NON-EQUILIBRIUM CONDENSATION OF CARBON DIOXIDE
IN SUPersonic NOZZLES

by

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ABSTRACT

Tests involving the condensation of CO₂ in three supersonic nozzles have been conducted. Data of streamwise pressure variation and local conditions at onset of condensation have been obtained and show that rate of expansion, as determined by nozzle geometry, has a significant influence on the supersaturation obtained prior to condensation.

Interferometric density measurements for non-condensing flow verified the applicability of 1-dimensional gas dynamics to the expansions, and the need for consideration of departure from perfect gas behavior of CO₂. Non-condensing density-pressure measurements and total pressure measurements substantiated the use of the Plank Equation of State in the low temperature region of testing.

Classical nucleation and drop growth theory, as applied in this study to nozzle flow, will predict the conditions of supersaturation at onset of condensation reasonably well, provided small corrections to the flat film liquid surface tension of CO₂ are applied. These corrections amount to increases of from 17% to 38%, depending upon values of thermal and mass accommodation coefficients which are also applied.

The applied theory does not successfully predict the experimental streamwise variation in pressure caused by condensation, nor the extent of effects of nozzle geometry noted experimentally. These failings are thought to indicate inadequacies in drop growth theory, or in its application.

Thesis Supervisor: Philip G. Hill
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BIographies NOTES

The author was born February 18, 1936 in Fort Worth, Texas and received his high school diploma from Edmonds High School, Edmonds, Washington in June, 1953. He entered M.I.T. that fall and received his Bachelor of Science Degree in Mechanical Engineering in June 1957. After graduating from M.I.T., he entered Naval Officer's Candidate School at Newport, Rhode Island in September, 1957 and was commissioned in January, 1958. He served for three years aboard the U.S.S. Brown (DD-546) in the Pacific Fleet, and obtained the positions of Engineering Officer and Senior Watch Officer prior to completing his duty aboard. In June, 1961 he returned to M.I.T. under the Navy's post-graduate school program and in September, 1964 obtained a Master of Science Degree in Naval Architecture and Marine Engineering.

The author is a member of Pi Tau Sigma, honorary Mechanical Engineering Fraternity, and the American Society of Naval Engineers. In June, 1964 he received the Brand Award from the Society for having stood first in his class, course XIII-A, the post-graduate curricula entered into in 1961.

He is married to the former Gretchen M. Raddatz of La Jolla, California, and presently has one daughter, age one year.
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The programmed computations were carried out with the assistance of the M. I. T. Computation Center's IBM 7094 digital computer, with many valuable contributions by Mrs. Joan Kukolich.
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NOMENCLATURE

A Nozzle Flow Area
Cp Specific Heat at Constant Pressure
Cv Specific Heat at Constant Volume
g Number of Molecules in a Droplet Cluster
g* Number of Molecules in a "Critical" Cluster
h Specific Enthalpy
hfg Specific Enthalpy of Vaporization
H Total Enthalpy
J Number of Nuclei Produced per Unit Time per Unit Volume
k Boltzman Constant
m Molecular Weight
m' "Effective" Molecular Weight
. m Mass Flux, Particles/Unit Area-sec.
M Mach Number
n Number of Vapor Molecules/Unit Volume
ni Index of Refraction of a Vapor in State i
N(g) Equilibrium Size Density Distribution of g-molecule Clusters
P, p Static Pressure
P o Stagnation Pressure
P' o Stagnation Pressure behind a Normal Shock
Pm>Pm Flat Film Equilibrium Vapor Pressure
PD Droplet Equilibrium Vapor Pressure
Q Heat
r Droplet Radius
r* Helmholtz "Critical" Radius
R Gas Constant
R' Effective Gas Constant
s  Entropy
S  Fringe Shift, Number of Fringes
t  Time
T  Static Temperature
T_o  Stagnation Temperature
T_D  Drop Temperature
T_R  Temperature of Molecules Reflected from a Droplet
u  Streamwise Velocity
U_{fd}  Internal Energy of a Droplet
v  Specific Volume
V  Total Volume
x  Streamwise Variation of Distance in Nozzle
y  Vertical Cross-Stream Variation of Distance in Nozzle
z  Horizontal Cross-Stream Variation of Distance in Nozzle

\alpha  \text{ Thermal Accommodation Coefficient}
\gamma  \text{ Ratio of Specific Heats}
\gamma'  \text{ Effective Ratio of Specific Heats}
\delta  \text{ Boundary Layer Thickness}
\epsilon  \text{ Defined in Appendix B, page B2.}
\eta  \text{ Quasi-Equilibrium Droplet Density Distribution}
\theta  \text{ Angle, Radians}
\lambda  \frac{h_{fg}}{C_T}
\lambda_o  \text{ Wave Length of Interferometer Light Source - 5461 \text{ a}}
\mu  \text{ Mass Fraction of Condensed Moisture}
\xi  \text{ Condensation (Mass Accommodation) Coefficient}
\rho  \text{ Local Density}
\rho_o  \text{ Stagnation Density}
\( \rho_L \)  Droplet (Cluster) Density

\( \sigma \)  Drop Surface Tension

\( \sigma_\infty \)  Liquid Flat Film Surface Tension

\( \tau \)  \( \frac{1}{T} \)

\( \phi \)  Free Energy per Molecule (Subscripts; \( L = \text{Liquid} \), \( V = \text{Vapor} \))

\( \phi \)  Total Free Energy of \( g \) Molecules

\( z \)  Distance \( z \) Between Optical Glass Side Walls, Nozzle I.
I. INTRODUCTION

In recent years, increasing attention has been given to the problem of homogeneous nucleation and condensation of supersaturated vapors.

Particular interest has arisen in connection with high temperature, low vapor pressure fluids, capable of high Carnot efficiencies in power plant application. The possible space application of systems employing alkali metals, and recent reports of experiments involving the flow of sodium and potassium through nozzles\(^4\),\(^5\),\(^4\) underlines the need to understand the condensation behavior of these gases well enough to provide a basis for optimum turbine and cycle design.

Early investigations in cloud chambers showed that, after initial expansions and settling had removed dust particles and other nuclei, subsequent expansions into the saturated region would result in the spontaneous formation of a condensation cloud at some limiting expansion ratio. In the absence of other nucleation sites, it was concluded that such clouds were the result of homogeneous nucleation of the super-saturated vapor. Similar condensation occurs in vapors expanded in nozzles, where rates of expansion are generally much more rapid than those of cloud chambers.

Despite extensive literature on the subject, there exists little agreement as to the correct formulation of nucleation and drop growth rates, and within the theoretical framework there exist many other uncertainties regarding the fluid properties pertinent to numerical evaluation. Experimental work is relatively limited in terms of range of test conditions, and numbers of fluids tested. Although a variety of gases have been studied in cloud chamber expansions, applicable and informative studies of nozzle expansions are somewhat sparse outside of steam and airborne water vapor for which both cloud chamber and nozzle data is extensive.\(^7\),\(^62\),\(^63\),\(^66\) & \(^67\) This data as well as the cloud chamber data for several other vapors tends to substantiate
so-called "classical" nucleation theory, as opposed to revised models of nucleation theory offered by more recent authors, as a better means of predicting condensation behavior. However, there is doubt regarding the extent to which cloud chamber data may be applied to the determination of nucleation rates, and too little data outside of water vapor has been obtained for flow in nozzles to allow a comprehensive test of theory over a variety of gases.

The purpose of this work has been to extend the limits of existing experimental data involving condensation in nozzles, through experimentation with CO₂, to study by means of a computational program the predictions of existing theory and make some judgments regarding the importance of the various physical parameters and uncertainties pertaining thereto, and to reduce these uncertainties to a point where a valid comparison of existing theory and experimental results can be made for CO₂.
II. **NOZZLE FLOW PHENOMENA**

Converging-diverging nozzles lend themselves particularly nicely to the study of condensation behavior of gases. Following are some of the aspects of gas behavior in nozzle flow and features which recommend it as a means of experimental study of condensation.

A. **Isentropic Effects**

It is general knowledge that for sufficiently high pressure ratios, an ideal gas will expand isentropically in a converging-diverging nozzle, becoming sonic at the point of minimum flow area and supersonic downstream, with continuously increasing velocities and decreasing pressures, densities and temperatures all along the expansion. A non-ideal gas behaves similarly, and depending upon the stagnation conditions, the extent of the expansion, and the properties of the vapor being tested, the isentrope may approach saturation conditions of pressure and temperature. As illustrated on the T-S diagram of Figure 1, it is possible for conditions to exist for certain gases when the opposite may be true and an isentropic expansion will result only in increasing the degree of superheat of the vapor (i.e. as with many of the FREONS). However, in most cases it is a simple matter to devise stagnation conditions which result in the rapid arrival of the gas to conditions of supersaturation during its isentropic expansion. For a supersonic nozzle a few inches in length expansion time is of the order $10^{-4}$ seconds.

B. **Wall and Boundary Layer Effects**

In all real gases there are departures from ideal non-viscous behavior which must be considered. Fortunately, in nozzle flow these effects are easily accounted for, provided fully developed flow does not occur. Viscous effects are accounted for by considering the nozzle flow to be an isentropic expansion contained between wall boundary layers. The boundary layers are greatly influenced by the shape of the pressure gradient and are generally
depressed to a very small minimum value near the most critical point, the
geomeric throat. Further, viscous and recovery effects on the nozzle walls
will keep the wall temperature close to stagnation value in the absence of
very strong wall cooling or heating, and, at decreased downstream pressures,
is adequate guarantee that there will be no condensation of liquid on the
walls which could interfere with the mid-stream process.

In experimentation where nozzle size might be limited by mass flow or
other restrictions, considerations of Reynold's Numbers and boundary layer
thicknesses which may be expected in the throat region are important since
large boundary layers in relation to nozzle size could invalidate the
assumption of an isentropic region of flow. A means of estimating the
boundary layer thickness in the throat region of a supersonic nozzle was
devised (Appendix E) and showed good agreement with interferometric photos
that the boundary layers involved in the CO₂ experiments of this investiga-
tion were about .006" at the nozzle throat. In all nozzles tested this was
less than 11% of the throat width. Additional discussion of boundary layer
treatment in this investigation is included later.

The isentropic region between boundary layers may be described by one-
dimensional gas dynamics, and, by means of measurement of some vapor
property (such as static pressure) along the nozzle, a complete time-
history of the expansion may be obtained.

Condensation in a nozzle expansion has been shown by many investiga-
tions to have a large influence on the thermodynamic properties of the
fluid. In particular, the static pressure records large variation from the
non-condensing profile as a result of the heat released by moisture forma-
tion. For the addition of heat to an ideal gas, it may be shown that

\[
\frac{dP}{P} = \frac{\gamma M^2}{1 - M^2} \left[ \frac{dA}{A} - \frac{dQ}{CT} \right]
\]
For water vapor, it is easily estimated that at $M = 2$, a formation of 1% moisture at constant area is accompanied by about 7% variation in static pressure. As is later shown, it is possible to detect the onset of condensation at about .1% presence of moisture through simple static pressure measurements.

It may be seen from the above equations that pressure variation is increased by establishing conditions such that condensation occurs with Mach Number close to unity.

Willmarth and Nagamatsu, $^{64}$ and Arthur, $^3$ have shown that the ratio $P/P^'$ plotted against the streamwise variation of $P^'/P_0$ is an even more sensitive indicator of condensation, where $P$ is the measured static pressure, $P^'$ is the measured total pressure behind a normal shock, and $P_0$ is the original stagnation pressure.

An important consideration in any system designed to test homogeneous nucleation theory is the possible effects of contamination. As will be shown later, nozzle expansions from superheated to supersaturated state proceed in such an extremely short time that spontaneous nucleation will produce nuclei many orders of magnitude greater in number than those which could be present in the form of dust or other particles and the amount of moisture formed on such contaminants is entirely negligible. As will also be mentioned later, an exception to this is the case of a pre-condensing vapor.

The proceeding observations indicate that, with fairly simple instrumentation of a nozzle system, it is possible to obtain a complete history of a gas expansion and any condensation occurring within, with the reasonable assumption that no outside influences exist which would invalidate testing of homogeneous nucleation and drop growth theory in the same system.

C. Test Vapor Considerations

The selection of a vapor for testing is subject to the following
considerations. First, a large number of vapors are toxic, reactive to instrumentation materials, or do not exist as vapors at conditions most convenient for laboratory testing. Second, the state relations and other properties of many are so poorly defined, that application to any meaningful test of theory would be ill-founded. Neither of these considerations would be restrictive if the condensation data for a particular vapor were particularly needed, or if a test of theory were incidental to the experimental data. Third, two particular properties are desirable in a test vapor, both of which are more common to gases of low molecular weight. The first has been mentioned previously, and involves the shape of the coexistence line in relation to the isentrope. A preferred test gas has a coexistence line having rapid convergence with the isentrope. Also, the measure of heat release resulting from condensation compared with that needed to alter its vapor properties is important. This may be expressed through the parameter $h_{fg}/C_p$, for which typical values for various gases are presented in Table 1. Variations in the gas properties caused by condensation are increased with larger values of this parameter. A more applicable use of these values is, as seen from Equation 1, the form $h_{fg}/C_p T$ at the test conditions of interest. For vapors such as alkali metals, higher values of $h_{fg}/C_p$ are decreased in significance due to the high temperatures of the gas.

$CO_2$ was selected for this study because it satisfies all the foregoing criteria and in addition may be obtained in high purity at relative low cost, thus allowing its use in a system not necessitating recovery after expansion. Also, such physical properties as $CO_2$ vapor pressure, liquid surface tension, and solid and liquid density, necessary in the application of theory, are well established and readily available in numerous publications of physical property data.
Discussion of the application of these properties, and details of the theoretical mechanism and behavior of condensation are presented later.

The question could be raised concerning possible non-equilibrium of energy modes of CO₂ during such a rapid expansion. Kantrowitz has shown that this would not be a problem with CO₂ in this case since 1)vibrational energy modes are not activated at the low temperatures of experimental test conditions and 2)the relaxation time constant of vibration is considerably less (~10⁻⁶ sec.) than that of the gas expansion. Camac has recently formulated the pressure-temperature dependence of CO₂ vibrational relaxation time and his expression also yields times of the order 10⁻⁶ sec. in the region of experimental testing.
III. EXPERIMENTAL INVESTIGATIONS

A. Test Apparatus

Industrial Welding CO₂ having very high (moisture content less than 0.0045% by weight) purity was introduced via a manifold from a bank of nine 50 lb cylinders to a needle valve used for regulation of flow rate (See Figure 2). Thence the flow entered a stagnation tank and then the test nozzle. Exhaust was to atmosphere in earlier tests and later to a steam ejector system maintaining about 23" Hg. of vacuum. This modification facilitated lower minimum stagnation pressures without the interference of compression shock systems in the supersonic test section. Nine cylinders were employed in order to obtain a large mass flow source providing nearly constant conditions to the nozzles for extended periods of flow, despite the quasi-steady nature of the "blow-down" method.

The stagnation tank contained a calibrated pressure gauge and, extending into the flow just upstream of the nozzle entrance, a copper-constantan thermocouple using an ice reference. A 100 inch 25-tube mercury filled manometer board was used in connection with static wall taps to determine pressure readings along the nozzle. Pressure data was recorded by polaroid photograph of the manometer board. Test time durations were of the order of 1 minute.

The stagnation tank had a removable end plate and contained screens which allowed the packing of large amounts of steel wool inside a nylon filter bag upstream of the nozzle entrance. (Figure 5) When high stagnation temperatures were desired (i.e., non-condensing tests) an electric heating tape wrapped around the stagnation tank was energized and the steel wool used for heat storage and thermal inertia to decrease the rate of the temperature transient. When low temperatures were desired, the steel wool was removed and the heating tape not used.
B. Test Nozzles

Two test nozzles were employed for experimental measurements, henceforth referred to as Nozzle I and Nozzle II.

