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ABSTRACT

Despite gasdynamic non-idealities in the flow produced in a shock tube, pressure measurements at three different locations in the driver section of the shock tube revealed that the expansion wave generated in relatively weak expansions could be viewed effectively as a simple centered expansion fan after an empirical shift of the actual origin of the expansion wave to a "virtual" origin. The resulting centered expansion fan was used to study at two locations the condensation of H_2O and D_2O vapors in an excess of the carrier gas argon, with simultaneous pressure and light scattering measurements. The isentropic flow within the centered expansion fan was found to be preserved up to the point of the detectable onset of condensation by tailoring the onset conditions to occur at the tail of the expansion fan, thus rendering a simple analysis of the experiments possible. The onset conditions for H_2O vapor were found to be in agreement with previous findings in supersonic nozzles and shock tubes, and they were well predicted by the so-called classical theory of homogeneous nucleation. The condensation of D_2O vapor was found to exhibit similar trends as those of H_2O vapor condensation despite the slight differences in physical properties between them due to isotopy.

LIST OF SYMBOLS

a	sound speed
c	wave speed
g	condensate mass fraction
J	nucleation rate
p	pressure
t	time
T	temperature
u	fluid velocity
x	axial coordinate
γ	specific heat ratio
ρ	density
λ	wavelength
Γ	empirical adjustment factor

Subscripts

1	undisturbed state in the driven section
4	undisturbed state in the driver section
cl	classical theory of homogeneous nucleation
exp	experiment
k	state of condensation onset condition
o	origin of particle path
obs	observation station

INTRODUCTION

In addition to the well-known cloud chamber and supersonic nozzle methods of condensation research, the shock tube offers a technique that has many desirable features. As a batch experiment like the cloud chamber it has the advantage of accurately controlled experimental conditions and easy exchange of test substances. Only small amounts of condensing substances are required which can be prepared with great care. Thus, toxic as well as costly materials can be studied. The wholly confined system can be thoroughly cleaned by pumping it to high vacuum prior to an experiment. This greatly reduces the problems of contamination and seeding by foreign substances which may lead to undesirable complications such as binary nucleation or heterogeneous nucleation, particularly at low cooling rates. With appropriate cooling and heating facilities a wide range of substances may be investigated over a wide range of experimental conditions.

The shock tube was first applied to condensation studies by Wegener and Lundquist¹; and the first streak photographs showing condensation zones in the expansion fan in the driver section were provided by Glass and Patterson². Since then there have been many different applications of different parts of the shock tube flow to condensation studies. For example, Homer^{3,4} utilized the constant thermodynamic condition behind the incident shock wave to study the nucleation and growth of lead particles. Kung and Bauer⁵ investigated the nucleation of iron vapor with the so-called tailored interface technique which exploited the expansion fan produced as a result of the

tailored interaction between the reflected shock and the contact surface. The method described in this work makes use of the cooling capabilities of the unsteady isentropic expansion wave in the driver section to induce condensation. The onset of condensation as indicated by the first appearance of condensation aerosols can be detected by Rayleigh scattering of laser light whose wavelength ($\lambda = 4416 \text{ \AA}$ for a Helium-Cadmium laser) is much larger than the size of the particles generated under these experimental conditions (normally a few hundred Angstroms); while the sudden release of the latent heat of condensation gives rise to a pressure "bump". Such a method has been used by Barschdorff⁶ to study the carrier gas effects on the homogeneous nucleation of water vapor. Courtney⁷ has also used the expansion wave to study the condensation of water vapor, methanol and carbon tetrachloride. Kawada and Mori⁸ has applied a similar method to droplet growth measurements using light transmission as well as Mach-Zehnder Interferometry. More recently Kalra⁹ has investigated the condensation of water vapor in an expansion wave using both laser Fabry-Perot Interferometry as well as differential interferometry.

Since the flow history as observed at a fixed location is different depending on its distance from the diaphragm location, the expansion wave offers the possibility of studying the effects of cooling rates on condensation processes by making observations at different locations along the driver section thus covering a range of cooling rates in a single experiment. The average cooling rates in most supersonic nozzle condensation experiments are 10^6 C/s^{10} and in Barschdorff's shock tube experiments $5 \times 10^5 \text{ C/s}$. In the experiments described in this work the typical average

cooling rates of 5×10^4 C/s were lower, approaching those of the cloud chamber.

THE CENTERED EXPANSION FAN

The heart of the shock tube technique used in the condensation experiments here lies in the understanding of the gasdynamics associated with the unsteady expansion flow in the driver section. It is recalled that a simple, isentropic, centered expansion wave is produced in the driver section provided that: (i) the complete removal of the diaphragm is instantaneous and the flow is set into motion abruptly; (ii) there are no three-dimensional and viscous effects; and (iii) the fluids employed behave like perfect gases. It is well-known that in such an isentropic expansion wave the flow properties of fluid velocity, density, pressure, temperature, etc. are constant along the characteristics of the governing differential equations^{11,12}. In an $x - t$ plane, these characteristics are straight lines of constant x/t , whose slope corresponds to the local wave speed, c , given by

$$c = a_4 \left[-\frac{2}{\gamma_4 - 1} + \frac{\gamma_4 - 1}{\gamma_4 + 1} \cdot \left(\frac{p}{p_4} \right)^{\frac{\gamma_4 - 1}{2\gamma_4}} \right], \quad (1)$$

- where
- a_4 = sound speed of the undisturbed medium in the driver section,
 - γ_4 = ratio of specific heats of the undisturbed medium in the driver section,
 - p_4 = pressure of the undisturbed medium in the driver section,
 - p - local pressure within the expansion wave along the characteristic in question.

The static pressure variation with time, $p(t)$, at a fixed location, x_{obs} , is given by,

$$\frac{p(t)}{p_4} = \left[\frac{2}{\gamma_4 + 1} + \frac{\gamma_4 - 1}{\gamma_4 + 1} \cdot \left(\frac{x_{\text{obs}}}{a_4 t} \right) \right]^{\frac{2\gamma_4}{\gamma_4 - 1}}, \quad (2)$$

and the time taken by the first characteristic (the head of the expansion wave) to reach x_{obs} is given by,

$$t_{\text{obs}} = x_{\text{obs}} / a_4. \quad (3)$$

The isentropic relationships from Poisson's Equations relate the remaining flow variables by

$$\frac{p}{p_4} = \left(\frac{\rho}{\rho_4} \right)^{\gamma_4} = \left(\frac{T}{T_4} \right)^{\frac{2\gamma_4}{\gamma_4 - 1}} = \left(\frac{a}{a_4} \right)^{\frac{2\gamma_4}{\gamma_4 - 1}}. \quad (4)$$

Each fluid element traverses the expansion fan with a local velocity, u , given by

$$u = (c - a_4) \cdot \frac{2}{\gamma_4 + 1} \quad (5)$$

along a particle path given by

$$x(t) = - \frac{a_4 t}{(\gamma_4 - 1)} \left[2 - (\gamma_4 - 1) \left(\frac{x_0}{a_4 t} \right)^{\frac{\gamma_4 - 1}{\gamma_4 + 1}} \right], \quad (6)$$

where x_0 defines the initial position of the particle path.

However, possible non-idealities in the real flow due to the bulging of the diaphragm under pressure before

rupture, finite opening time, three-dimensional initial flow, boundary layer effects, etc. may cause the real expansion flow to deviate from that predicted by the ideal theory. Moreover, for condensation studies, one has to trace each fluid element under observation back to its initial position in the driver section and integrate the amount condensed along the particle path in order to compute the total condensate mass fraction, $g(x_{\text{obs}}, t)$, at a given time and at a given location, x_{obs} . In order to define the particle path uniquely the starting point or origin of the expansion fan has to be known. Therefore, prior to the condensation study, experiments on the flow itself using dry gases were made on the expansion flow actually produced in the shock tube.

The experiments were performed in a 76.2 mm (3 inches) I.D. steel shock tube which was later also used for condensation studies. Prior to an experiment, the tube was outgassed by evacuating it to 10^{-6} torr for an hour with a cold-trapped diffusion pump. The tube was then filled with high-purity scientific-grade argon. After recording the initial temperatures and pressures in the two section the flow was started by rupturing the stretched diaphragm with a spring-loaded needle.

Pressure time histories at three different locations situated at 127 mm (5 inches), 178 mm (7 inches), and 495 mm (19.5 inches) respectively upstream of the diaphragm (i.e. into the driver section) were measured by three Kistler Model 606L pressure transducers mounted flush with the inner tube wall and electrically insulated from it. The charge outputs from the transducers were

converted to amplified voltage signals by Kistler Dual Mode Amplifiers (Model 504E) and were displayed on the storage screen of a fast single sweep oscilloscope (Tektronix Model 5103N).

This procedure was repeated for many different values of initial diaphragm pressure ratios ranging from $p_4/p_1 = 2.0$ to $p_4/p_1 = 8.0$. Whereas the initial temperatures in both sections were kept at ambient temperatures, the initial pressure in the driver section was varied from one to two atmospheres while that in the driver section never went below 150 torr. Under these conditions, the terminating characteristics of the relatively weak expansion waves produced would remain in the driver section. Hence the outflow from the driver section remains subsonic.

Figure 1 shows a typical oscillogram from which points of equal pressure at different locations and times were mapped in an $x - t$ plane as shown in Figure 2. The isobars drawn through these experimental points are found to be straight lines merging at a single point not far from the origin of the $x - t$ diagram. This result seen in Figure 2 indicated that viscous effects were negligible since a boundary layer would give rise to an effectively variable-area duct causing the characteristics to curve. Moreover, the interaction between a wall boundary layer and an initially centered wave would lead to a non-centered wave¹³. Figure 3 shows the remarkable agreement between the wave speeds determined from the slopes of the isobars and the ideal centered-wave theory. This result further demonstrates the fact that the real expansion flow produced can be viewed effectively as a simple centered wave

with a "virtual" origin shifted with respect to the actual diaphragm location. The virtual origin can be determined best by noting the value of x_{obs} which gives a best fit between the experimental pressure variation and the theoretical curve of Equation (2). The values of t_{obs} for each of the three observation stations determined in this manner agreed well with those found from Figure 2. Using experimental values of t_{obs} , the pressure measurements from Figure 1 can finally be represented by a single curve of p/p_4 versus t/t_{obs} as shown in Figure 4.

It is worthy of note that the relatively low initial diaphragm pressure ratios ($2 \leq p_4/p_1 \leq 8$) employed in these experiments produced only weak expansion waves ($0.32 \leq p_3/p_4 \leq 0.70$). Therefore the flow occurred at high Reynolds numbers (Re). It is well-known¹² that deviation from theory grows with increasing expansion wave strength, namely low p_3 and low Re, which would produce more serious viscous effects resulting in thicker boundary layers. Our primary error results from the diaphragm that actually bulges under pressure before rupture, thus physically contributing to a shift of the starting point of the expansion wave to the right in Figure 2.

CONDENSATION EXPERIMENTS

The experiments on the condensation of H_2O and D_2O were performed in the same shock tube as shown schematically in Figure 5. As before, prior to an experiment the whole tube was evacuated to 10^{-6} torr. The condensing substance normally in a liquid state was first injected

into a vaporizing chamber heated by electrical heating tapes. The vapor was then carefully metered into the shock tube via a needle valve which was electrically heated to avoid condensation. The H_2O used was triply distilled water and in the case of D_2O , a 99.8% Deuterium heavy water was used. (Wilmad Glass Co., Inc. of New Jersey). The initial partial pressure of the condensing vapor was measured with a calibrated Statham electric pressure transducer while the temperature was measured with six calibrated thermocouples distributed along the driver section. The tube was next filled with filtered and cold-trapped ultra-high-purity argon (min. purity 99.999%). The pressure in the driver section also filled with argon was preset. The gas mixture in the driver section was then stirred for 15 minutes by circulation through a sealed Metal Bellows pump. After a steady state of all properties was reached the experiment was initiated by rupturing the diaphragm.

Two Kistler transducers measured the static pressure variation at 415 mm and 810 mm upstream from the diaphragm. Rayleigh light scattering at 90° of a 15 mW Helium-Cadmium laser ($\lambda = 4416$ Angstrom) was measured by two photomultiplier tubes (RCA 7265) having a fast response and a high sensitivity at the wavelength of the laser. The pressure and light scattering signals were recorded by an oscilloscope triggered by a third Kistler transducer mounted 50 mm upstream of the first observation station.

Onset of condensation was indicated by a pressure bump and the simultaneous first appearance of the light scattering signal.