Nozzle I appears schematically in Figure 7 and in photos in Figures 6 and 9. It featured two removable parallel side walls which were plates of optical glass, used for interferometric measurements. By exchanging one of the plates for an identical metal one containing pressure taps, static pressure profiles could be obtained and the flow observed simultaneously through the remaining glass walls. Slotted "O-rings" provided sealing of the side walls. The nozzle was made wide in comparison to its height in order to minimize any contribution to error caused by boundary layer effects of the side walls on the interferometric measurements. This error was later estimated to be less than .2% of the measured densities.

The nozzle shape was also intended to provide nearly 2-dimensional flow, and in fact, interferometric photos to be discussed later show the flow to be effectively 1-dimensional between boundary layers. A region of isentropic flow was thus obtained between the boundary layers, which were of thickness ± .005" - .007" at the throat (Appendix E).

Pressure Taps in Nozzle I were spaced .2" apart, and later during the program, additional taps, resulting in .1" spacing, were added in the test section region.

Nozzle II is illustrated in Figures 8 and 10 and featured hinged upper and lower nozzle walls which could be placed at arbitrary angles between the fixed parallel side walls. This facilitated variable expansion rates in the test section, depending on the divergence angle of the test section, and it was hoped to thus establish a "best" angle of divergence to obtain the most informative pressure profiles. Since pressure variations due to heat addition approach maximum when Mach
Number approaches 1.00, it was expected that this result would be obtained for a very low angle of divergence.

Static pressure taps located in the side wall were located .25 inches apart for early runs, after which a new test plate with taps .1 inches apart in the test region was substituted.

C. **Experimental Procedures**

1. **Pressure-Density Measurements**

Tests were conducted using two persons and the following procedure.

1) The needle valve was opened until the approximate value of desired stagnation pressure was obtained.

2) One observer maintained a null-balance on the thermocouple-potentiometer circuit, while the other, manning a tripod-mounted polaroid camera, observed the manometer board.

3) When the manometer board reached a steady state, a polaroid photo was taken of the manometer board by the second observer while the first recorded the stagnation gauge pressure and terminated the null-balancing procedure on the thermocouple potentiometer.

4) The reading of the potentiometer at time of photograph was converted to a stagnation temperature reading, and the polaroid display of the pressure data developed, and all pertinent data involving the test run affixed to it (including local barometric pressure).

Interferometer runs were made in a similar manner, except for the use of 5" x 7" sheet film in the built-in camera of the interferometer, and requiring only the potentiometer null-balance prior to recording data.

Discussion of interferometry theory and the methods applied in this investigation plus related references are presented in Appendix G.
2. **Stagnation Pressure Measurements**

The stagnation pressure behind a normal shock was measured by means of a .030" O.D. pitot tube extending upstream from a micrometer mounted on the elbow downstream of the nozzle. (See Figure 6) All measurements were made on Nozzle I, by visually aligning (with the aid of lighting, and machinist's templates) the tip of the tube with the center of the corresponding wall tap with which static pressure was measured. Since the location of the shock was actually located a small distance upstream, a correction was made to the static pressure reading corresponding to this distance, (about .63 tube diameters at Mach 1.65 and about .30 tube diameters at Mach 2.45, decreasing further as Mach Number increases.)

Upstream stagnation pressures were measured with the calibrated gauge in the stagnation tank. As in the case of the static pressure profile measurements, photographs of the manometer board were taken simultaneously with stagnation temperature and pressure measurements, after steady pressure readings and a null potentiometer reading were obtained.

Total pressure measurements were made with all manometer tubes disconnected except the following 1) pitot tube 2) static wall tap at tip of pitot tube 3) throat static wall tap (recorded only for supplementary purposes).

Discussion and estimates of possible experimental error are contained in Appendix I.

D. **Non-Condensing Measurements**

1. **Real Gas Behavior**

It became necessary to consider the departure from perfect gas behavior of CO\textsubscript{2} near the very outset of this investigation, since one of the objects of experimental work was to compare the agreement of measured density profiles with measured pressure profiles. Failure to consider the proper
p-v-T relation for reduction of interferometric data resulted in considerable error in the density profile. It also introduced doubt as to how the proper density ratios should be converted to the corresponding isentropic pressure ratios for purposes of making comparison between measurements.

Existing tabulated properties of CO$_2$ also showed that the degree of departure from perfect gas behavior for CO$_2$ (as reflected by the compressibility factor, $z = \frac{P_v}{RT}$) increased markedly near the saturated region, and that the values of compressibility factor could easily be as low as .9 during an expansion. Also, existing tables did not extend to pressures and temperatures low enough to make adequate determination of what the real gas behavior of CO$_2$ is.

Since a meaningful test of the nucleation and drop growth predictions which were to follow must rest on the most precisely accurate information available regarding the gas dynamics, and since, as will be shown, the predictions are quite sensitive to errors in this respect, it was decided to establish the real gas behavior of CO$_2$ through use of an applicable equation of state.

Unfortunately, though the literature abounds with work, much of it recent, on high pressure and temperature properties of CO$_2$, it was possible to find only one equation of state that is applicable in the lower P-T range of these experiments, that being Plank's (1929), which fits the data of Lowry and Erikson, and Maass and Mennie very well. Comparison of Plank's Equation of State and of their data is shown in Figure 14, showing maximum disagreement of about .1% of $\frac{P_v}{RT}$. For comparison, the equation of State of Martin & Hoa, and Benedict, Webb, and Rubin applied well outside their applicable region to the area of experimental interest, vary by as much as 10% from Plank's.
Plank's equation is stated and was applied as follows:
\[ v = \frac{RT}{p} + \frac{(0.0825 + 0.0012657p) \times 10^3}{(0.01T)^3} \] (2)

Using the relations:
\[ \frac{\partial h}{\partial p} = v = \frac{1}{\rho} \] (3)
\[ dh = (\frac{\partial h}{\partial T})p dT + (\frac{\partial h}{\partial p})_T dp \] (4a)
\[ = C_p \frac{dT}{\rho} + (\frac{\partial (vl)}{\partial T})_p dp \] (4b)

where \( \tau = \frac{1}{T} \), it is possible to use the zero pressure value \( C_p \) for the result
\[ h = \int_0^T C_p dT + \int_0^p [\frac{\partial (vl)}{\partial T}]_p dp \] (4c)

The right hand term may be evaluated from the equation of state and hence it is possible to determine enthalpy at any desired pressure and temperature. To determine an isentrope, equation (3) is used and the change in \( h \) calculated for an arbitrary change in \( p \). Thence, knowing new values of \( p \) and \( h \), the corresponding value of \( T \) may be determined, either through the use of pre-calculated enthalpy tables, or by iterative integration of equation (4). It is then a simple matter to calculate the new value of density from equation (2) and to thus determine a series of pressure, temperature, and density ratios along an isentrope for any given set of stagnation conditions.

Table 2 contains values of enthalpies calculated in this manner. Agreement within .1° to .3°K occurs at the juncture of those presented by Din.15

In addition to the comparison of pressure and density measurements, it was decided to apply an additional experimental test to the ability of the Plank Equation of State to predict the behavior of \( \text{CO}_2 \) gas dynamics in regions of low pressure and temperature yet below its known region of applicability, (0°C to -70°C). Measurement of the normal shock pressure
characteristics were made and compared with the predictions of "real" gas dynamics. For this purpose, it was necessary to include a computation scheme for solution of the continuity, momentum, energy, and state equations across a normal shock and to return along a new isentrope to the new stagnation conditions. A listing of this computation procedure is given in Appendix H.

2. Pressure-Density Measurements

Photographs of two interferometer fringe patterns for non-condensing flow are seen in Figures 11 and 12 and the resulting densities given in Table 3 and 4. The photographs are marked by an extended region of near verticle interference fringes between the upper and lower nozzle walls, indicating that this region is in 1-dimensional flow, and that the boundary layers are quite small (approximately .006" at the throat).

Results of the two sets of measured density and the corresponding pressure profiles of Nozzle I are shown in Figures 15a, 15b and 16a, 16b. Each comparison has been illustrated in two ways. Figures 15a and 16a show the density and pressure profiles in relation to nozzle position. Pressures have been directly measured and plotted at their tap locations. Measured density ratios have been converted to their corresponding pressure ratios through use of the real gas isentropic values based on the experimental stagnation conditions.

Figures 15b and 16b show the same data plotted in somewhat different form, illustrating the theoretical isentropic values of $P/P_o$ vs. $\rho/\rho_o$ and those values of $P/P_o$ and $\rho/\rho_o$ which were measured at identical points in the nozzle. On Figure 16b, the corresponding perfect gas conversion from density to pressure (based on zero pressure value of $C_p/C_v$) is also shown.
Figures 15 and 16 show very good agreement between measured pressures and densities, and those predicted from the Plank Equation of State. The fact that the conversion of density to pressure is not very significantly affected by the use of the equation of state rather than perfect gas relations, is indicated by the close proximity of the perfect gas line in Figure 16b, where the applied ratio of specific heats is the zero pressure value at stagnation temperature, $\gamma = 1.28$. However, it should be emphasized that even this use of the perfect gas approximations is preceded by the use of real gas p-v-T relations in the reduction of interferometric fringe shift data to obtain values of density. The only noticeable departure from exact real gas agreement consistent to both tests occurs near the throat region where the measured pressures are slightly higher ($= 1.8''$ Hg) than those values based on measured density. This departure is concluded to be due to departure from 1-dimensional flow over the boundary layer of the lateral side walls in this region and an approximate numerical estimate of this effect in good agreement with the noted error is made in Appendix F.

Attempts to apply the interferometer to direct study of condensation were frustrated by increasing fluctuation and blurring of the interference fringes as stagnation pressure was increased. This effect was apparently due to thermal turbulence. Above pressures of 85 psia, complete blurring resulted upstream of the throat, although the interference bands again became visible downstream, where acceleration of the flow dampens the effects of turbulence. Consequently, no density profiles involving condensation were obtained although photographs of the condensation ice cloud showing the interference fringes within were obtained.

3. Normal Shock Total Pressure Measurements

A more sensitive test of the Plank Equation of State predictions versus those of perfect gas was made by applying each to the gas dynamics
of normal shock behavior and comparing experimental results with theoretical predictions based on the experimental stagnation conditions of each test.

Results of experimental normal shock measurements are shown in Figure 17, using the display of Willmarth and Nagamatsu, and Arthur, who made similar measurements for Nitrogen and Helium. The predictions computed with Plank's Equation of State are shown in Figure 18. Test series No. 2 was taken with a great deal of regard for better accuracy in pitot tube positioning and establishment of steady state readings prior to taking data, with a resulting decrease in the amount of scatter. One-dimensional perfect gas predictions are shown for reference in the lines of constant $\gamma$, the appropriate zero pressure value for most of the test conditions being about $\gamma = 1.28$. It is seen that the real gas predictions are considerably more accurate than those of perfect gas in predicting the normal shock characteristics of CO$_2$, although there still exists some departure of the experimental results from those predicted with the Plank Equation of State. The experimental results indicate behavior very close to that of a perfect gas having a value of $\gamma = 1.40$. However, this is misleading, since treatment of CO$_2$ as a perfect gas, using this value of ratio of specific heats, does not successfully correlate the pressure-density measurements, or correctly predict local temperatures along an isentrope. In terms of the measured properties, the experiment indicated about 2% larger values of stagnation pressure behind the normal shock than those predicted.

Despite the above-mentioned lack of complete agreement between Plank's Equation of State and measured normal shock pressure characteristics, it appears that such a "real gas" treatment of the gas dynamics is a considerably better description than perfect gas and so warranted application to the prediction of local conditions along the isentrope.
4. Influence of Equation of State Upon Isentropes

The influence of real gas treatment to the computation of \( \text{CO}_2 \) isentropes has quite a significant effect upon the values of local temperature calculated. Figures 19 and 20 compare two typical perfect gas isentropes with those predicted using Plank's Equation of State. These curves indicate that an error of 50K could easily occur by attempting to describe the isentrope by a constant ratio of specific heats, and that a decrease in stagnation temperature could cause errors of possibly 100K if one isentrope were applied to all experimental test conditions.

The resulting uncertainty in predicted nucleation rate, depending upon location within the supersaturated region, can be seen by referring to the classical nucleation rate profiles for \( \text{CO}_2 \) illustrated in Figure 21. Errors of 15 orders of magnitude could easily occur.

E. Condensation Measurements

1. Pressure Profiles and Local Conditions at Onset of Condensation

Results of experimental measurements involving condensation in Nozzle I, pressure tap plates 1 & 2 are shown in Figures 22 and 23. Results of Nozzle II, pressure tap plate 1 are shown in Figure 24, and of the same nozzle using pressure tap plate 2 in Figures 25 through 30. Figures 24, 25, 26, 27 and 28 are for Nozzle II at an included divergence angle of approximately 2.2° while Figures 29 & 30 are for Nozzle II at an angle of 0.80°. For convenience and due to manometer board limitations, pressure ratios are expressed in terms of the pressure at the tap nearest the throat, but may be easily converted to total pressure ratio. Data for each Figure is adjoined in Tables 5 through 13.

All experimental profiles showed a marked occurrence of pressure variation from non-condensing values at the point of occurrence of
condensation (verified visually in Nozzle I). By noting the pressure ratio at which condensation occurs, it is possible to determine the local conditions at occurrence using the isentropic relations for the equation of state previously discussed. Based on the range of applicability of Plank's Equation of State and comparison with the tables of Din,\textsuperscript{15} it is estimated that temperatures in the vicinity of 190°K are accurate to within .3°K and temperatures in the region of 160°K accurate to perhaps 1° or 2°K. A plot of these points for all three nozzle geometries tested is shown in Figure 31.

Several characteristics of the condensation profiles and the local conditions at point of occurrence may be noted.

First, the shape of the profiles contain no sharp minimum at the point of occurrence of condensation such as those noted in the condensation of pure water vapor.\textsuperscript{7,23} i.e. it is not appropriate to speak of condensation shock in this case, since the formation of moisture occurs along an extended region.

Second it appears that pressure level (or temperature level, or some dependent property) may influence the shape of the profile, i.e. the rate of formation of moisture, to some extent since the shapes of the condensation profiles appear to be more "spread out" for high stagnation pressures than for low stagnation pressures. See Figure 27. These effects are not explainable in terms of gas dynamics influences, and the cause is not apparent.

Third, a very definite influence of nozzle geometry upon the point of onset of condensation occurs, as seen in Figure 31. The rapid rate of nozzle divergence and expansion of Nozzle I results in higher supersaturation ratios and relatively greater delay of condensation. Progressively decreasing the rate of expansion through decrease of nozzle divergence
results in earlier condensation. Comparative geometry and non-condensing pressure profiles of the three nozzles may be seen in Figure 32.

Fourth, the extremely low rate of divergence of Nozzle II in the 0.80° case (subsonic flow results at about 0.50° divergence) results in notable axial fluctuation of the pressure profile data (Figure 29) and some scatter of the point of onset of occurrence of condensation. (Figure 31) It should be remembered that differential changes in gas properties are proportional to \((1/1-M^2)\) and that at the pressure ratios present in these tests Mach Number remains very close to unity. (\(M = 1.25\) at \(P/P_0 = .40\)). Also, as heat of condensation is released, Mach Number is driven even closer to unity. This is coupled with the fact that at low angles of divergence, the value of the negative pressure gradient is greatly decreased, downstream of the throat, from those values which existed for the other two test nozzles. This would tend to create more rapid boundary layer growth and greater departure from 1-dimensional flow in the narrow angle case. It might be expected that larger departures from 1-dimensional flow, plus the magnifying effects of near unity Mach Numbers could contribute to larger pressure fluctuations.

2. Visual Observations

Experimental observations indicated that the pressure hump accompanying condensation occurred at a point coincident with the appearance of the ice cloud.

This at first seemed at variance with the expectation that pressure variation should occur prior to the appearance of visible condensate, due to the theoretically small sizes of forming nuclei. (\(r \sim 10^{-7}\) cm).

Yellot's\(^{66,67}\) investigation of condensing steam was also accompanied by visual observation of the water cloud coincident with the location of onset of condensation as determined by pressure measurements. He
reported that this observation remained valid despite changes which resulted in the axial shift of condensation in the nozzle. The same was true in this investigation.

It is also mentioned that the data illustrated a very high degree of consistency and reproduceability, that illustrated in Figures 24 through 28 involving three separate sets of measurements over a period of six months, each set being separated by nozzle dismantlement, cleaning, and pressure tap plate alterations.
IV. INTERPRETATION OF EXPERIMENTAL RESULTS

As discussed in Sections I and II, there are reasons for supposing that the process of nozzle condensation is one involving homogeneous nucleation within the super-saturated vapor. The purpose of this section is to discuss homogeneous nucleation theory and the subsequent drop growth process and apply these processes to nozzle expansions for the purpose of testing the ability of existing theory to interpret the experimental results of CO\textsubscript{2} condensation. Attention is given to the physical properties upon which nucleation and drop growth theory depend, and to the uncertainties which exist in the applicable values of these properties as well as to the theory itself.

A. Nucleation Theory

A review of classical nucleation theory, following the development of Frenkel\textsuperscript{19} is presented in Appendix B. For more comprehensive review of theory see Reference Numbers 5, 6, 12, 18, 38, 56, 58, 65 and 68. The discussion of Yang,\textsuperscript{65} accomplished while working under P.P. Wegener at Yale University, is particularly thorough.

1. General Form of Nucleation Rate Equation

The rate of production of nuclei in a supersaturated vapor has been shown by various authors to be of the form\textsuperscript{*}

\[ J = A \cdot \exp\left(-\frac{\Delta G^*}{KT}\right) \]  

where \( J \) = number of nuclei produced per unit time per unit volume,

\( \Delta G^* \) = free energy of formation of a critical cluster (a cluster in unstable equilibrium with its supersaturated vapor)

And A is proportional to the incident flux of vapor molecules on a critical-sized cluster.

\[ A = C_n \left[ \frac{P}{\sqrt{2 \pi mKT}} \right] 4\pi r^2 \]  

* See Appendix B for a review of nucleation theory development
The value of $C$ varies somewhat as derived in the classical treatments of Volmer, Becker and Doring, Frenkel, Barnard, and others. Though variations of one to two orders of magnitude occur over the range of values presented, the effect on nucleation rate is small, since for only a small additional expansion along a nozzle, the value of $J$ can easily vary by 5 - 10 orders of magnitude. Since the change in local conditions is small for a variation in nucleation rate of this sort and, in fact, is scarcely noticeable for 1 to 2 order of magnitude variations in $J$, the differences in classical treatments of the theory are not significant.

In the classical treatments of the theory, the nucleus is considered as a stationary object, and the free energy of formation is merely:

$$
\Delta G = 4\pi r^2 \sigma - g k T \ln \frac{P}{P_\infty}
$$

(7)

where the first term is the work of formation of surface area for a liquid having surface tension $\sigma$, and the second is the constant-temperature free energy change of $g$ molecules which change state from vapor at $P, T$ to saturated liquid at $T$. $P_\infty$ is the flat film vapor pressure of the liquid at temperature $T$.

The critical cluster(*) is that for which the function $\Delta G$ is a maximum, and results in

$$
\ln \frac{P}{P_\infty} = \frac{2\sigma}{\rho_L KT r^*}
$$

(8)

Using the procedure presented by Frenkel, for which the value of $c$ becomes

$$
C = \left[ \frac{\ln P}{6\pi g^*} \right]^{1/2}
$$

(9)

the expression for nucleation rate is

$$
J = \left( \frac{P}{K T} \right)^2 \frac{m}{\rho_L} \left( \frac{2\sigma}{\rho_m} \right)^{1/2} \exp \left[ -\frac{\ln g^*}{3kT} \right]
$$

(10)
As mentioned, Yang has shown that the results of the authors previously mentioned yield expressions for classical nucleation that are much alike (in some cases identical) and which yield quantitative predictions that are not significantly different.

More recent authors have pointed out the need for inclusion of additional terms to the free energy of formation of a critical cluster, which account for the fact that the cluster may have translational, rotational and vibrational energy modes. Kuhrt, Rodebush \textsuperscript{53} Lothe and Pound, \textsuperscript{38} and Courtney \textsuperscript{12,13} have treated this problem but reach widely different conclusions as to the effect upon nucleation rate. The postulated correction factors for this "gassification" of the condensed clusters range from $10^4$ to $10^{20}$, depending on the treatment and molecular configuration chosen.

More recently, Feder, Russell, Lothe and Pound\textsuperscript{18} have jointly made a very comprehensive review of this problem, and offered some new thoughts on the subject. For water vapor, they conclude that the effects of gassification are equivalent to a correction factor of about $10^{15}$, but admit that this is not borne out by cloud chamber data. In general, such experimental data as does exist tends to more strongly verify classical theory\textsuperscript{5,23} than the improved gassified theory, despite strong arguments for the greater theoretical validity of the latter.

2. Evaluation of Classical Nucleation Rate Equation

Essentially the same physical properties appear both in the exponential and pre-exponential terms of the nucleation rate equation. Of course, uncertainties in these properties will effect the calculated value of nucleation rate chiefly through the exponential. By substitution for the value of $r^*$ it is seen that the exponential is

$$\exp\frac{-4\pi r^*}{3kT} = \exp\left\{ \frac{-4\pi}{3} \left( \frac{2m}{\rho_L\ln P/P_w} \right)^2 \left( \frac{\sigma}{kT} \right)^3 \right\}$$

(11)
In evaluating this term correctly it is clear that four physical properties of the vapor must be known: 1) the liquid drop surface tension 2) local temperature 3) liquid drop density and 4) local supersaturation ratio.

Much attention has been given to the applicable values of surface energy for small droplets. However, here again there is little agreement as to what corrections should be made to flat film values to take account of the effect of droplet curvature, and the sign of the correction is itself a subject of controversy.

Effects of curvature are considered by some to cause a surface tension decreasing with drop radius such as in the Kirkwood-Buff Equation.

$$\sigma = \frac{\sigma_\infty}{(1 + 2\delta/r)}$$

(12)

where $\delta$ is a length, nearly constant, lying between .25 and 0.6 of the molecular radius.

In addition, Stever and Rathbun have calculated a correction to take account of the radius dependency of surface tension which also results in a decrease for small radii. Compared with uncorrected surface tension these corrections lead to lower energies of formation of nuclei and hence higher nucleation rates for the same supersaturated conditions.

However, Orian and Sundquist predict an increase in surface energy of about 25% in the case of water, using an analysis based on the breaking of intermolecular bonds. A bonding model used by Benson and Shuttleworth, however, predicts a 15% reduction in surface energy of correspondingly small water ice crystals. Bogdonoff and Lees, also using considerations of bonding energies of sodium and of nitrogen obtained a correction that results in larger critical clusters and also larger energies of formation, hence lower nucleation rates from the uncorrected case. For lack of a more
definite indication of the effects of small droplet sizes, most investigators use the flat film value of surface energy, and estimate the effects on the theoretical predictions of departure from it.

An additional uncertainty in surface energy occurs with CO$_2$ due to the fact that, at pressures below its ice point, 75.1 psia, it apparently condenses in the form of ice crystals rather than liquid droplets. Bondi$^9$ has shown that for metal crystals, which involve valence bonding, increases in surface energy from the liquid value are closely proportional to the heat of fusion and, depending also on the crystal planes involved, amount to increases of about 25% for those metals which he discusses. He further presumes that for molecular solids one might expect increases of from approximately 1/8 to 1/3 these amounts.

In the case of some gases there is doubt as to the proper value of droplet density to be applied. Though density does not vary too rapidly with temperature, there is doubt, in the case of CO$_2$, as to whether solid or liquid density should be used. Although the CO$_2$ may condense as ice particles, there is some argument for use of liquid density in order to be consistent with the spherical geometry and liquid surface energy applied to the liquid drop model, and in the absence of any knowledge of CO$_2$ crystal surface energies. Also, studies of Dorsch and Hacker$^{16}$ with very small water droplets indicated that the spontaneous freezing temperature decreases rapidly with decreasing droplet size. (Figure 33) This might tend to justify doubt on the reality of attempting to distinguish the difference between liquid and solid states at droplet sizes of critical clusters. [For CO$_2$, typical critical radii are of $7.5 \times 10^{-8}$ cm to $10^{-7}$ cm (about 30 - 50 molecules) at supersaturation ratios of the order of 10, where noticeable quantities of moisture are condensed. Typical nucleation rates are $10^{17}$ to $10^{21}$ nuclei/cm$^3$ - sec.]
Figure 34 illustrates existing data on vapor pressures of CO₂ in the region of experimental condensation, and the computational approximations used in the condensation calculations. Accuracy of the vapor pressure data and the approximation below temperatures of about 180⁰K is uncertain, but is probably within 5% for higher temperatures. Supersaturation ratio may be determined within these limits of accuracy for a given local pressure if the temperature is also known.

Surface tension is considered to be described for flat film liquid by the function \( \sigma_\infty = 5.00 - 0.193T(°C) \) dynes/cm, based on data of References 21, 27, and 50, for which a linear dependence of \( \sigma_\infty \) on \( T \) down to 220⁰K is justified. An alternative function \( \sigma = 75(1 - \frac{T}{T_c})^{1.25} \) (Reference 27) yields values of \( \sigma_\infty \) which vary by only 2% to 9%, in the 200⁰K to 160⁰K region of interest, from those of the linear function above.

Knowledge of local temperature, previously discussed, is important in evaluating the nucleation rate equation, not only because of its presence as a cubic in the exponent, but because the other variables are also dependent in value upon temperature. Effects of uncertainty in temperature do not cancel appreciably in their influence upon theoretical nucleation rate. At 200⁰K, a 1% error in temperature will result in about a 2% error of the opposite sign in surface tension and hence about a 3% error in the ratio \( \sigma/T \) for CO₂. These effects were illustrated in the earlier discussion of the potential uncertainty of local temperature unless an equation of state is used to determine the isentrope for CO₂. It is possible that some past applications of theory to experimental condensation data have erred through failure to consider these real gas effects.

Departures from perfect gas behavior affect the evaluation of the free energy required for the formation of a critical cluster, and also the drop growth relations dealt with in the following section, since both nucleation
and drop growth developments are based on perfect gas assumptions. This problem is dealt with in Section IVD, following the discussion of drop growth theory, and the application of nucleation and drop growth to condensation in nozzles.

B. Drop Growth Theory

Subsequent to the production of nuclei, additional formation of moisture will occur through the growth of the clusters due to vapor impingement. Nuclei may by this process grow to radii of up to about an order of magnitude greater than their original size, depending upon two important drop growth parameters introduced below. In the theoretical treatment, the distinction between nucleation and commencement of drop growth treatment is somewhat artificial, as it is necessary to establish some size of cluster at which the nucleated droplet exists and may commence growing. Becker and Doring have shown that at a cluster radius 1.3 times critical, the probability of clusters continuing to grow reaches better than 99%, and hence it is considered that this is a reasonable starting point for the application of drop growth equations. (Note: Computations showed that results were relatively insensitive to this starting radius.)

The growth of a droplet depends predominantly on two factors: 1) The rate of condensing mass flux 2) The rate of droplet energy transfer to the vapor environment.

It would appear that the first factor is itself a description of the drop growth process, but in actuality, it is greatly dependent upon not only local vapor conditions, but also the difference between droplet and vapor conditions. Both of these are strongly influenced by the second factor.
Two coefficients are defined to describe this behavior:

\[ \xi = \text{Condensation Coefficient}. \] The fraction of impinging vapor molecules which are absorbed at the drop surface.

\[ \alpha = \text{Thermal Accommodation Coefficient}. \] The fractional temperature change which takes place in the reflected portion of incident molecules.

\[ \alpha = \frac{T_r - T}{T_D - T} \quad (13) \]

Temperature variation across the droplet is assumed to be negligible, and the droplet is assumed to be spherical.

Based on mass and energy balance considerations presented and developed in Appendix C the following drop growth equations are obtained.

**Conservation of Mass**

\[ \rho_L 4\pi r^2 \frac{dr}{dt} = 4\pi r^2 \left( \frac{\xi \rho}{(2\pi RT)^{1/2}} - \frac{\rho P_D}{(2\pi RT_D)^{1/2}} \right) \quad (14) \]

**Conservation of Energy**

\[ \frac{4}{3} \pi \rho_L C_L \frac{dT_D}{dt} + 4\pi r^2 \frac{dr}{dt} \rho_L U_f = \beta (2RT) - \xi \beta_D (2RT_D) \]

\[ - (1 - \xi) \beta_2 R[T + \alpha (T_D - T)] \quad (15) \]

These equations may be reduced, respectively, to

\[ \frac{dr}{dt} = \frac{\xi}{\rho_L} (\beta - \beta_D) \quad (16) \]

\[ 0 = 2\xi (1 - \frac{\beta D}{\beta D \frac{T_D}{T}}) - 2(1 - \xi) \alpha \left( \frac{TD}{T} - 1 \right) - \xi (1 - \frac{\beta_D}{\beta_D} \frac{Y}{Y - 1}) (\lambda - 1) \quad (17) \]

where

\[ \lambda = \frac{Y - 1}{Y} \quad \frac{hfg}{RT} \]

\[ \beta = \frac{\rho P}{(2\pi RT)^{1/2}} \]

\[ \beta_D = \frac{P_D}{(2\pi RT_D)^{1/2}} \]

And \( P_D \) is the equilibrium vapor pressure for a spherical liquid drop of temperature \( T_D \) in an environment of temperature \( T_D \).

\[ P_D = P_\infty (T_D) \exp \left( \frac{\delta}{\rho_L RT_D r} \right) \quad (18) \]

In the further application of these equations to streamwise growth of drops in a nozzle, additional assumptions are made. First, for an
acceptably simple computation scheme it is convenient to treat droplets of uniform size, where the radius is an average based either on surface area or volume. Ignoring the effects of coagulation, there is evidence that the actual distribution of drop sizes is narrow, and that the average droplet treatment does not introduce appreciable error.

One possible error that may occur as a result of the above assumption becomes likely when the supersaturation ratio $P/P_\infty$ is reduced so rapidly as a result of condensation, that droplets, which have recently nucleated and begun to grow, will suddenly be of size less than the critical cluster size. In this case, considerable re-evaporation would occur among the smaller droplets of the actual size distribution, and the treatment of an "average" droplet does not account for this.

Relative motion between droplets and the gas stream is ignored, and though this is generally accepted, it may be possible that the same translational modes of energy required by recent authors in nucleation theory might also affect to some degree such things as incident mass and energy fluxes upon newly formed nuclei.

Values of $a$ and $\xi$ have been presented by numerous investigators for several vapors, excluding CO$_2$. Although earlier presented values of $\xi$ were fairly low, there has been more recent substantiation of values close to 1.00 for most materials, including liquid metals, the previous errors having been caused by neglect of surface temperature depressions and the effects upon evaporation flux. However, there is also indication that molecules of higher dipole moment tend to have lower values of $\xi$, as with liquid water for which Alty and MacKay,\textsuperscript{1,2} and Prueger\textsuperscript{49} present values of less than $\xi = 0.4$. The influence of crystallization upon $\xi$ may also be large, as Tschudin presents a value close to unity for water ice. Effects
of surface contamination and surface diffusion barriers (to recently evaporated particles due to insufficient vacuum and mean free path lengths,) are also thought to play a role in experimentation which tends to lower apparent values of $\xi$. No experimental values of $\xi$ have been measured for CO$_2$, which has a dipole moment of about 1.5 compared with 80.0 for that of water.