The heat addition due to condensation in the subsonic flow field generates waves which can propagate both upstream

and downstream, thus causing the pressure profile to deviate from the isentropic pressure variation. Inside the condensation zone the characteristics become curved due to increasing sound speed caused by heat addition. If condensation onset, i.e. the critical supersaturation, is the major interest the problems due to heat addition can be avoided. The initial diaphragm pressure ratio can be adjusted such that the tail of the expansion fan produced coincides with the condensation onset locus at the observation station as shown schematically in Figure 6. Here the isentropic flow is preserved at all locations in the driver section up to the point of condensation onset, a state that coincides with the tail of the centered expansion fan⁶. Since the condensation onset conditions are not known before hand an iterative experimental procedure is required to "home in" on the onset conditions. Figure 7 shows such an onset experiment where pressure and light scattering traces from an expansion with argon only under the same experimental conditions, are superimposed on the experiment where condensation has just begun. In analyzing the data in such a situation it is reasonable to assume that the accumulation of condensate and hence the heat addition due to condensation are negligible in the expansion fan prior to the instant of detectable onset of condensation. Therefore no departure from ideal flow has taken place.

RESULTS AND DISCUSSION

Experimental results are presented in Table 1 and condensation onset conditions are presented in a plot of pressure versus temperature as shown in Figure 8. Here the

equilibrium vapor pressure curves of both H_2O and D_2O as well as some condensation onset data of H_2O from other sources in supersonic nozzles¹⁴⁻¹⁷

and a shock tube⁶ are shown. Apparently no condensation results of D_2O are to date available in the literature. In Figure 8 we note a partial overlap between this work and Barschdorff's shock tube data. In general the nozzle results lie to the left of the shock tube results. This difference is to be expected. It can be explained on the basis of a difference in cooling rates. The average cooling rates were 10^6 C/s in nozzles, 5×10^5 C/s in Barschdorff's work and 5×10^4 C/s in this work. It is well-known that for a given initial relative humidity greater supersaturation can be achieved at higher cooling rates^{10,18}. The same argument also sheds light on the observation (Table 1) that the onset data measured at the first station have a greater supercooling compared to those at the second station. The first observation station is closer to the diaphragm location and therefore it has a cooling rate roughly twice that at the second station as can be deduced from data such as those of Figure 2. The described cooling rate effect was found for both H_2O and D_2O condensation.

In order to compare experiments with nucleation theory one needs to evaluate the condensate mass fraction accumulated along a particle path finally reaching the experimentally determined point of condensation onset. The formulation of this problem leads to a system of integro-differential equations similar to those first proposed by Oswatitsch¹⁹ for nozzle flow. These equations have been treated elsewhere^{6,20} and they will not be repeated here.

The comparison between theory and experiment is finally accomplished in terms of a single empirical adjustment factor, Γ , such that the actual nucleation in a given experiment, J_{exp} , is given by²¹

$$J_{\text{exp}} = \Gamma \cdot J_{\text{cl}}. \quad (7)$$

where J_{cl} is the nucleation rate based on the classical theory of homogeneous nucleation developed by Becker and Döring (1935)²², Volmer (1939)²³, and Frenkel (1946)²⁴, et al. For each experiment, the value of Γ is varied in the corresponding calculation until $g(x_{\text{obs}}, t_k) \approx 10^{-3}$ at the instant of time when the computed J_{exp} from Eq.(7) matches well with the measured onset conditions. Since there is doubt as to whether the state of the condensate at onset conditions is of a solid or supercooled liquid, the experimental results were evaluated for both cases of solid and liquid condensates.²¹

Despite the lack of reliable property data below the melting point, (e.g. surface tension of solid H₂O and D₂O), and the uncertainty of extrapolation when supercooled liquid property data are not available (e.g. D₂O), it can be seen from Table 2 that the values of $\log_{10}\Gamma$ obtained in this work are in reasonable agreement with those found in previous studies^{17,6}. The relatively low values of Γ further confirm the remarkable fact that the classical homogeneous nucleation theory in the case of H₂O and D₂O serves as a good prediction of the onset of condensation.

ACKNOWLEDGEMENTS

The author wishes to thank Professor Peter P. Wegener and Benjamin J.C. Wu of Yale University for their valuable advice and encouragement.

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FIGURE CAPTIONS

- Figure 1. Oscilloscope traces showing experimentally measured pressure variations at three different locations situated at 127 mm, 178 mm and 495 mm respectively upstream from the diaphragm location. For the first (bottom) and second (top) stations, 99 Torr/div. vertical; for the third station (middle), 109 Torr/div. vertical. The common time base is 0.5 ms/div. horizontal. The gas employed was argon. $p_4 = 777.1$ Torr; $p_4/p_1 = 3.70$; $T_4 = T_1 = 297.4$ K.
- Figure 2. Pressure measurements of Fig. 1 mapped into an x-t plane.
- Figure 3. Wave speeds determined from the x-t diagram in Fig. 2. ——— Centered Expansion Wave Theory.
- Figure 4. p/p_4 versus t/t_{obs} plot. \circ first station; \square second station; \triangle third station; ——— centered expansion wave theory.
- Figure 5. Schematic diagram of the shock tube for condensation experiments. (Dimensions in mm.)
- Figure 6. Condensation zone within the centered expansion fan. Pressure variations at x_{obs} and along a particle path reaching the point of condensation onset.
- Figure 7. Oscilloscope traces showing isentropic flow in the expansion fan is preserved in a condensation experiment up to the point of condensation onset.

Figure 8. Plot of pressure versus temperature showing experimentally determined condensation onset conditions. ● H₂O, this work; ■ D₂O, this work; ▲ H₂O, Nozzles¹⁴⁻¹⁷; □ H₂O shock tubes⁶

Table 1. Initial and onset conditions of a selected number of experiments.

Table 2. Summary of H₂O and D₂O condensation onset data.

TABLE 1. INITIAL AND ONSET CONDITIONS OF A SELECTED NUMBER OF EXPERIMENTS

Condensation of Water (H₂O) in Argon

Expt. No.	Initial Conditions			Onset Conditions at Station 1			Onset Conditions at Station 2				
	P ₄ (torr)	P _{v4} (torr)	T ₄ (K)	ω ₄	P _{vk} (torr)	T _k (K)	(P _v /P _∞) _k	P _{vk} (torr)	T _k (K)	(P _v /P _∞) _k	T _k -T ₄ (°C)
67	1216.0	17.3	299.5	0.0065	10.2	254.6	9.6	11.5	256.9	8.9	-16.3
98	1414.6	15.5	298.1	0.0050	9.2	251.6	11.2	9.2	252.8	10.1	-20.4
70	1330.0	13.9	297.8	0.0047	8.3	249.7	11.9	7.9	252.9	8.5	-20.3
73	1370.9	12.2	297.9	0.0040	5.9	247.4	10.3	6.8	249.2	10.1	-24.0
71	1345.5	10.5	297.8	0.0035	5.6	245.1	12.1	6.4	247.3	11.3	-25.9
77	1392.0	8.8	298.4	0.0029	5.1	243.7	12.6	5.1	244.8	11.4	-28.4
78	1398.2	7.1	298.3	0.0023	4.0	239.9	14.2	4.1	242.3	14.1	-30.9
80	1407.2	5.4	298.0	0.0017	2.7	233.8	17.6	2.6	234.9	15.2	-38.3
82	1454.8	3.8	298.0	0.0012	1.8	231.8	21.8	1.7	232.9	17.8	-40.3