The thermal accommodation coefficient for vapors and their liquids is generally accepted to be around unity. However, values may be much lower between vapors and solids, as Wachman$^{60}$ indicates for gases on clean metal surfaces. For CO$_2$, the value of $\alpha$ for the vapor and its solid crystal is unknown, and there is uncertainty as to which of the two situations above is the most applicable description.

Departures from perfect gas behavior also influence drop growth. Again, correct values of local temperature and supersaturation ratio will be influenced, and the perfect gas assumptions used in treating mass and energy balances leading to equations (16) and (17) may require modification.

In this study, considerable attention has been given to the behavior and influence of the above mentioned factors.

C. Application of Nucleation and Drop Growth Equations

The predicted effects of moisture formation during a nozzle expansion are determined by applying the time rate release of heat to the gas dynamics of the related expansion. By considering the process with respect to variable nozzle position, rather than time, a result is obtained which can be directly compared with experiment.

Interferometric measurements of density variations in the nozzle verified that the flow between boundary layers is in fact one-dimensional and allows the use of the one-dimensional gas dynamics relations;
Continuity
\[ \dot{m} = \frac{\rho A u}{1 - \mu} \]  \hspace{1cm} (19)

Momentum
\[ -Adp = \dot{m}du \]  \hspace{1cm} (20)

Energy
\[ C_p \Delta T + udu = h_{fg} du \]  \hspace{1cm} (21)

where \( \dot{m} \) is the mass rate of flow through the nozzle, \( \mu \) the mass fraction of condensed moisture, \( u \) is the streamwise velocity of the vapor, and \( \rho, P, \) and \( T \) are the vapor density, pressure and temperature, respectively.

\( A \) is the effective area of flow between boundary layer thickness, and is determined experimentally for non-condensing flow.

The rate of moisture formation is determined as follows and assumes the droplet velocities to be those of the vapor. A nucleus of radius \( r_0 \) formed at some point \( x_0 \) will grow to a larger size at some future time and distance \( x \).

\[ r = r_0 + \int_{x_0}^{x} \frac{1}{u} \frac{dr}{dt} \, dx \]  \hspace{1cm} (22)

and the surface area will be

\[ a = 4 \pi [r_0 + \int_{x_0}^{x} \frac{1}{u} \frac{dr}{dt} \, dx]^2 \]  \hspace{1cm} (23)

For a rate of growth \( \frac{dr}{dt} \) predicted by drop growth, the increase in liquid mass per droplet will be

\[ \rho_L \frac{d\dot{r}}{dt} = \rho_L 4 \pi [r_0 + \int_{x_0}^{x} \frac{1}{u} \frac{dr}{dt} \, dx]^2 \frac{dr}{dt} \frac{1}{u} \, dx \]  \hspace{1cm} (24)

And the fractional moisture change that occurs at \( x \) for the

\[ \frac{dn_o}{n_o} = \frac{J(x_0)A(x_0)}{u_0} \, dx_0 \]  \hspace{1cm} (25)

droplets \( \frac{dx_0}{u_0} \) particles formed in the volume \( A(x_0) \, dx_0 \) is

\[ \frac{dn_o}{n_o} = \frac{J(x_0)A(x_0)}{u_0} \, dx_0 \rho_L \pi [r_0^+ x_0] \frac{1}{u} \frac{dr}{dt} \, dx \frac{dr}{dt} \frac{dx}{u} \]

Inclusion of the drop growth on all previously formed nuclei necessitates integrating the right hand term over all \( x_0 \). Noting that
\[ \dot{m} = \rho_{o} A_{o} u_{o} / (1 - \mu_{o}) \] and considering also the moisture produced through additional nucleation at \( x \), there results

\[ \frac{d\dot{u}}{dx} = \int_{0}^{x} J(x_{0}) \frac{(A(x_{0})dx_{0})p_{l} m_{m}}{\dot{u}} \left[ r_{o} + \int_{x_{0}}^{x} \frac{1}{u} \frac{dr}{dx} dx_{0} \right]^{2} \frac{dr}{dt} dx_{0} \]

\[ + \frac{4 \pi r^{3}}{3} \frac{\dot{p}}{m} J(x)A(x) \]

\[ = \frac{4 \pi r^{3}}{m} \left[ \int_{0}^{x} (r_{o} + \int_{x_{0}}^{x} \frac{1}{u} \frac{dr}{dt} dx_{0})^{2} J(x_{0})A(x_{0}) \frac{1}{u} \frac{dr}{dt} dx_{0} \right] \]

\[ + 1/3 \ r^{3} J(x)A(x) \] \hspace{1cm} (26)

For nuclei generally only slightly larger than critical size, the theoretical drop growth rate is essentially independent of size. \(^{23}\) Hence \( \frac{dr}{dt} \) is assumed to have a value equal to that for large drops. Also, since it is practically impossible to keep account of the radii of each group of growing nuclei which formed at each section \( dx \), it is assumed that the droplets can be represented by the "surface-area averaged" radius of droplets. As previously mentioned, volume averaging has little influence on the results. In addition, Lundgren has given experimental evidence that the actual size distribution of particles condensed in a nozzle expansion is quite narrow.

The foregoing equations are reduced to the following form for use with a Runge-Kutta-Merson integration procedure along the expansion path. (The detailed listing of computation procedure may be seen in Appendix H).

\[ dY_{1} = 8 \pi JA \]

\[ dY_{2} = Y_{1} \frac{dr}{dt} + r^{*}dY_{1} \]

\[ dY_{3} = Y_{2} \frac{dr}{dt} + JA(4 \pi r^{*}^{2}) \]

\[ dY_{4} = \frac{d\dot{u}}{dx} = \frac{\rho_{l} m_{m}}{m} \left[ Y_{3} \frac{dr}{dt} + JA(\frac{4 \pi r^{*}^{3}}{3}) \right] \]

\[ dY_{5} = \frac{dp}{dx} = P(\lambda - \frac{1}{1 - \mu})d\mu - \frac{1}{A} \frac{dA}{dx} / [1 - (1 - \mu)(\frac{1}{\gamma} - \frac{1}{\gamma_{m}^{2}})] \]

\[ dY_{6} = \frac{dT}{dx} = T[\lambda d\mu + \gamma - \frac{1}{\gamma}(1 - \mu) \frac{dp}{P}] \]

\[ dY_{7} = \frac{dM}{dx} = M[(- \frac{1}{\gamma_{m}^{2}}) \frac{dp}{P} - \frac{dp}{2T}] \]
where

\[
\lambda = \frac{h_{fg}}{C_p T} \\
Y = \frac{C_p}{C_v} \\
M = \frac{u}{\sqrt{YRT}}
\]

Initial values of the stagnation conditions, physical properties of the vapor and its liquid, values of \(a\) and \(\xi\), and calculation starting point are given for each computation. Effects of contamination may also be considered by giving non-zero values to \(Y(1), Y(2)\) and \(Y(3)\), based on an initial concentration and size of contaminant. Effects of possible droplet energy modes upon free energy of cluster formation ("gassified" behavior of droplets — page 23) may be considered by multiplying nucleation rate by constant arbitrary factors, equivalent to the values proposed by various authors.

The various sub-routines of the listed computation scheme accomplish the determination of such things as the change in nozzle area, vapor and liquid properties, supersaturation ratios and solution to the drop growth equations when step-wise changes are made along the stream of the expansion. An error estimation is made for each computation and the size of the step, \(dx\), is adjusted to reduce the error bounds to within the desired limits.

D. Real Gas Effects

The preceding developments of nucleation and drop growth theory and application to nozzle condensation computations assume perfect gas behavior during the expansion. However, as earlier discussion illustrated, experimental measurements were being made for which values of \(\frac{PV}{RT}\) which were considerably less than unity. Consequently, the computational scheme was modified so that the effects of departure from perfect gas behavior could also be considered.

A feature of an isentropic expansion which facilitates fairly simple corrections to the gas dynamics, drop growth and nucleation equations is the
fact that for this particular thermodynamic process, the compressibility factor remains fairly constant, changes of less than 2% in the value of \( z \) generally occurring for expansion pressure ratios of 10 or more. This was found to be true for several other gases, including ammonia, and for CO\(_2\) is valid throughout the superheated and supersaturated regions for which compressibility factors are in excess of about .88. Using this fact, it may be shown (Appendix D) that the following relations are approximately true for an isentropic expansion.

\[
P\varepsilon \frac{P}{RT} = z = \text{constant} \tag{27}
\]

\[
\gamma' = \frac{\gamma}{1 + (\gamma-1)(1-z)} = \frac{dP}{d\rho} \left( \frac{\rho}{P} \right) \tag{28}
\]

\[
R' = \frac{R_z}{m} = zR \tag{29}
\]

where \( \gamma' \) and \( R' \) are the effective ratio of specific heats and gas constant, respectively, that are applicable. It should be noted here, that the nucleation rate is effected in the following manner:

\[
J \propto \exp \left( \frac{-\frac{4\pi r^2 \gamma}{3kT}}{\rho_L zRT \ln \frac{P}{P_\infty}} \right); \quad r^* = \frac{2\pi \sigma}{\rho_L zKT \ln \frac{P}{P_\infty}}
\]

due to the change in free energy required to form a critical cluster.

To apply these effects to the perfect gas relations of the condensation computations, two additional steps were necessary. First, modifications which allowed for use of a variable ratio of specific heats (\( \gamma = \gamma(T) \)) were made. Second, each set of stagnation conditions which was to be placed in the condensation scheme first had its "real" isentrope, \( z \), and effective isentropic exponent \( \gamma'(T) \) determined, and from these the \( \gamma(T) \) which corresponded to \( z \) and \( \gamma' \). Computation input was \( \gamma(T) \) and correction (compressibility factor) \( z \). This resulted in an isentrope, providing the correct values of \( P \) vs. \( T \) being employed in each condensation calculation, plus the use of effective values of \( \gamma \) and \( R \) in the drop growth and nucleation calculations. Additional details of this procedure are presented in Appendix D.
E. Results of Application of Theory to Condensation of CO₂

1. Comparison of Theory and Experimental Data

Data shown in Table 14 and Fig. 31 illustrate the local conditions at the occurrence of condensation for each of the experimental tests made. Local temperature was determined through computation of the isentrope for each test case, using the procedure discussed on page 13 based on Plank's Equation of State for CO₂. Reference to Figure 35 illustrates that .1% presence of moisture is a convenient and reasonable criteria for theoretical onset of condensation. Comparing the data and the predictions of theory involved two main comparisons; 1) The location of the onset of condensation, defined as the theoretical occurrence of .1% moisture, and 2) The shape of the pressure profiles. For lack of better knowledge, α, ξ and \( \frac{\sigma}{\sigma_m} \) were originally assumed to be unity. Specific comparisons were made of the predictions of 1) Classical nucleation theory 2) Droplet "gassification" corrections to the theory 3) "Real" gas vs. perfect gas treatment of the classical theory (to determine whether or not a simplified approach, containing known errors, might yet yield a better prediction of observed behavior.) 4) Use of liquid vs. solid droplet density and 5) the predictions of the theory in nozzles of different geometry. Figures 36 through 39 illustrate these comparisons and support the following conclusions:

1) All predictions based on values of \( \sigma/\sigma_m \), α and ξ of unity predict condensation at considerably lower supersaturation ratios than those observed experimentally.

2) Real Gas treatment of the classical theory yields a closer prediction than perfect gas treatment, and the "gassification" corrections are in the wrong direction. (Fig. 36)

3) Use of liquid rather than solid droplet density yields a prediction nearer to experimental results, lacking any better knowledge of
what corrections might be applied to surface energies of small CO₂ crystals. (Fig. 37)

4) The unmodified theory does extremely poorly in predicting any effects of nozzle geometry. (Fig. 38)

5) The shape of condensation profiles predicted by values of α and ξ of unity are in extreme disagreement with those observed experimentally. (Figs. 45 and 46)

2. Influence of Nucleation and Drop Growth Theory on Theoretical Predictions.

It is concluded that homogeneous nucleation and drop growth theory fail in their ability to predict observed condensation behavior of CO₂, unless some modification in values of the nucleation and drop growth parameters σ, ρ_L, α, and ξ can be independently established. Toward better understanding of how these parameters influence predicted condensation phenomena, and with the idea in mind of finding corrections for the prediction of CO₂ condensation, a series of calculations based on arbitrary variations of σ, ρ_L, α and ξ were made.

Figures 39 and 40 show typical pressure profiles for condensation of CO₂, for various condensation and thermal accommodation coefficients. Shown in the lower portion of the figures is the nucleation rate, indicating how nucleation rate remains high for longer duration when drop growth is retarded. This results in condensation clouds of greater particle density and smaller particle size than in the case of ready drop growth. In addition, smaller total deflection in the static pressures (from non-condensing), indicating lesser amounts of moisture, exist for the downstream portions of profiles having lower drop growth parameters.

An important observation in these figures is that, beyond a certain reduction in the drop growth parameters, appreciable proportions of moisture
may be present primarily from nucleation. This is manifested in the form of a so-called "ramp" deflection in the pressure profile to which additional drop growth effects are super-imposed. Thus, for example in Figure 39, for an assumed $\alpha$ of .10, variations in $\xi$ from 1.0 to .3 and then to .1 have progressively less effect on the point of departure of the condensation curve, and for lower values of $\xi$, the nucleation "ramp" becomes increasingly apparent. The same behavior occurs for decreasing values of $\alpha$, (shown for an assumed value of $\xi = .04$) in Figure 40. It would be possible to obtain a deflection curve involving only nucleation, if drop growth were set at zero. 

Figure 41 illustrates the influence of increasing or decreasing the nucleation rate by changing the free energy of formation of nuclei. Indicated are variations in the surface energy from flat film liquid value, though similar changes may be introduced by varying the droplet density $\rho_L$. The important parameter in the nucleation rate exponent is $(\rho^3/\rho_L^2)$, since, with knowledge of temperature and supersaturation ratio, the uncertainties of surface energy corrections, small cluster densities, and non-spherical geometry may all be contained in this term.

Two interesting features may be noted in Figure 41. First is the effects of higher downstream velocities, which tend to "wash out" the curves further downstream, and second, the important observation that, for $\alpha$ and $\xi$ of unity, condensation results in a return to near-equilibrium conditions, seen by the fact that condensation commencing at any point in the nozzle results in the same pressure deflection immediately further downstream.

a) Onset of Condensation

As the previous figures suggest, influence of the drop growth coefficients upon the point of onset of condensation is relatively small compared to that caused by changes in surface tension. However, uncertainties in $\alpha$
and ξ still have appreciable effect upon values of surface energy necessary to bring the predictions of theory into agreement with experiment. Figures 42, 43, and 44 show the corrections to flat film surface tension necessary to provide agreement between theory and experiment (based on fitting the data of nozzle IX(2.20°)). If the uncertainty in the values of α and ξ for CO₂ crystals, is given limits of from perhaps .01 to 1.0 for each, then the respective surface energy corrections necessary may range from about 20% to 33% increases in the flat film surface tension. For the absolute lower bound of uncertainty on α and ξ of zero, the necessary correction in surface energy is reduced to +17%.

It is noted in these figures that the theoretical effects of nozzle geometry are increased to some extent by use of small, but non-zero values of the drop growth parameters, but that the theory in all cases predicts less effect than that recorded experimentally.

b) Profile Shapes

Figure 45 illustrates the pressure profiles for a single expansion corresponding to the same values of α and ξ, and corresponding σ determined above, and indicates an additional distinct failing of the theory in its description of the early stages of condensation. It is apparently impossible to reconcile the experimental downstream pressure deflections, which indicate a considerable degree of supersaturation, or departure from equilibrium, with the apparent rates of formation of moisture immediately after onset. When drop growth parameters are adjusted to bring theoretical profiles into agreement regarding the amount of downstream pressure deflection, then the upstream theoretical rate of formation of moisture lags considerably behind the experimental results.

This problem may result from some of the approximations discussed in the numerical calculations, or may in fact indicate some physical "arrest"
of the condensation process, perhaps due to decreasing mass or thermal accommodation characteristics of crystals which grow rapidly colder during their passage downstream.

c) **Downstream Moisture Formation**

For purposes of calculating the amounts of carbon dioxide that will form, or the downstream pressure variations which will occur during a nozzle expansion of CO₂, the following parameters were determined, having arbitrarily set thermal accommodation coefficient, $\alpha$, at unity. There results, as a best description of the three nozzles, the profiles shown in Figures 46, 47, 48 and 49 for $\alpha = 1.0$, $\xi = .00008$, $\rho_L = 1.18 \text{ g/cm}^3$, $\rho/\rho_\infty = 1.18$. These drop growth parameters have no physical significance, and are presented only as an aid in determining approximate amounts of CO₂ moisture formation subsequent to the occurrence of condensation in a nozzle expansion.

The best compromise to profile shape soon after onset has been a recipe which yields theoretical profiles lagging behind and below the experimental data early in the condensation, and which then pass slightly to the other side of the data by about the same amount for the remainder of the profile.

The condensation data of Nozzle II (0.80) indicates the presence of some cyclic axial variation in the pressure ratios of all profiles. This suggests that at this narrow angle, some departure from 2-dimensional flow may be interfering with the condensation process. Friction, or compression shocks may be creating these fluctuations, and would also result in decreased supersaturation ratios in this region, offering a possible explanation as to why considerably less moisture is forming than the recipe predicts for that nozzle.
3. Influence of Contamination

Computations attempting to determine the influence of contamination of vapor in nozzles were carried out at intervals during this investigation. Original calculations were based on perfect gas assumptions and values of $\alpha$, $\xi$, and $\frac{a}{a_\infty}$ of unity, in an attempt to establish whether or not condensation could possibly be due to reasonable amounts of contamination present in the test vapor. Such contaminants might be present in the form of dust, charged particles, or the result of an earlier condensation of some other vapor. Contamination of this sort would be expected to cause earlier condensation than would occur if the test vapor were perfectly homogeneous.

Junge\textsuperscript{27} presents data for typical size density distributions of contamination in atmosphere. Upper limits of particle concentration in dirty atmosphere of a large city reach values of $5 \times 10^5$ cm$^{-3}$ for particles of size less than $10^{-5}$ cm (.1 micron), about $10^3$ cm$^{-3}$ for sizes from .1 to 1.0 microns and 1.0 cm$^{-3}$ for particles exceeding one micron in size.

Typical profiles of supersaturation obtained for increasing concentrations of contamination with microscopic particles are shown in Fig. 50. Beyond a limiting concentration of particles, (depending on size) the supersaturation ratio obtains lesser and lesser values, until the flow reaches a state where it is close to equilibrium throughout the expansion, due to the ready formation of moisture on contaminant particles.

Subsequent to the establishment of the "recipe" described in the previous section, additional computations for both microscopic and macroscopic contamination were made.

The results are shown graphically in Figures 51 and 52, and are briefly summarized:

1) Micron-sized particles in excess of $10^8$/ cm$^3$ are required to influence the theoretical condensation of CO$_2$. This is at least 5 orders of
magnitude larger than upper atmospheric concentrations of particles of this size.

2) Microscopic particles, such as might occur from the precondensation of another vapor, of size the order of $10^{-7}$ cm are required in excess of $10^{12}$/cm$^3$ to influence theoretical condensation of CO$_2$.

3) No appreciable difference between empirically fitted real and perfect gas theories results.

4) The difference between amounts of contamination necessary to influence condensation in the rapid expansion of Nozzle I and the slow expansion of Nozzle II (0.80") is approximately one order of magnitude, independent of particle size.

5) The empirically fitted recipe predicts necessary concentrations of particles roughly two orders of magnitude larger than that predicted using values of $\alpha$, $\xi$, and $\sigma/\sigma_\infty$ of unity.

When the curves of Fig 52 are replotted in terms of $\left[ \log_{10} \frac{V_{\text{CNTAM}}}{V_{\text{TOTAL}}} \right]$, the two curves tend to move together, since the first curve to drop downward represents larger volume contamination. This results in fairly localized values of supersaturation ratio at occurrence of condensation as a function of volume contamination, regardless of particle size.

Applying the volume amounts of contamination of CO$_2$ in nitrogen presented by Willmarth and Nagamatsu, and Arthur, a computation of the predicted effects of theoretical influence of contamination on nitrogen was made, using values of $\alpha$, $\xi$, and $\alpha/\alpha_\infty$ of unity and perfect gas assumptions for nitrogen. The results are shown in Figure 53, for various size particles typical of cluster sizes for CO$_2$, indicating reasonably good agreement with their data. This is taken to indicate some substantiation of the above calculations for contamination in CO$_2$, perhaps within one or two orders of magnitude of the calculated contamination levels needed to influence homogeneous condensation.
V. SUMMARY OF FINDINGS AND CONCLUSIONS

A. Experimental Findings

1. Consistent data regarding the supersaturation of CO$_2$ at onset of condensation plus the shape of pressure profiles has been obtained for three different nozzles.

2. Interferogram photographs and an approximate theoretical estimation show agreement that the boundary layer in the nozzle throat was of the order .006" and substantiates the treatment of flow in the mid-stream as 1-dimensional isentropic flow.

3. Departures from perfect gas behavior by CO$_2$ in the P-T range tested are significant, and must be accounted for in the calculation of local conditions along an isentropic expansion. Density and static pressure measurements plus total pressure measurements for non-condensing flow indicate that the Plank Equation of State gives a good description of low temperature CO$_2$ behavior.

4. Rates of CO$_2$ expansion, as determined by nozzle geometry have a large influence upon the degree of supersaturation obtained prior to occurrence of condensation.

B. Application of Nucleation and Condensation Theory

1. Classical nucleation and drop growth theory, as applied herein, will reasonably predict the conditions at which onset of CO$_2$ condensation occurs, if flat film liquid surface energies are corrected by amounts within suggested ranges of uncertainty for small droplets or crystals. In these calculations a droplet density equal to that of CO$_2$ liquid at the triple point, 1.18 g/m/cm$^3$, has been assumed.

2. The appropriate correction to surface energy is dependent upon the values of mass and thermal accommodation coefficients, $\xi$ and $\alpha$, and has a correction uncertainty of about 21% of flat film values for maximum possible
uncertainty in $\xi$ and $\alpha$. $(\frac{\sigma}{\sigma_\infty} = 1.17$ for $\xi = \alpha = 0.0; \frac{\sigma}{\sigma_\infty} = 1.38$ for $\xi = \alpha = 1.0.)$ Consequently, the applicable value of the parameter $(\sigma^3/\rho_L^2)$ to be applied to the nucleation rate equation corresponds to values of $(\sigma^3/\rho_L^2)(\rho_L^2/\sigma_\infty^3)$ which lie between the limits $(1.17)^3$ and $(1.38)^3$ where $\rho_L = 1.18$ gm./cm$^3$ and $\sigma_\infty$ is the linear temperature dependent function $\sigma_\infty = 5.00 - .193T(\circ\text{C})$ dyne/cm.

3. Pressure profiles obtained experimentally indicate that considerably less moisture is forming than that necessary to return the mixture of vapor and condensate to near equilibrium values, or zero supersaturation downstream of the onset of condensation. Values of $\xi$ and $\alpha$ necessary to produce agreement between the theory and experiment in this respect require that one or both of these parameters be considerably less than what are thought to be minimal values (about .01, for each.)

4. Even at their best empirical fit, classical nucleation and drop growth theory as applied herein do not seem to provide any means of describing the exact shape of the pressure profiles obtained experimentally or the extent of effects of geometry noted experimentally, These failings, in addition to that of B3, above, are thought to indicate inadequate or incorrect assumptions in the treatment of the drop growth calculations.

5. Application of the so-called "gassification" corrections to classical nucleation theory results in corrections in the wrong direction, unless even larger corrections in the surface energy of formation of CO$_2$ ice crystals (than indicated above) can be established. Use of solid, rather than liquid, droplet density in the parameter $(\sigma^3/\rho_L^2)$ further increases the necessary corrections to surface energies required to provide agreement with experiment.

6. The influence of dirt particles or other foreign particles on the
condensation of CO₂, and probably most other gases, may be completely disregarded in supersonic nozzles, unless the particles result from the precondensation of another gas in the stream. Amounts of micron-sized particles necessary to influence CO₂ condensation are about five orders of magnitude in excess of upper atmospheric limits of concentration, and particles of the order of 20 a° are necessary in excess of about 10¹² particles/cm.³ to influence homogeneous nucleation and condensation.

Testing the condensation of nitrogen with pre-condensed particles of CO₂ in quantities mentioned by Willmarth and Nagamatsu, and Arthur, yields theoretical predictions which are in reasonable agreement with their experimental observations.

7. Additional knowledge of thermal and mass accommodation coefficients are necessary to allow better testing of homogeneous nucleation and condensation theory. In addition, comprehensive testing of the theory will await some better establishment of surface energy corrections for small particles. Until this information becomes available and an established treatment of nucleation and drop growth can be applied with confidence to many vapors, "recipe" fitting may provide the best means for predicting nozzle condensation behavior.
VI. SUGGESTIONS FOR FURTHER STUDY

One of the greatest difficulties in application of experimental results to a meaningful test of theoretical predictions results from the uncertainties in value of many of the important variables and parameters contained in nucleation and drop growth theory. Consequently, any methods or devices of experimentation which would decrease any of the uncertainties, or the effects of uncertainty, of such quantities as \( \sigma, \rho_l, a, \xi \), local temperature and the gas dynamics in general would be of great assistance. One means of proceeding in this direction is to contain the test gas within large quantities of another gas which is non-condensing in the region of interest of the test gas, and whose gas properties are well-known, preferably being as near those of a perfect gas as possible. An immediate benefit of this is to provide a means of accurately determining local properties at all points of the isentrope with ease, since, the temperature of the test gas should be that of the ideal "carrier" gas, which dominates the gas dynamics. Second, the influence of thermal accommodation is greatly reduced due to the fact that the huge preponderance of incident molecules upon the surface of a formed droplet are those of "carrier" gas, and hence are reflected. The drop is consequently kept at or nearly at environmental temperature regardless of the value of \( a \). Hence, there are advantages to be gained by testing gases for which these uncertainties exist (the huge preponderance of all gases) in a carrier stream of such gases as nitrogen, helium, or dry air. Several complications arise from this method, including the need for very precise measurement of mass fraction of test gas in the mixture, and more sensitive instrumentation to detect the lessened effects of condensation upon the gas properties.
Highly steady flow systems would aid in obtaining the accuracy necessary.

With regard to some of the uncertainties pertaining to CO₂, it would be of considerable interest to determine whether or not the experimental behavior of condensation is influenced by establishing liquid rather than ice formation of condensate particles. This could presumably be accomplished by raising pressure levels to a point where condensation occurs above the ice point, 75.1 psia. This would necessitate stagnation pressure in the range of 175 - 250 psia, mass rates of flow and a source of CO₂ which are proportionally larger, heavier construction of some important fixtures in the system, (such as the stagnation tank) and a means of pressure profile procurement other than manometer board.

It is felt that interferometric means of study offer enough advantages, particularly with more toxic or reactive gases, to warrant further application to study of condensation in nozzles. The inability to distinguish interference lines in the upstream region of the nozzle encountered at high pressure levels in this investigation could possibly be solved by facilitating higher camera shutter speeds (1/150 sec., the maximum possible, was used in these studies) or by establishing wall temperatures which are close to stream stagnation temperature. Two-dimensional nozzles, where interferometric study could be successfully employed, would decrease the problems of instrumentation and would be readily adaptable to studies of light scattering and polarization, yielding experimental information regarding droplet sizes. Interferometric photos involving condensation, although not applied in this report, were obtained, and showed interference bands very clearly within the region of condensation,
this region being marked plainly by the decreased light intensity resulting from the presence of the ice cloud.

Light scattering or other techniques which yield information regarding the average size of the ice particles at their initial appearance and after subsequent downstream growth would be very useful. Since, as previously mentioned, there is some indication that nucleation is the major contributor to moisture in its early stages of formation for CO₂, this information might be directly related to the critical cluster size of CO₂.

Additional investigation of crystal thermal and mass accommodation coefficients would be of help in further establishing the apparent indication from this investigation that either one or both have very low values for crystalline CO₂ at low temperatures. The carrier gas technique previously mentioned could isolate the influence of α and might facilitate a better means of studying ξ and its role in condensation.
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**TYPE I**

- $V + L$
- ISENTROPE

**TYPE II**

- $V + L$
- ISENTROPE

Axis labels:
- T (Temperature)
- S (Entropy)
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<th>C&lt;sub&gt;p&lt;/sub&gt; Specific Heat</th>
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FIG 2 SCHEMATIC ARRANGEMENT OF TEST APPARATUS

9 - 50LB. CO₂ (INDUSTRIAL WELDING)

ATMOSPHERIC EXHAUST
(7" HG ABS.)

STAGNATION TANK

NOZZLE

NEEDLE VALVE
FIGURE 5, STAGNATION TANK WITH END PLATE REMOVED, SHOWING SCREEN AND FILTER BAG CONTAINING STEEL WOOL.
FIGURE 6, NOZZLE 1 SHOWING TOTAL PRESSURE TUBE IN TEST POSITION.
Distance between plates .956" (+.003"

\[
\begin{array}{cccccccc}
 x(\pm 0.005) & b & x & b & x & b & x & b \\
 0.63 & .950 & 1.11 & .061 & 1.325 & .085 & 2.07 & .074 \\
 0.84 & .300 & 1.165 & .086 & 1.400 + .015 & .053 & 1.001 & 2.21 & .086 \\
 0.89 & .250 & 1.190 & .078 & 1.500 & .054 & 2.27 & .090 \\
 0.92 & .225 & 1.200 & .074 & 1.570 & .055 & 2.38 & .101 \\
 0.95 & .200 & 1.220 & .069 & 1.620 & .056 & 2.46 & .110 \\
 0.97 & .175 & 1.235 & .066 & 1.670 & .057 & 2.52 & .120 \\
 1.01 & .150 & 1.250 & .063 & 1.735 & .060 & 2.63 & .135 \\
 1.04 & .135 & 1.270 & .060 & 1.820 & .063 & 2.71 & .150 \\
 1.07 & .120 & 1.290 & .057 & 1.920 & .066 & 2.84 & .175 \\
 1.09 & .110 & 1.305 & .056 & 1.990 & .069 & 2.98 & .200 \\
\end{array}
\]

FIGURE 7, ASSEMBLY VIEW AND GEOMETRY OF NOZZLE I.
FIGURE 9, NOZZLE I; DISASSEMBLED, SHOWING OPTICAL GLASS AND METAL PRESSURE TAP PLATE SIDE WALLS, AND METAL CLAMP BARS.
FIGURE 10, NOZZLE II; DISASSEMBLED, SHOWING HINGED UPPER AND LOWER WALLS, AND PRESSURE TAP PLATES 1 AND 2.
(A) NO FLOW

(B) SCALING PLATE (1.00")

(C) FLOW, SHOWING SHIFT IN INTERFERENCE BANDS

FIGURE 11, INTERFEROMETRIC PHOTOGRAPHS FOR TEST 1 OF DENSITY PROFILES.
(A) NO FLOW

(B) SCALING PLATE (1.00"")

(C) FLOW, SHOWING SHIFT IN INTERFERENCE BANDS

FIGURE 12, INTERFEROMETRIC PHOTOGRAPHS FOR TEST 2 OF DENSITY PROFILES.
FIG 13 GRAPHICAL REPRESENTATION OF DETERMINATION OF FRINGE SHIFTS - TEST NO 1
Figure 14, Comparison of the Plank Equation of State and Data Upon Which It Was Formulated
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TABLE 3
INTERFEROMETRIC MEASUREMENT AND CALCULATION OF DENSITY

Test No. 1

$T_o = -9.5^\circ F$

$P_o = 83.7$ psia

No Flow: Average 28.1 fringes/inch

$\lambda = \frac{5461 \times 10^{-8}}{(0.956)(2.54)} = 2248 \times 10^{-8}$

Photograph Scale: 2.3 in./in.