Condensation of Heavy Water (D₂O) in Argon

10	1399.7	17.3	298.5	0.0062	11.0	255.5	12.1	9.0	255.6	9.8	-21.4
14	1401.7	15.6	298.0	0.0056	10.1	253.5	13.2	10.1	254.9	11.8	-22.1
19	1437.1	13.8	298.4	0.0048	7.5	249.8	13.7	7.8	251.5	12.2	-25.5
24	1421.6	12.2	298.6	0.0043	7.3	248.0	15.8	7.3	250.9	12.1	-26.1
44	807.5	9.7	297.3	0.0061	5.4	244.6	15.9	5.3	246.0	13.8	-31.0
48	804.5	8.0	297.4	0.0050	4.1	242.1	15.5	4.2	243.7	13.7	-33.3
54	797.1	6.3	297.4	0.0040	3.4	238.2	19.0	3.5	240.1	16.5	-36.9
55	823.7	4.6	297.5	0.0028	2.4	235.0	18.5	2.1	235.8	15.1	-41.2
57	822.9	2.9	297.5	0.0018	1.5	226.3	29.6	1.5	230.7	19.1	-46.3

Table 2. Summary of H₂O and D₂O condensation onset data

Substance	$\Delta T = T_k - T_{mp}$ (°C)	$\log_{10} \Gamma$		References
		solid	liquid	
Steam	0 to + 40	—	-2 to 1	Nozzles (Experiments ²⁵⁻²⁸ and Γ calculations ²¹)
H ₂ O	-62 to -19	5 to 7	3 to 4	Nozzles (Experiments ¹⁴⁻¹⁶ and Γ calculations ²¹)
H ₂ O	-48 to -12	—	1 to 6	Shock Tube ⁶
H ₂ O	-41 to -16	1 to 3	3 to 6	This work
D ₂ O	-51 to -21	1 to 2	1 to 3	This work