$\rho_o = \frac{P}{RT} = .306$ lbm/ft$^3$

$Gp_o = N_o^{-1} = (.3673 \times 10^{-2})(.806) = 296.0 \times 10^{-5}$

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TABLE IV
INTERFEROMETRIC MEASUREMENT AND CALCULATION OF DENSITY

Test No. 2

\[ T_o = 117.0^\circ F \quad P_o = 58.7 \text{ psia} \]

\[ \rho_o = \frac{P}{2RT} = 0.4235 \text{ lbm/ft}^3 \]

\[ \lambda = \frac{5461 \times 10^{-8}}{2.54(0.970)} = 2.215 \times 10^{-5} \]

\[ G_{p_o} = N_o - 1 = (0.3673 \times 10^{-2})(0.4235) = 155.6 \times 10^{-5} \]

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FIGURE 15a, NOZZLE I PROFILES OF MEASURED PRESSURE RATIO AND ISENTROPICALLY CONVERTED MEASURED DENSITY RATIO, TEST #1.

- MEASURED PRESSURE, (PRESSURE TAP PLATE #1)
  \( P_0 = 5.09 \text{ ATM., } T_0 = 282.2^\circ\text{K} \)

- CONVERTED DENSITY, (USING PLANK EQUATION OF STATE)
  \( P_0 = 5.71 \text{ ATM., } T_0 = 250.0^\circ\text{K} \)
Figure 16a, Nozzle I Profiles of Measured Pressure Ratio and Isontrically Converted Measured Density Ratio, Test #2.

- Measured Pressure, (Pressure Tap Plate #2)
  \( P_0 = 3.83 \text{ atm}, \ T_0 = 315.0^\circ K \)

- Converted Density (Using Planck Equation of State)
  \( P_0 = 4.00 \text{ atm}, \ T_0 = 320.5^\circ K \)
**Figure 16b** Measured versus theoretical pressure and density ratios, test #2.

- **Experimental**
  - Pressures at $P_0 = 3.83$ atm, $T_0 = 315.0^\circ K$
  - Densities at $P_0 = 4.00$ atm, $T_0 = 320.5^\circ K$

- **Plank equation of state**
- **Perfect gas**
FIGURE 17, EXPERIMENTAL NORMAL SHOCK CHARACTERISTICS OF CO$_2$.

- **TEST SERIES # 1**
- **TEST SERIES # 2**
FIGURE 18, PREDICTED NORMAL SHOCK CHARACTERISTICS OF CO\textsubscript{2} - PLANKE EQUATION OF STATE.

NOTE: POINTS CORRESPOND TO NUMBERED EXPERIMENTAL $P_0$, $T_0$, AND $P$, EXCEPTING POINT #13.
FIGURE 19, COMPARISON OF PREDICTED CO₂ ISENTROPES

PERFECT GAS, $\gamma = 1.28$
PERFECT GAS, $\gamma = 1.32$
PLANK EQ. OF STATE

$P_0 = 7.5$ ATM.
$T_0 = +30.0^\circ$C
FIGURE 20, COMPARISON OF PREDICTED CO₂ ISENTROPES

PERFECT GAS, $\gamma = 1.28$

PERFECT GAS, $\gamma = 1.32$

PLANK EQ. OF STATE

$P_o = 7.5$ ATM.

$T_o = -30.0{^\circ}C$
FIGURE 21, CONSTANT NUCLEATION RATE PROFILES OF CLASSICAL NUCLEATION THEORY FOR CO$_2$. $\theta_{in} = 1.00$.

$J = \text{NUCLEI/CM}^3\cdot\text{SEC.}$

$P \, \text{- ATMOSPHERES}$

$T \, \text{- °K}$

$J = 10^{20}$
$J = 10^{15}$
$J = 1$

SATURATION LINE
FIGURE 22, EXPERIMENTAL PRESSURE RATIO PROFILES. NOZZLE I, PRESSURE PLATES 1 & 2

- TEST # 1
- TEST # 3
- TEST # 5
- TEST # 7

--- NON-CONDENSING (TEST # 8 AND TOTAL PRESSURE MEASUREMENTS)
### TABLE 5
Pressure Data for Figure No. 22

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<th>Run No. 3</th>
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<th>Run No. 7</th>
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<td>( P = 228.5''\text{Hg} ) ((\text{calculated}))</td>
<td>( P_0 = 104.7 \text{ psia} )</td>
<td>( P = 152.7''\text{Hg} ) ((\text{calculated}))</td>
<td>( P_0 = 152.7''\text{Hg} ) ((\text{calculated}))</td>
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<td>( T_0 = 50.6^\circ\text{F} )</td>
<td>( T_0 = -47.2^\circ\text{F} )</td>
<td>( T_0 = -45^\circ\text{F} \pm 2^\circ\text{F} )</td>
<td>( T_0 = -35^\circ\text{F} \pm 5^\circ\text{F} )</td>
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<td>( \frac{P}{P_0} )</td>
<td>( P(''\text{Hg}) )</td>
<td>( \frac{P}{P_0} )</td>
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FIGURE 23, EXPERIMENTAL PRESSURE RATIO PROFILES, NOZZLE I, PRESSURE PLATES 1 & 2

- TEST # 2
- TEST # 4
- △ TEST # 6

--- NON-CONDENSING (TEST # 8 AND TOTAL PRESSURE MEASUREMENTS)
**TABLE 6**

Pressure Data for Figure No. 23

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<th>Run No. 8</th>
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<td>( P_0 = 75.7 \text{ psia} )</td>
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<td>( T_0 = -45^\circ F )</td>
<td>( T_0 = -40^\circ F \pm 2^\circ F )</td>
<td>( T_0 = +48.4^\circ F )</td>
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*Indicates improved values determined from subsequent total pressure tests.*
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* Assumed Station 8 Total Pressure Ratio $P_8/P_0 = .560$
### Table 10

Pressure Data for Figure No. 27

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<th>Test No. 19&lt;sub&gt;II&lt;/sub&gt;</th>
<th>Test No. 12&lt;sub&gt;II&lt;/sub&gt;</th>
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(See Table 9)
### Table II
Pressure Data for Figure No. 28

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(See Table 9.)
FIGURE 29, EXPERIMENTAL PRESSURE RATIO PROFILES. NOZZLE II, PRESSURE PLATE 2, ANGLE OF DIVERGENCE = 0.80°

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TEST # 24
TEST # 25
TEST # 26
TEST # 27
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* Assumed Station 8 Total pressure ratio $P_0/P_0 = 0.627$
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* Assumed Station 8 Total Pressure Ratio P₈/P₀ = .627
FIGURE 31. CONDENSATION DATA.
LOCAL CONDITIONS AT ONSET OF CONDENSATION.

- NOZZLE I
- NOZZLE II (2.20°)
- NOZZLE II (0.80°)

(DOTTED LINES SHOW EXPERIMENTAL MEAN)
TABLE 14
EXPERIMENTAL DATA
LOCAL CONDITIONS AT ONSET OF CONDENSATION

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Figure 33, spontaneous freezing temperature of supercooled water droplets on platinum and copper surfaces.

Experimental data of Dorse & Hacker.
FIGURE 34, LOW TEMPERATURE VAPOR PRESSURE VALUES

- EXISTING DATA

- COMPUTATIONAL APPROXIMATION
FIGURE 35, PRESENCE OF MOISTURE IN A TYPICAL CONDENSATION PROFILE.
FIGURE 36, ONSET OF CONDENSATION, NOZZLE II, COMPARISON OF THEORY AND EXPERIMENT.

\[ \frac{\sigma}{\sigma_0} = \alpha = \xi = 1.00 \]
FIGURE 37, INFLUENCE UPON THEORY OF USE OF LIQUID VERSUS SOLID DROPLET DENSITY. NOZZLE II (2.20°)

\[
\frac{\sigma}{\sigma_\infty} = \xi = \alpha = 1.00
\]

- \( \rho_L = 1.18 \text{ gm./cm}^3 \)
- \( \rho_L = 1.55 \text{ gm./cm}^3 \)
FIGURE 38, PREDICTED INFLUENCE OF NOZZLE GEOMETRY ON OCCURRENCE OF CONDENSATION, USING $\xi, \alpha, \frac{C}{\alpha}$ OF UNITY. $p_L = 1.18 \text{ gm./cm}^2$.
FIGURE 39, INFLUENCE OF MASS ACCOMMODATION COEFFICIENT ON CONDENSATION OF CO$_2$ (THEORETICAL).

$T_0 = -26.0\ C \quad \rho_L = 1.18\ \text{GM./CM}^3$

$P_o = 5.39\ \text{ATM.}$

$\sigma = 1.00$

$\sigma / \sigma_{\infty}$

$\alpha = .10$

$P / P_o$

$\xi = 1.0$

$\xi = .30$

$\xi = .10$

$\xi = .04$

$X - \text{CM (NOZZLE I)}$

$\log_{10} J$

$4.0 \quad 4.5 \quad 5.0 \quad 5.5 \quad 6.0$
Figure 40, Influence of Thermal Accommodation on Theoretical Condensation of CO₂.

\( P = 5.39 \text{ atm.} \)
\( T = -26.0^\circ \text{C} \)
PERFECT GAS, \( \gamma = 1.32 \)
\( \xi = .04, \rho_L = 1.18, \frac{\sigma}{\sigma_\infty} = 1.00 \)
FIGURE 42, THEORETICAL OCCURRENCE OF 1% MOISTURE FOR $\alpha = \xi = 1.00$, CORRECTED SURFACE TENSION $\sigma / \sigma_{\infty} = 1.38$.

**EXP.**
- Δ NOZZLE I
- □ NOZZLE II (2.20°)
- ○ NOZZLE II (0.80°)

$\sigma \pm 3\%$

$T - ^{\circ}K$

$P - \text{ATMOSPHERES}$
FIGURE 43, THEORETICAL OCCURRENCE OF .1% MOISTURE FOR $\alpha = \xi = .01$, CORRECTED SURFACE TENSION $\sigma/\sigma_\infty = 1.20$

EXP.

- NOZZLE I
- NOZZLE II (2.20°)
- NOZZLE II (0.80°)

$\sigma \pm 3\%$

P - ATMOSPHERES

1.0

2.0

3.0

4.0

140 150 160 170 180 190 200 210

T - K

SATURATION

NOZZLE I

NOZZLE II (2.20°)

NOZZLE II (0.80°)
FIGURE 44, THEORETICAL OCCURRENCE OF .1% MOISTURE FOR $\alpha = \xi = 0.0$, CORRECTED SURFACE TENSION $\sigma/\sigma_0 = 1.17$.

EXP,

- \( \Delta \) NOZZLE I
- \( \square \) NOZZLE II (2.20°)
- \( \circ \) NOZZLE II (0.80°)

$\sigma \pm 3\%$

SATURATION

P - ATMOSPHERES

T - °K

140 150 160 170 180 190 200 210
FIGURE 45, COMPARISON OF EXPERIMENTAL AND THEORETICAL PRESSURE PROFILES FOR VARIOUS $\xi$ AND $\alpha$.

- EXPERIMENTAL, $P_o = 6.48$ ATM., $T_o = -42.3^\circ$C

$\alpha = \xi = 1.0, \frac{\sigma}{\sigma_0} = 1.38$

$\alpha = \xi = 0.1, \frac{\sigma}{\sigma_0} = 1.20$

$\alpha = \xi = 0.0, \frac{\sigma}{\sigma_0} = 1.17$
FIGURE 46, COMPARISON OF EMPIRICALLY FITTED THEORETICAL CONDENSATION AND EXPERIMENTAL DATA - NOZZLE I. $a = 1.0$, $\xi = 0.00008$, $\sigma = 1.18$

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<td>$\frac{P}{P_o}$</td>
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- $P_o = 7.64$ ATM. $T_o = -46.0^\circ$C
- $P_o = 7.42$ ATM. $T_o = -42.9^\circ$C
- $P_o = 5.14$ ATM. $T_o = -42.9^\circ$C

X - CENTIMETERS

4.0 4.5 5.0 5.5
FIGURE 47, COMPARISON OF EMPIRICALLY FITTED THEORETICAL CONDENSATION AND EXPERIMENTAL DATA, NOZZLE II (2.20"") PRESSURE TAP PLATE NO. 1. $a = 1.00$, $\frac{P}{P_0} = 1.18$, $\xi = .00008$

EXP. THEORY

- $P_0 = 8.01$ ATM. $T_o = -42.9^\circ$C
- $P_0 = 8.01$ ATM. $T_o = -40.8^\circ$C
- $P_0 = 7.81$ ATM. $T_o = -33.3^\circ$C

$P/P_0$ vs. $X$ - CM. (NOZZLE II)
FIGURE 49, COMPARISON OF EMPIRICALLY FITTED THEORETICAL CONDENSATION AND EXPERIMENTAL DATA - NOZZLE II (0.80°). $a = 1.0$, $\xi = 0.00008$, $\frac{q}{q_\infty} = 1.18$

EXP. THEORY

$P_o = 8.72$ ATM. $T_o = -40.4^\circ$C
$P_o = 8.95$ ATM. $T_o = -36.2^\circ$C
**Figure 51. Theoretical Influence of Macroscopic Contamination on Condensation of CO₂, Using Theory Empirically Fitted to Homogeneous Condensation Data.**

Particle Diameter = $10^{-4}$ cm.

- $P_o = 5.39$ atm.
- $T_o = -26.0^\circ$C
- Perfect gas $\gamma = 1.35$

- $P_o = 7.42$ atm.
- $T_o = -42.9^\circ$C
- Perfect gas $\gamma = 1.35$

$\xi = 0.0008$

$\alpha = 1.0$

$\frac{\sigma}{\sigma_0} = 1.18$

$\rho_L = 1.18 \text{ cm.}^3/\text{cm.}^3$

---

**Diagram Details:**

- **Super-saturation Ratio at .1%**
- **Log₁₀(Particles/cm³)**
- **Nozzle I**
- **Nozzle II (0.80°)**
- **Real Gas (Nozzle I)**
- **Perfect Gas (Nozzle I)**
FIGURE 52, THEORETICAL INFLUENCE OF MICROSCOPIC CONTAMINATION ON CONDENSATION OF CO₂, USING THEORY EMPIRICALLY FITTED TO HOMOGENEOUS CONDENSATION DATA. $\xi = 0.00008$, $a = 1.0$, $d_0 = 1.18$, $\rho_L = 1.18 \text{ gm./cm}^3$.

$P_0 = 5.39 \text{ atm.}$
$T_0 = -26.0 ^\circ\text{C}$

PERFECT GAS, $\gamma = 1.35$

$V_0 = 10^{-7} \text{ cm.}$

$r_0 = 1.5 \times 10^{-7} \text{ cm.}$

$\log_{10}(\text{PARTICLES/CM}^3)$

SUPERSATURATION RATIO AT 1.1%
FIGURE 53, THEORETICAL EFFECT OF CO₂ CONTAMINATION ON NITROGEN CONDENSATION. SUPERSATURATION AT 1% MOISTURE.

- 7 x 10⁻⁸ cm³ PARTICLE RADIUS
- 10 x 10⁻⁸ cm³ PARTICLE RADIUS
- 13 x 10⁻⁸ cm³ PARTICLE RADIUS

EXPERIMENTAL (ARTHUR; WILMARTH & NAGAMATSU)

LOG₁₀(V_CO₂/V_total)

SUPERSATURATION RATIO AT CONDENSATION

10⁻⁴  10⁻³  10⁻²  10⁻¹
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APPENDIX B

REVIEW OF NUCLEATION RATE EQUATION DEVELOPMENT

The following is a review of the classical nucleation rate equation as developed by J. Frenkel\(^{19}\). Although Frenkel was not the first to investigate nucleation theory, his result is not greatly different from earlier authors mentioned in the text, and his method provides relatively straightforward consideration of the important aspects and assumptions in classical nucleation theory.

Frenkel's droplet model was based on Volmer's\(^{58}\) earlier model of droplets as generalized molecules, neglecting intermolecular forces, and his method of development was based on that of Zeldovich\(^{68}\). He showed that the distribution of clusters of vapor molecules, resulting from the kinetic interaction of the molecules could be described by a Boltzmann distribution. That is, that (in equilibrium)

\[
N(g) = N_0 \exp \left(-\frac{\Delta \phi}{kT}\right)
\]  

(B-1)

where

- \(N_0\) = total number of molecules
- \(\phi = H - TS\)
- \(\Delta \phi = \phi_L - \phi_V\) = change in free energy of a group of \(g\) free molecules when forming a liquid embryo.

Several simplifying assumptions of debatable validity are made at this point. First, the droplet model is taken to be a liquid sphere, having surface energy described by the flat-film liquid surface tension (or some corrected function thereof). Second, the total change in free energy is assumed to be the work of formation of the surface of the droplet, plus the change in free energy resulting from the change in state of \(g\) molecules condensing from vapor to liquid, with the spherical droplet assumed to be stationary in the vapor system, and having no rotational or vibrational energy. This is the important distinction between classical and more recent "gassified"
droplet models of nucleation theory. Third, the droplet is assumed to form at environmental temperature, an assumption which may also be invalid \(^{(32)}\).

Using these assumptions, and defining \(\phi\) as the free energy per molecule, \(\Delta \phi\) can then be written:

\[
\Delta \phi = g(\phi_L - \phi_V) + 4\pi r^2 \sigma \\
= -T(\Delta S) + 4\pi r^2 \sigma \\
= -gKT \ln \frac{P}{P_\infty} + 4\pi r^2 \sigma
\]  

(B-2)

(B-3)

where \(P_\infty\) equals the flat surface equilibrium pressure of a formed embryo of \((g)\) molecules and \(p\) is the existing local pressure.

Hence,

\[
N(g) = N_0 \exp(g \ln \frac{P}{P_\infty} - \frac{4\pi r^2 \sigma}{KT})
\]  

(B-4)

Saying that:

\[
gm = \frac{4}{3} \pi r^3 \rho_L
\]  

(B-5)

yields

\[
N(g) = N_0 \exp \{ g \ln \frac{P}{P_\infty} - \frac{4\pi \sigma}{KT} \left(\frac{3gm}{4\pi \rho}\right)^{2/3}\}
\]  

(B-6)

where \(\varepsilon = \frac{4\pi \sigma}{\left(\frac{3m}{4\pi \rho}\right)^{2/3}}\)

Inspection of Eq. (B-6) shows that a reasonable equilibrium distribution results when \(p\) is less than \(P_\infty\), that is, in a superheated vapor. However, in a supersaturated vapor, an unreasonable and, in fact, impossible equilibrium distribution is predicted as seen in Fig. (B-1), since it predicts \(N(g) \to \infty\) as \(g \to \infty\). Acknowledging that an equilibrium distribution of drop sizes is unreasonable under supersaturated conditions, early authors treated the system as one in a state of quasi-equilibrium, where droplets are imagined to be removed after reaching some size larger than the critical radius, \(r^*\), which occurs at the maximum of the supersaturated free energy change. This maximum in \(\Delta \phi\) (minimum in \(N(g)\)) occurs when;
FIGURE B1, GENERAL FORM OF CLUSTER SIZE DENSITY DISTRIBUTIONS.
\[ \frac{d\phi}{d\epsilon} (\Delta \phi) = 0 = -KT \ln \frac{P}{P_\infty} + \frac{2}{3} \epsilon g^{-1/3} \]

\[ KT \ln \frac{P}{P_\infty} = \frac{2}{3} \epsilon g^{-1/3} \]

\[ \Delta \phi_{\text{max}} = \frac{1}{3} \epsilon g^{2/3} = \frac{4}{3} \pi r^2 \sigma \]  

Then

\[ \Delta \phi_{\text{max}} = -gKT \ln \frac{P}{P_\infty} + 4\pi r^2 \sigma = \frac{4}{3} \pi r^2 \sigma \]  

(B-7)

Hence

\[ KT(\frac{1}{3} \pi r^3 \frac{L}{m}) \ln \frac{P}{P_\infty} = \frac{8}{3} \pi r^2 \sigma \]

\[ \ln \frac{P}{P_\infty} = \frac{2m \sigma}{\rho L KT r^*} \]  

(B-8)

Equation (B-8) is the Kelvin-Helmholtz expression describing the conditions under which a liquid drop may remain in equilibrium with its vapor phase.

The radius \((r^*)\) and the number \((g^*)\) of atoms of a critical embryo are thus determined. A variation of \(r\) (or \(g\)) in either direction results in a decrease of free energy from the maximum, and consequently the drop is in unstable equilibrium at this point.

In considering the quasi-equilibrium state, and ignoring interaction between nuclei, it is said that nucleation rate is the net rate at which particles of a given size \((g)\) are formed. To maintain quasi-equilibrium, it is further assumed the net rate at which \((g+1)\)-sized droplets are formed from \((g)\)-sized droplets is equal to the rate at which \((g)\)-sized droplets are formed from \((g-1)\)-sized droplets, and so on. That is, nucleation rate is independent of \(g\), in order that the number of droplets of any size remains constant and the rate at which droplets of larger size are imagined to be removed is also equal to the nucleation rate. Correspondingly, it is imagined that single molecules are added to the system at a rate equal to the depletion due to nucleation.

Using this model then, and assuming that growth of nuclei is due only to interaction with free molecules, the nucleation rate is
\[ J(g + 1, t) = \beta \cdot s(g) \times n(g, t) - \gamma(g + 1) \cdot s(g + 1) \cdot n(g + 1, t) \]  
(B-9)

where \( \beta \) = rate at which molecules strike unit surface area.
\( \gamma(g) \) = rate at which molecules leave the surface per unit area
\( n(g,t) \) = distribution of non-equilibrium drops
\( s(g) \) = surface area of a \( g \)-sized drop.

Assuming that no effects of droplet curvature exist, and neglecting the cluster motion,

\[ \beta = \frac{p}{(2\pi m KT)^{1/2}} \]

Also, it is said that if nucleation rate were zero, then the non-equilibrium (or quasi-equilibrium) distribution would in that case have to be equal to the equilibrium distribution \( N(g) \). This is easy enough to see in the superheated case, but perhaps a bit difficult to accept when \( N(g) \) is the fictitious mathematical result corresponding to supersaturation.

Applying this, when \( J(g, t) = 0 \)
\[ \gamma(g + 1) \cdot s(g + 1) \cdot n(g + 1) = \beta \cdot s(g) \cdot n(g) \]
when \( n(g) = N(g) \)

Hence,
\[ \gamma(g + 1) \cdot s(g + 1) \cdot N(g + 1) = \beta \cdot s(g) \cdot N(g) \]

and Equation (B-9) becomes

\[ J(g, t) = \beta \cdot s(g) \cdot N(g) \left\{ \frac{n(g, t)}{N(g)} - \frac{n(g + 1, t)}{N(g + 1)} \right\} \]  
(B-10)

Here some rather notable simplifications and manipulations facilitate a solution to (B-10). For large values of \( (g) \) where these functions may be treated as continuous, this is approximately:

\[ J(g, t) = \beta \cdot s(g) \cdot N(g) \frac{d}{dg} \left( \frac{n}{N} \right) \]

Hence, since it is assumed that \( J(g, t) = J(t) \)

\[ \int d\left( \frac{n}{N} \right) = -\frac{J}{\beta} \int \frac{dg}{s(g) \cdot N(g)} \]  
(B-11)

Using the boundary conditions
(a) \( \frac{n}{N} = 1.0 \) at \( g = 1 \) (b) \( \lim_{g \to \infty} \frac{n}{N} = 0 \)

One obtains

\[
J = \frac{\beta}{\int_1^{\infty} \exp (\Delta \phi/\kappa T) \, dg} \tag{B-12}
\]

Since \( \frac{1}{N} = \frac{1}{N_0} \exp \left( \frac{\Delta \phi}{\kappa T} \right) \) has a sharp maximum in the vicinity of \( g = g^* \), it is possible to approximate (B-12) as:

\[
J = \frac{\beta \, s(g^*) \, N_0}{\int_1^{\infty} \exp (\Delta \phi/\kappa T) \, dg} \tag{B-13}
\]

Using Equations (B-5), (B-6) and (B-8), the exponent may be written:

\[
\frac{\Delta \phi}{\kappa T} = - g \ln \frac{p_0}{p_\infty} + \frac{e}{\kappa T} g^{2/3}
\]

\[
= - g \left[ \frac{2m \sigma}{\rho_L K T \mu^*} \right] + \frac{4 \pi \sigma}{3 K T} \left( \frac{3m}{4 \pi \rho_L} \right)^{2/3} g^{2/3}
\]

\[
= A \left[ 3(g/g^*)^{2/3} - 2(g/g^*) \right]
\]

where

\[
A = \frac{\sigma m}{\rho_L K T} \left( \frac{4 \pi \rho_L}{3m} \right)^{1/3} \left( g^* \right)^{2/3} = \frac{4 \pi \sigma g^*}{3 K T}
\]

\[
J = \int_1^{\infty} \exp \left\{ A \left[ 3(g/g^*)^{2/3} - 2(g/g^*) \right] \right\} \, dg \tag{B-14}
\]

This integral occurs in the Becker and Doring, and Frenkel derivations, and can be solved approximately with the introduction of

\[
\Omega = (g/g^*)^{1/3} - 1
\]

(B-15)

Since the integrand of (B-14) has a very sharp maximum \( e^A \) at \( g = g^* \), the important values of \( \Omega \) are small, and the limits of the integral may be taken at \( +\infty \) and \( -\infty \).

Also, since \( (g/g^*)^{2/3} = \Omega^2 + 2\Omega + 1 \)

\[ dg = 3(1 + \Omega)^2 \, g^* d\Omega \]

And

\[ 3(g/g^*)^{2/3} - 2(g/g^*) = - 3\Omega^2 - 2\Omega + 1 \]

The integral may be written approximately

\[
3g^* e^A \int_{-\infty}^{\infty} (1 + \Omega)^2 \exp (- 3A\Omega^2 - 2A\Omega^3) \, d\Omega
\]

\[ = 3g^* e^A \int_{-\infty}^{\infty} \exp (- 3A\Omega^2) \, d\Omega \]
\[ = 3g^* e^{A(\pi/3A)^{1/2}} \]  

\( J = \beta s(g^*) N_o \left[ \frac{A}{3\pi} \right]^{1/2} \exp(-A) \)  

This is an expression for the nucleation rate, where

- \( N_o = p/KT \)
- \( \beta = p/(2\pi m KT)^{1/2} \)
- \( s(g^*) = \frac{e(g^*)^{2/3}}{c} \)

and with the definitions of \( \epsilon \) and \( A \), (B17) can be shown equivalent to

\[ J = (\frac{p}{KT}) \frac{m}{\rho} \left( \frac{2\sigma}{nm} \right)^{1/2} \exp\left( \frac{-4\pi \sigma^*}{3KT} \right) \]  

This is an expression of the number of nuclei formed per unit volume per unit time, due to the existence of a non-equilibrium distribution \( \eta(g) \). By assuming \( (J) \) was independent of \( (g) \) as Becker and Doring did, it was evaluated using only the boundary conditions of \( \eta(g) \) and the properties of \( \Delta \phi(g) \).

Discussion and criticism of Eq. (B-17), and of the importance of its various constituent values is contained in the text.
APPENDIX C

REVIEW OF DEVELOPMENT OF DROP GROWTH EQUATIONS

The following development follows the method of Oswatitch\(^{145}\) in considering the necessary conservation of mass and energy of growing droplets.

Included are a correction of average incident energy to be \(2RT\) rather than \(3/2RT\), and with generalized non-unity values of both mass and thermal accommodation. An important assumption is that molecular velocity distribution are Maxwellian before and after collision and accommodation with the droplet surface.

Consider a drop of temperature \(T_D\) in an atmosphere \(p\) and \(T\). It has an incident mass flux equal to

\[
\frac{p}{(2\pi RT)^{1/2}}
\]

with a proportion \(\xi\) (condensation coefficient) condensing and the rest being reflected. Since the drop is very small, its temperature is assumed to be constant throughout. The mass transfer from the drop is equal to that which would be incident in an environment \(T_D\) and \(p_D\), the equilibrium values of temperature and pressure, where

\[
\ln \left( \frac{P_D}{p_{\text{sat}}(T_D)} \right) = \frac{2mg}{\rho_L K T r}
\]

Hence, the mass flux per unit area passing out through the surface is

\[
\frac{\xi P_D}{(2\pi RT_D)^{1/2}}
\]

Thus, the conservation of mass may be written

\[
p_L 4\pi r^2 \left( \frac{dr}{dt} \right) = 4\pi r^2 \left[ \frac{\xi P_D}{(2\pi RT)^{1/2}} - \frac{\xi P_D}{(2\pi RT_D)^{1/2}} \right]
\]

or

\[
\frac{dr}{dt} = \frac{\xi P_D}{(2\pi RT)^{1/2}} \left( \frac{p}{T^{1/2}} - \frac{P_D}{T_D^{1/2}} \right)
\]

assuming that \(\xi\) is constant, or at most, a function of the drop temperature only.

Considerations of energy conservation are made after first defining the
the thermal accommodation coefficient, involving the change in temperature of reflected incident molecules.

\[ \alpha = \frac{T_{\text{refl.}} - T}{T_D - T} \quad \text{(Thermal Accommodation Coefficient)} \]  

(C-2)

The energy flux to the drop is, per unit area

\[ \frac{P}{(2\pi RT)^{1/2}} \left[ 2RT \right] \]

and the energy leaving, upon reflection

\[ \frac{(1 - \xi)p}{(2\pi RT)^{1/2}} \left[ 2R \left[ T + \alpha(T_D - T) \right] \right] \]

In addition, the evaporating mass flux carries energy

\[ \frac{\xi p_D}{(2\pi RT_D)^{1/2}} \left[ 2RT_D \right] \]

assuming the evaporating mass flux also to be Maxwellian.