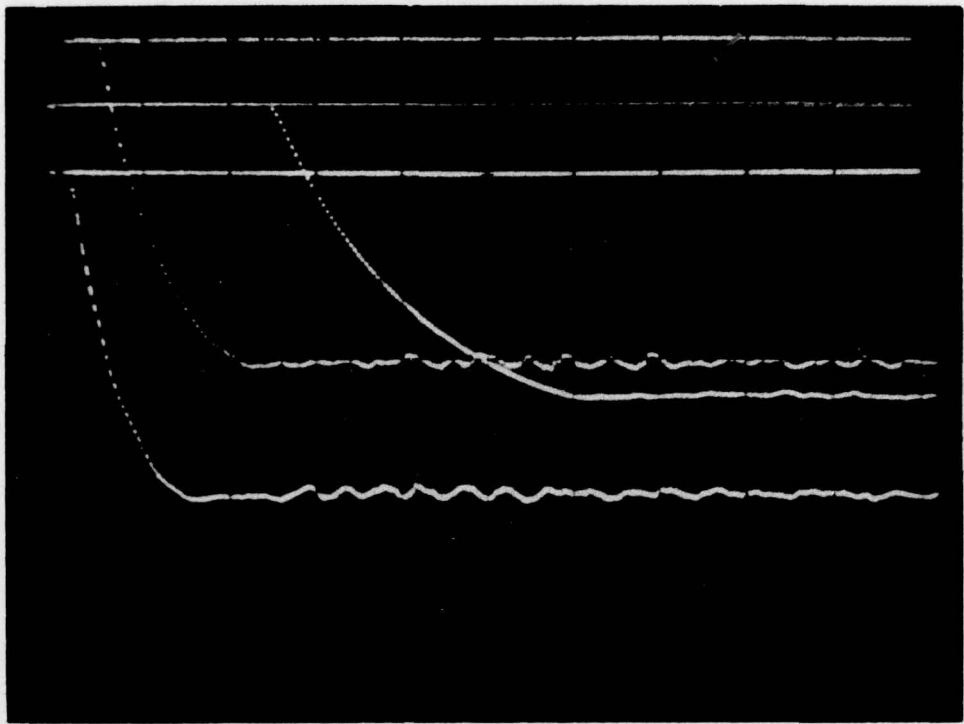


Figure 1

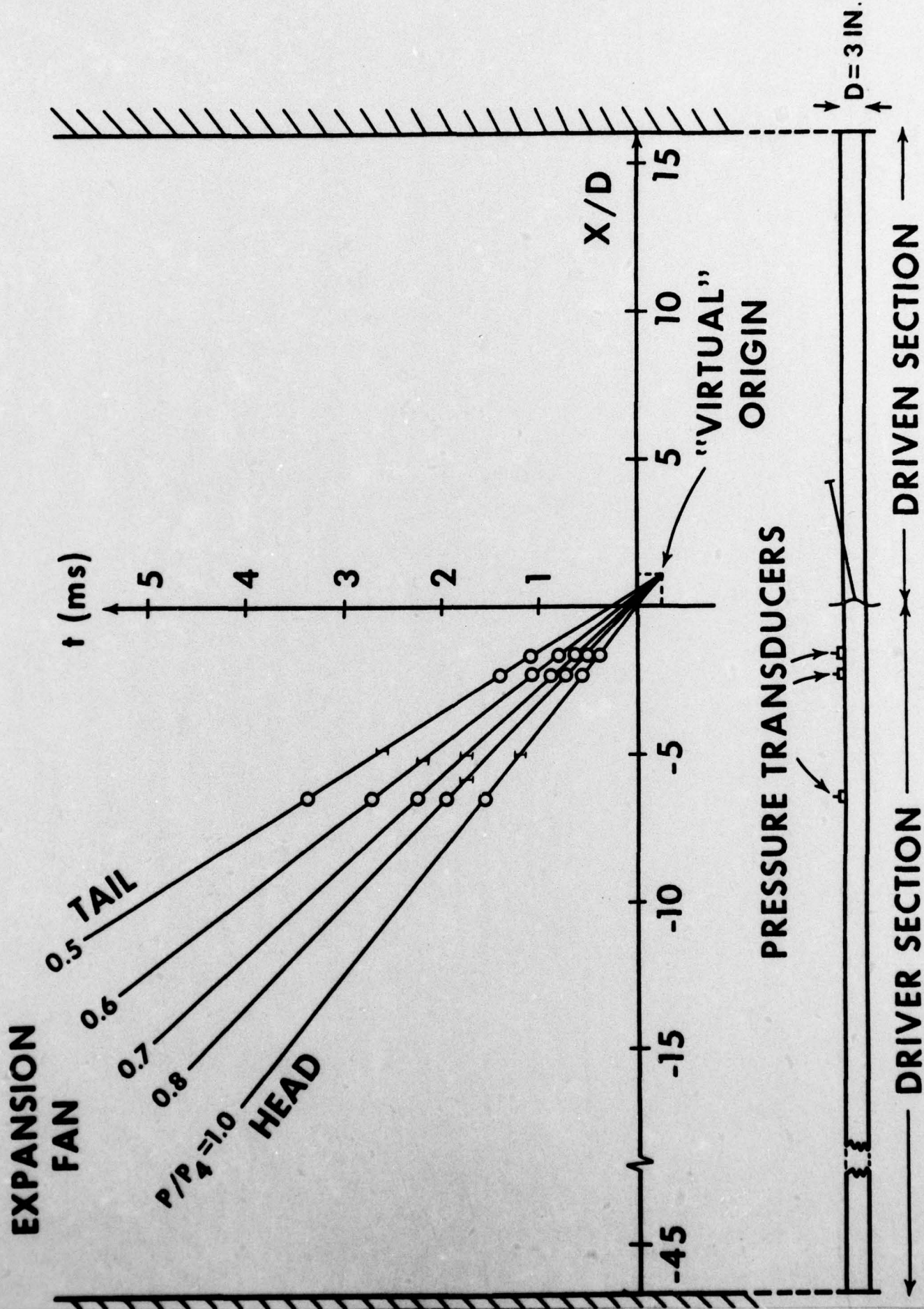


Figure 2

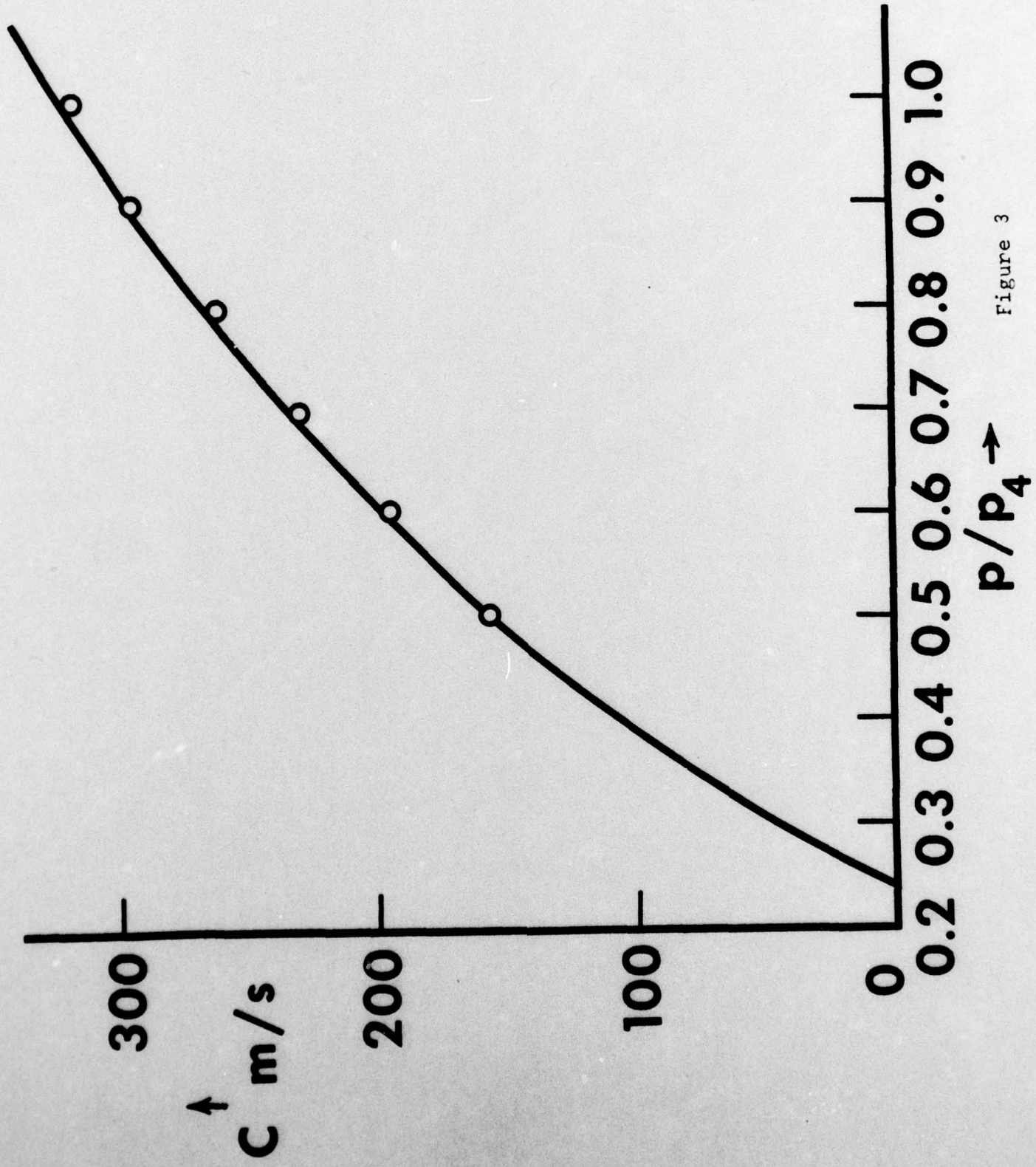


Figure 3

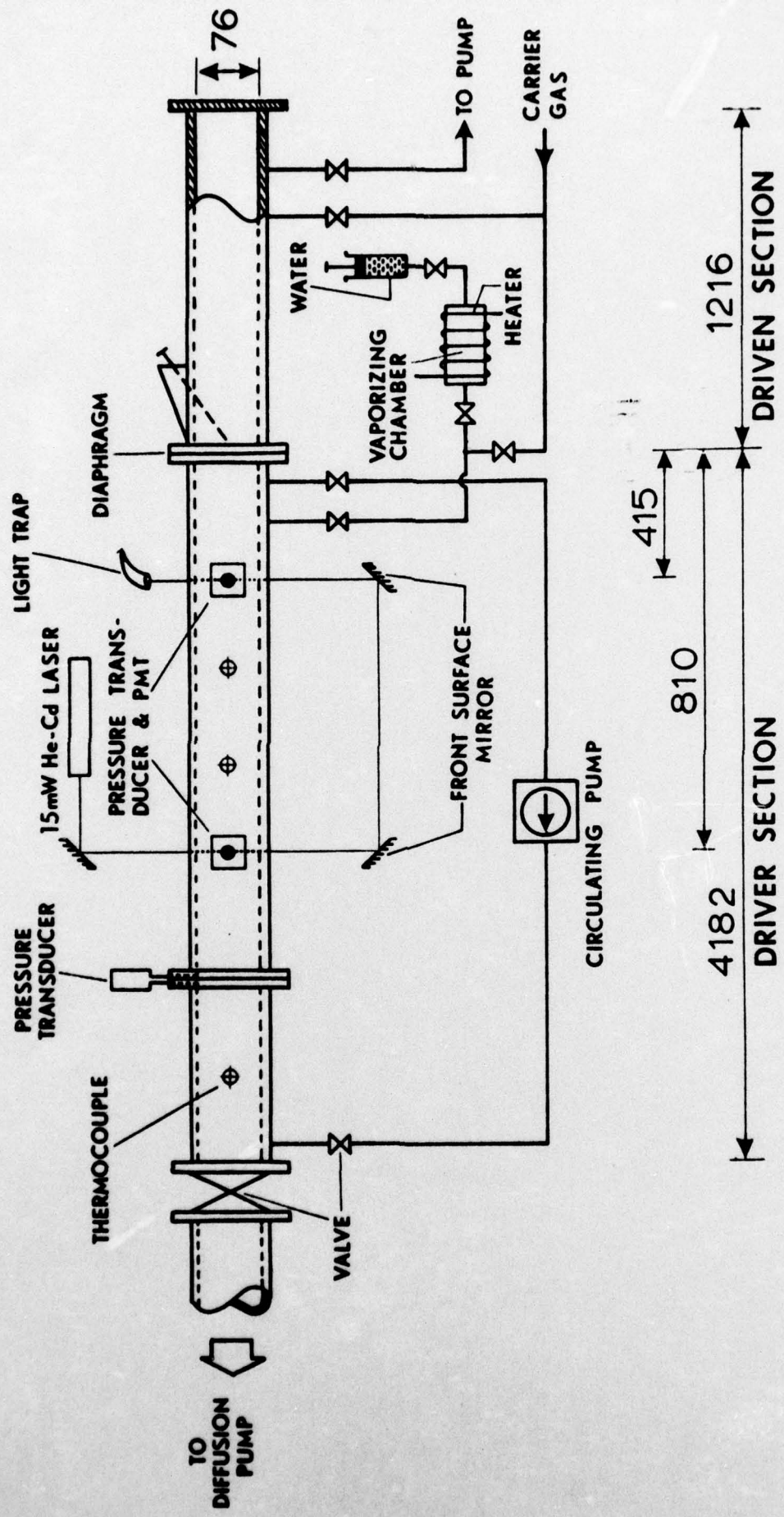


Figure 5

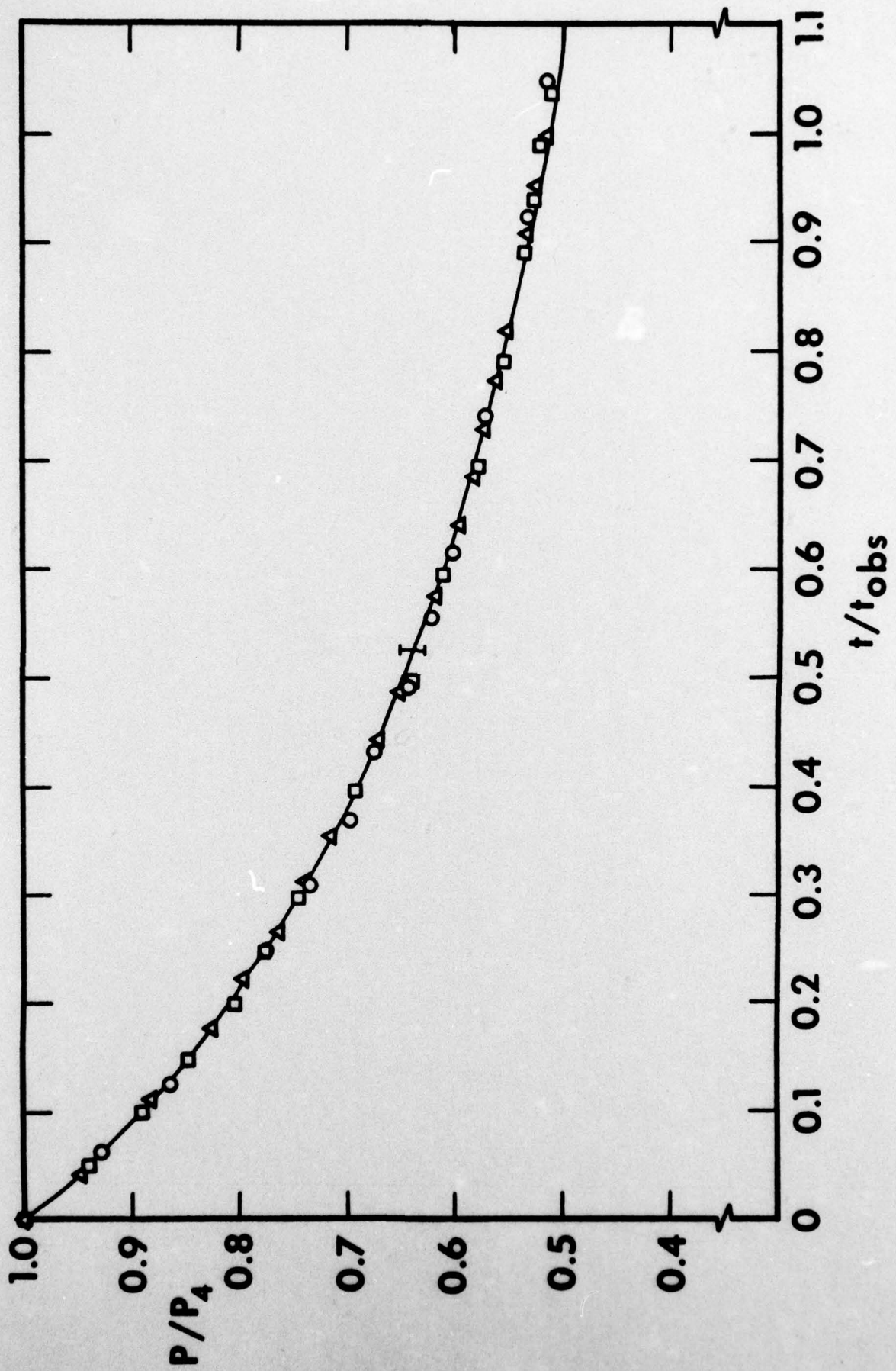


Figure 4

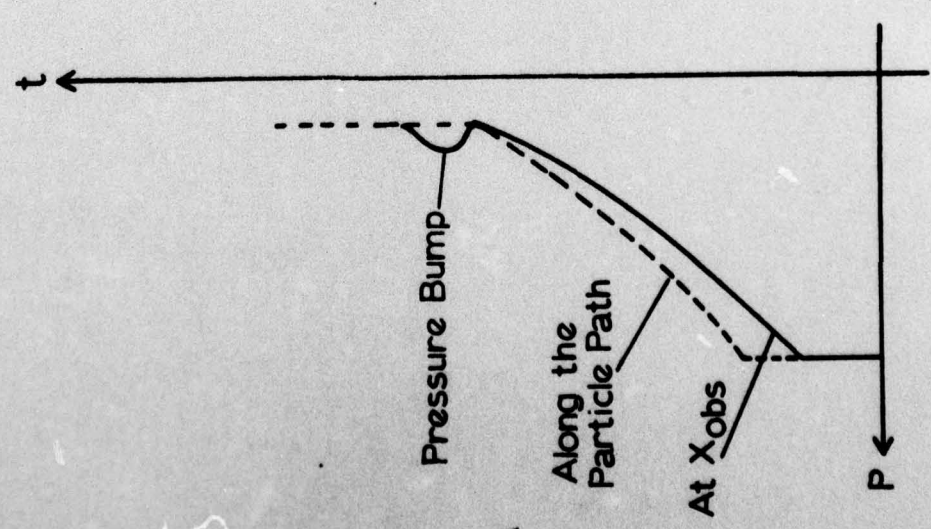
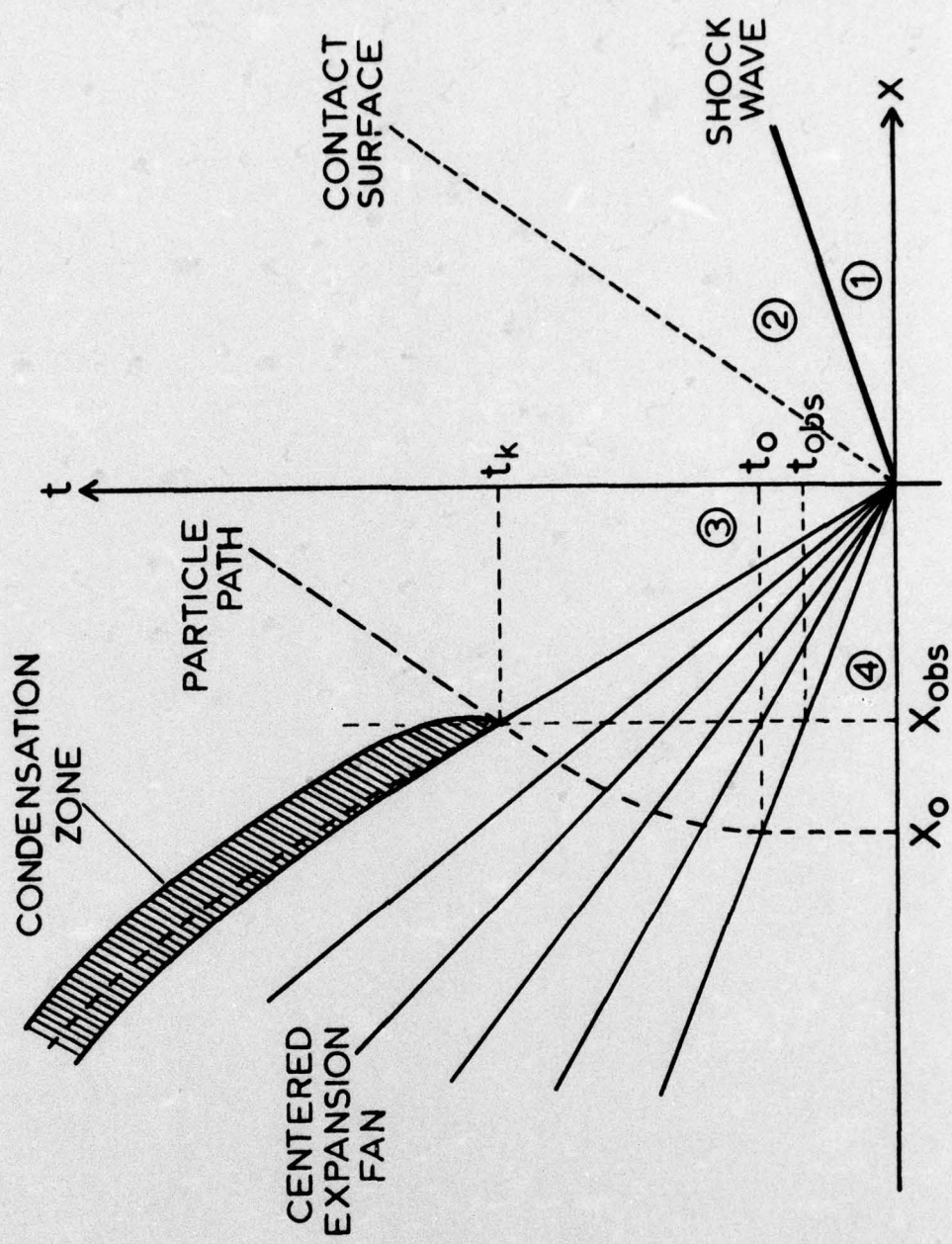
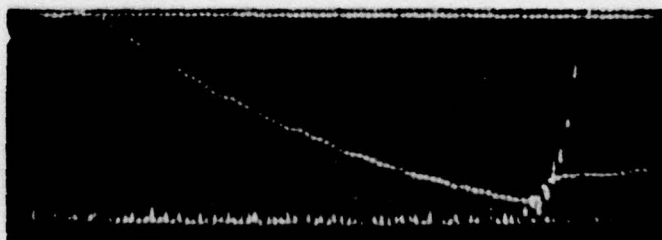


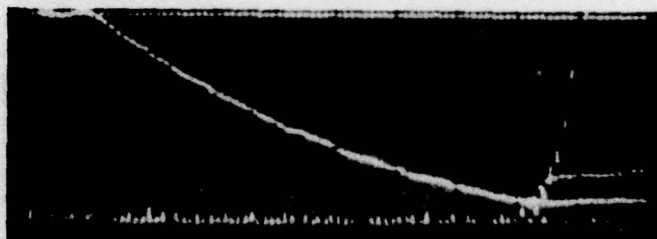
Figure 6

Condensation of Water in Argon



Superposed with Expansion in Pure Argon

Pressure
↑
155 Torr/div



0.2 ms/div → Time

EXP 73, H₂O/A, ST.2, 3/13/76

Condensing Vapor : Water

Carrier Gas : Argon

$P_4 = 1371 \text{ Torr} ; P_4 / P_1 = 2.41$

$T_4 = T_1 = 298 \text{ K} ; \omega_4 = 0.004$

Figure 7

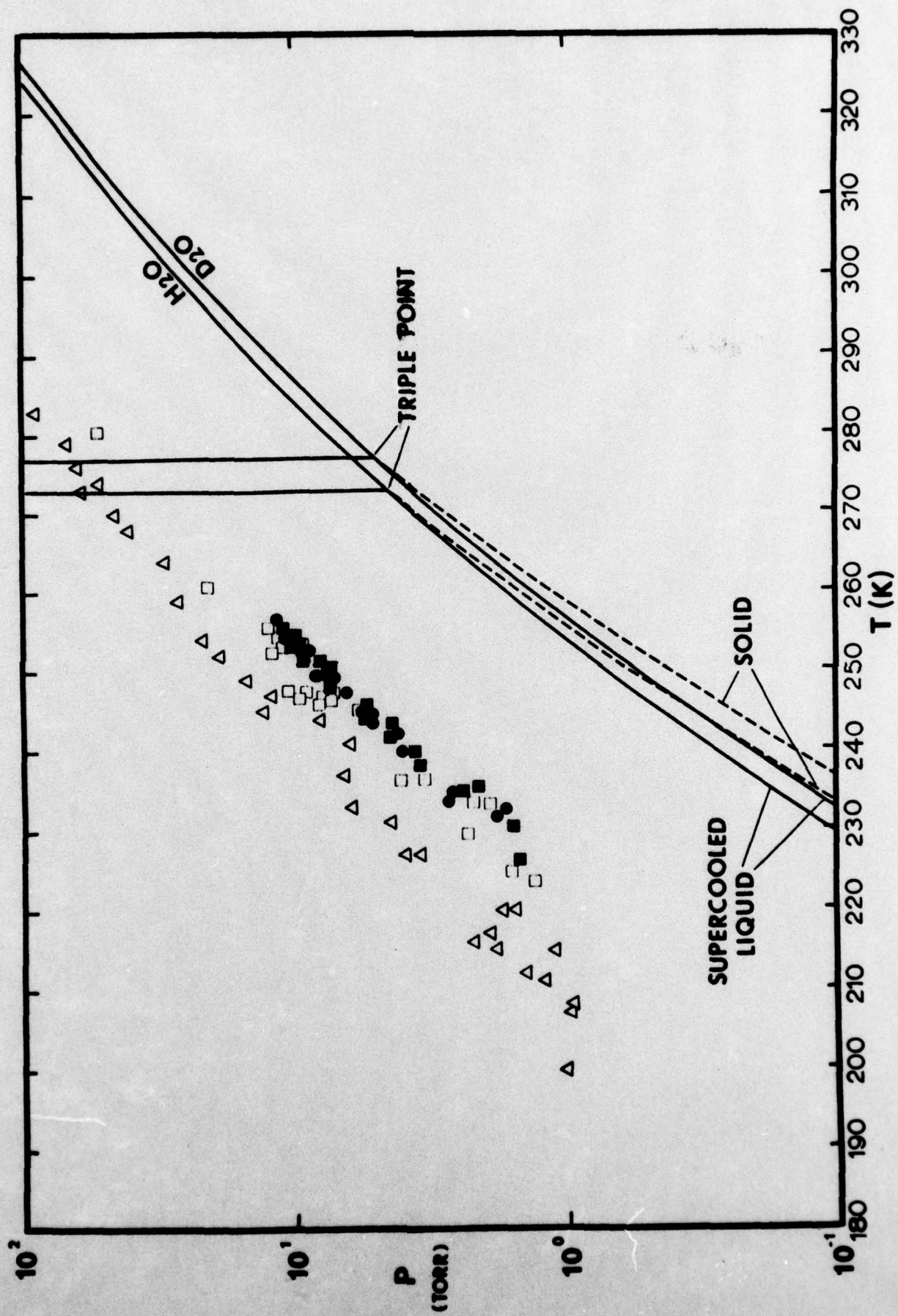


Figure 8

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) CONDENSATION OF H_2O AND D_2O IN ARGON IN THE CENTERED EXPANSION WAVE IN A SHOCK TUBE		5. TYPE OF REPORT & PERIOD COVERED
7. AUTHOR(s) C.F. Lee		6. PERFORMING ORG. REPORT NUMBER #36
8. PERFORMING ORGANIZATION NAME AND ADDRESS Department of Engineering and Applied Science Yale University New Haven, CT 06520		9. CONTRACT OR GRANT NUMBER(s) N00014-75-C-0263
11. CONTROLLING OFFICE NAME AND ADDRESS Power Program, Office of Naval Research Arlington, VA 22217		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 12. REPORT DATE January 1977 13. NUMBER OF PAGES 21
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office) 1436		15. SECURITY CLASS. (of this report) Unclassified 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release, distribution unlimited.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Flow, shock tubes, rarefaction waves, condensation, drops, growth, nucleation, water vapor, deuterium compounds.		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Despite gasdynamic non-idealities in the flow produced in a shock tube, pressure measurements at three different locations in the driver section of the shock tube revealed that the expansion wave generated in relatively weak expansions could be viewed effectively as a simple centered expansion fan after an empirical shift of the actual origin of the expansion wave to a "virtual" origin. The resulting centered (Continued on reverse side)		

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expansion fan was used to study at two locations the condensation of H_2O and D_2O vapors in an excess of the carrier gas argon, with simultaneous pressure and light scattering measurements. The isentropic flow within the centered expansion fan was found to be preserved up to the point of the detectable onset of condensation by tailoring the onset conditions to occur at the tail of the expansion fan, thus rendering a simple analysis of the experiments possible. The onset conditions of H_2O vapor were found to be in agreement with previous findings in supersonic nozzles and shock tubes, and they were well predicted by the so-called classical theory of homogeneous nucleation. The condensation of D_2O vapor was found to exhibit similar trends as those of H_2O vapor condensation despite the slight differences in physical properties between them due to isotopy.



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