The rate of change of droplet internal energy is

\[ \frac{h}{3} \pi r^3 \rho_L c \frac{dT}{dt} + 4\pi r^2 \frac{dr}{dt} \rho_L U_{fD} \]

\[ U_{fD} = h_{fD} = \frac{\gamma}{\gamma - 1} RT - h_{fg} \]

Hence, conservation of energy requires,

\[ \frac{h}{3} \pi r^3 \rho_L c \frac{dT_D}{dt} + 4\pi r^2 \frac{dr}{dt} \rho_L \left( \frac{\gamma}{\gamma - 1} RT - h_{fg} \right) = \]

\[ \beta(2RT) - \xi \beta_D(2RT_D) - (1 - \xi) \beta 2R \left[ T + \alpha(T_D - T) \right] \]

or

\[ \frac{\rho_L c}{3RT} \frac{dT_D}{dt} = 2\beta \xi \left[ 1 - \frac{\beta_D T_D}{\beta T} \right] - (1 - \xi) \beta \left[ 2\alpha \frac{T_D}{T} - 1 \right] \]

\[ - \frac{\rho_L}{RT} \frac{dr}{dt} \left[ \frac{\gamma}{\gamma - 1} RT - h_{fg} \right] \]

where

\[ \beta = \frac{P}{(2\pi RT)^{1/2}} \quad \beta_D = \frac{P_D}{(2\pi RT_D)^{1/2}} \]

An order of magnitude analysis shows that the left hand term is small compared to the last term on the right hand side. Taking typical values;

\[ r \sim 10^{-7} \text{ cm} \quad \frac{h_{fg}}{c_{p} T} \sim 3 \]

\[ c \sim .3 \text{ cal/gm } ^{\circ}K \quad R \sim .045 \text{ cal/gm } ^{\circ}K \quad T \sim 250^{\circ}K \]

\[ \frac{\gamma}{\gamma - 1} \sim 4 \quad \frac{dT_D}{dr} \sim 7 \times 10^7 ^{\circ}K/\text{cm} \]
\[
\frac{rc}{dr} \frac{dT_D}{dr} = \frac{h_{fg}}{C_p T} \left( \frac{\gamma}{\gamma - 1} \right) (3RT) (\gamma - 1)
\]

This signifies that the energy required to change the droplet temperature during the expansion is much less than the latent heat imparted from condensing incident molecules. Neglecting this term, the energy equation becomes:

\[
0 = \xi(1 - \frac{\theta_D}{\theta}) \frac{T_D}{T} - 2(1 - \xi) \frac{T_D}{T} - 1) + \frac{\rho_L}{\theta} \frac{dr}{dt} \left( \frac{\gamma}{\gamma - 1} \right) (\lambda - 1)
\]

where

\[
\lambda = \frac{h_{fg}}{C_p T}
\]

Since

\[
\frac{dr}{dt} = \frac{\xi}{\rho_L} (\gamma - \theta_D)
\]

The energy equation can be further reduced to

\[
0 = (1 - \frac{\theta_D}{\theta}) \frac{T_D}{T} - 1 - \alpha(\frac{1}{\xi} - 1) \frac{T_D}{T} - 1) + \frac{1}{2} \frac{1}{\gamma - 1} (\lambda - 1)
\]

(C-3)

Solution to the drop growth equations involves an iterative determination of drop temperature, \(T_D\), from Eq. (C-3) and the resulting value of growth rate from Eq. (C-1).

An interesting indication of the importance of considering both mass and energy balance may be seen by closer inspection of Eqs. (C-1) and (C-3). It is seen that the requirement for a drop to grow is that

\[
\frac{dr}{dt} = \frac{\xi}{\rho_L} (1 - \frac{\theta_D}{\theta}) > 1 \quad \therefore \frac{\theta_D}{\theta} < 1
\]

(C-4)

Re-arranging Eq. (C-3) into the form:

\[
\frac{\theta_D}{\theta} \left[ \frac{T_D}{T} + \frac{1}{2} \frac{1}{\gamma - 1} (\lambda - 1) \right] = 1 - \alpha(\frac{1}{\xi} - 1) \frac{T_D}{T} - 1) + \frac{1}{2} \frac{1}{\gamma - 1} (\lambda - 1)
\]

we obtain

\[
\frac{\theta_D}{\theta} = \frac{\left[ 1 + \frac{1}{2} \frac{1}{\gamma - 1} (\lambda - 1) \right] - \alpha(\frac{1}{\xi} - 1) \frac{T_D}{T} - 1) }{\frac{T_D}{T} + \frac{1}{2} \frac{1}{\gamma - 1} (\lambda - 1)}
\]

(C-5)
It is clear that whenever \( T_D > T \) the denominator of Eq. (C-5) is always greater than the numerator. Hence from Eq. (C-4) the droplet will grow. Even in the most unfavorable case, that of \( a = 0 \), where the drop temperature becomes much hotter than the surrounding environment, the value of \( \beta_D / \beta \) will remain less than unity and the drop will grow. This is really a proof of the intuitive fact that a droplet of size greater than critical radius must continue to grow in a supersaturated environment. Conversely when \( T_D < T \), \( \beta_D / \beta \) is greater than unity and the drop evaporates, supporting our knowledge that evaporating liquids are cooler than their superheated environments.
APPENDIX D.

MODIFICATION OF FOREGOING GAS DYNAMICS, NUCLEATION AND DROP GROWTH EQUATIONS FOR DEPARTURE FROM PERFECT GAS DURING AN ISENTROPIC EXPANSION.

The following corrections introduced into the perfect gas relations to account for departure from perfect gas behavior are based on the empirical observation that compressibility factor $z = \frac{P}{RT}$ is almost constant during an isentropic expansion. This is illustrated by Table 15 which lists the values of $P$, $T$ and $z$ for all of the condensation tests of this investigation. Changes in $z$ generally encountered were less than .5% from stagnation pressure to maximum supersaturation prior to condensation. This in effect allows introduction of "effective" perfect gas relations,

$$Pv = R' T \text{ where } R' = zR$$  \hspace{2cm} (D-1)

and the gas dynamics are affected accordingly:

$$Tds = 0 = C_p \frac{dT}{dp} - vdp$$

$$= C_p \frac{pdv}{Rz} + \frac{vdp}{Rz} - vdp$$

$$= \frac{Y}{Y-1} \left[ - \frac{dp}{p} + \frac{dp}{p} - \frac{Y-1}{Y} \frac{dp}{p} \right]$$

Hence

$$\frac{dp}{\rho} = \frac{dp}{p} \left[ 1 - z \left( \frac{Y-1}{Y} \right) \right] = \frac{dp}{p} \left( \frac{1}{Y} \right)$$  \hspace{2cm} (D-2)

Hence, the isentropic relations of a perfect gas may be modified by replacing $Y$ with $Y'$ as defined in equation (D-2).

Similarly, the free energy of formation of a nucleus is modified as follows. The free energy change from vapor to liquid of $(g)$ molecules (added to the free energy of surface formation in the classical droplet model) is

$$d\Phi = dH - d(TS)$$

If it is assumed that the nucleus forms at environmental temperature.

$$dH = SdT = 0$$

$$d\Phi = -TdS = vdp = zRT \cdot \frac{dp}{p}$$
\[ \phi_2 - \phi_1 = z g k T \frac{\ln \frac{P_2}{P_1}}{\ln \frac{P}{P_\infty}} = -z g k T \frac{\ln \frac{P}{P_\infty}}{\ln \frac{P}{P_\infty}} \]

or, the total change in free energy of formation of a classical model droplet becomes

\[ \phi_2 - \phi_1 = -z g k T \frac{\ln \frac{P}{P_\infty}}{\ln \frac{P}{P_\infty}} + 4 \pi r^2 \sigma \]

By proceeding as in Appendix B, it can be further shown that

\[ \Delta \Phi_{\text{max}} = \frac{1}{3} \pi r^2 \sigma \quad \text{at} \quad r = r^* \]

and

\[ \frac{\ln \frac{P}{P_\infty}}{\ln \frac{P}{P_\infty}} = \frac{2m^0}{P_L z k T r^*} \]

These, plus the corrections to the drop growth equations, may be introduced to the perfect gas equations by the simple substitutions

\[ m' = \frac{m}{z} \]  \hspace{1cm} (D-3)

\[ Y' = \frac{Y}{1 + (\gamma - 1)(1 - z)} \]  \hspace{1cm} (D-4)

In a computation involving condensation of a "real" gas, it is necessary to first determine the real isentrope, determined through use of the equation of state as mentioned in the text.

From this, by use of the equation of state and Equation D2, respectively, \( z \) and \( Y' \) are established at all points of the expansion and it is then a simple matter to determine the value of \( \gamma \) to be entered into the computation. The value of \( z \) is taken as the mean of stagnation and condensation values (indicated in Table 14) and a linear, temperature dependent approximation to \( \gamma \) between the same two points is made,

\[ \gamma = \gamma_a - \gamma_b T(\degree C) \]

where values of \( \gamma_a \) and \( \gamma_b \) are given in Table 15.

Consequently, the computation scheme treats a value of \( \gamma \) which will yield the real isentropic exponent \( \gamma' \) when treated with the corrections (D-3) and (D-4). This matches the isentrope prior to condensation with the best
estimate of local conditions and calculates corrected nucleation and drop
growth rates within the theoretical framework of perfect gas.

Perfect gas calculations may be made by simply setting \( z = 1.00 \), and
entering the desired value of \( \gamma = \gamma_a, \gamma_b = 0 \).

It might be pointed out that the compressibility factor begins to vary
more rapidly after condensation commences, and that the computation scheme
ignores this. Some error in profile shapes may result, but probably are no
more in doubt than those resulting from perfect gas treatment. The value
of this treatment lies in its attempt at establishment of the best estimate
of conditions and behavior at the point of \textit{onset} of condensation.

Another possibility for the consideration of real gas behavior in
condensation would be to completely rewrite the entire processes of nucleation,
drop growth and gas dynamics in terms of an equation of state, as was done
for the non-condensing isentropes and total pressure computations. This
could only be done by greatly increasing the complexity of an already complex
computation and at the cost of excluding the many perfect gas relationships
which so greatly simplify the mathematical treatment of condensation in gas
flow processes.
APPENDIX E. A CLOSED FORM SOLUTION TO THE MOMENTUM INTEGRAL BOUNDARY LAYER EQUATION INCLUDING APPROXIMATE EFFECTS OF STREAMWISE DENSITY VARIATION

In most cases involving the study of boundary layers, effects of changing density are negligible. However, in such cases as nozzle flow, where sonic velocities are approached or exceeded, acceleration forces due to the presence of the density gradient become appreciable, and near the throat of a supersonic nozzle are of the same order as those due to area change in their influence on main stream velocity and hence boundary layer growth. A study of the momentum integral equation has revealed that a closed form solution is possible for both laminar and turbulent flow. In the laminar case, an extremely simple form of solution is possible, enabling one to determine estimates of boundary layer growth and depression in supersonic nozzles, using only slide-rule calculations.

If it is assumed that density and pressure vary in the direction of flow, but not across the boundary layer, the momentum integral equation may be written as follows:

\[
\frac{d}{dx} \rho \int_0^\delta (U - u)udy + \rho \frac{dU}{dx} \int_0^\delta (U - u)dy = \tau_{y=0} \tag{E-1}
\]

For the general case, it is assumed that

\[
u/U = f(y/\delta) \quad (E-2a)
\]

\[
\tau_{\infty}/\rho U^2 = a_1 \left(\frac{V}{US}\right)^{1/n} \quad (E-2b)
\]

Then, Equation (1) may be reduced to:
\[ b_1 \frac{d}{dx}(\rho \delta u^2) + b_2 \rho \delta u \frac{du}{dx} = a_1 \left( \frac{v}{U_0} \right)^{1/n} \delta \rho u^2 \]  

(E-3)

where \( b_1, b_2 \) and \( a_1 \) are dependent upon the assumed profile.

Equation (E-3) may be manipulated to appear in the form:

\[ \frac{b_1}{2} \frac{d(\delta^2)}{dx} + \delta^2 \left[ \frac{2b_1 + b_2}{U} \frac{du}{dx} + \frac{b_1}{\rho} \frac{d\rho}{dx} \right] = a_1 \left( \frac{v}{U_0} \right)^{1/n} \delta \]

or

\[ \frac{d(\delta^2)}{dx} + \delta^2 \left[ \frac{C_1}{U} \frac{du}{dx} + \frac{2}{\rho} \frac{d\rho}{dx} \right] = C_2 \left( \frac{v}{U_0} \right)^{1/n} \delta \]  

(E-4)

where \( C_1 = \frac{2(b_2 + b_1)}{b_1} \) \quad \quad \quad \quad \quad C_2 = \frac{2a_1}{b_1}

Equation (E-4) may be put into integrable form by using the integration factor:

\[ m = C_1/\delta x, \quad \delta x = C_1 \frac{du}{U} + \frac{2d\rho}{\rho} \]

Hence, \( m \) takes the convenient form:

\[ m = U \delta \rho^2 \]  

(E-5)

or, more conveniently \( m = \left( \frac{U}{U_0} \right)^{1/2} \left( \frac{\rho}{\rho_0} \right)^2 \)

and the new form of the differential equation is:

\[ \frac{d}{dx}(m\delta^2) = mQ \]  

(E-6)

where \( Q = C_2 \left( \frac{v}{U_0} \right)^{1/n} \delta \)

Hence

\[ \frac{d}{dx}(m\delta^2) = C_2 m \left( \frac{v}{U} \right)^{1/n} \delta^{1-1/n} \]

\[ = C_2 m \left( 1 - \frac{n-1}{2} \right) \left( \frac{v}{U} \right)^{1/n} \left( m\delta^2 \right)^{\frac{n-1}{2n}} \]

and:

\[ (m\delta^2)^{\left( \frac{n-1}{2n} \right)} \frac{d}{dx}(m\delta^2) = C_2 m \frac{n+1}{2n} \left( \frac{v}{U} \right)^{1/n} \]
Integrating, one finally obtains:

$$
\left[ \frac{n+1}{(m^2)^{2n}} \right]^x_{x_0} = C \frac{(n+1)}{2n} \int_{x_0}^x \frac{n+1}{2n} \frac{v_i}{U} \frac{\partial \rho}{\rho} \, dx
$$

(E-7)

Equation (E-7) is the most general form of solution to the momentum integral equation, approximately valid for a compressible gas in either a laminar or turbulent boundary layer.

It should be noted that the integrating factor $m$ can often be more conveniently expressed as follows:

$$
pdx = \left[ C_1 \left( -\frac{dA}{A} - \frac{d\rho}{\rho} \right) + 2 \frac{d\rho}{\rho} \right]
$$

$$
m = e^{\int pdx} = A^{C_1} \rho^{2-C_1}
$$

or, more conveniently:

$$
m = \left( \frac{A}{A^*} \right)^{-C_1} \left( \frac{\rho}{\rho_o} \right)^{2-C_1}
$$

(E-8)

Laminar Boundary Layer

Considering Equation (E-7) for the case of a laminar boundary layer, where $n = 1$, we immediately have:

$$
\left[ m^2 \right]^x_{x_0} = C_2 \int_{x_0}^x m(\frac{\nu}{U}) \, dx
$$

(E-7a)

Assuming that viscosity $\mu$ is nearly constant and substituting for $m$ with Equation (8) yields:

$$
\delta^2 = \frac{C_2 \nu}{m} \int_{x_0}^x m(\frac{1}{\rho U}) \, dx + \frac{m_o}{m} \delta_o^2
$$

$$
= C_2 \nu \left( \frac{A}{A^*} \right)^{C_1} \left( \frac{\rho}{\rho_o} \right)^{C_1-2} \int_{x_0}^x \left( \frac{A}{A^*} \right)^{-C_1} \left( \frac{\rho}{\rho_o} \right)^{2-C_1} \, dx + \frac{m_o}{m} \delta_o^2
$$

Further, for a nozzle: mass flow rate = constant = $\rho^* A^* = \rho^* U^* A^*$

Hence:

$$
\delta^2 = \left\{ \frac{C_2 \nu}{\rho^* U^* A^*} \int_{x_0}^x \left( \frac{A}{A^*} \right)^{1-C_1} \left( \frac{\rho}{\rho_o} \right)^{2-C_1} \, dx \right\} + \frac{m_o}{m} \delta_o^2
$$

(E-9)
Equation (E-9) is the specialized form of Equation (E-7) for a laminar boundary layer. Reviewing Equations (E-3) and (E-4), it is seen that the constants $C_1$ and $C_2$ will depend on the polynomial used to approximate the shape of the boundary layer velocity profile.

For flow in a nozzle, $(A/A^*)$ is known and $(\rho/\rho_0)$ can be easily determined. Hence, the function:

$$F = m\left(\frac{A}{A^*}\right)^{1-C_1} \left(\frac{\rho}{\rho_0}\right)^{2-C_1}$$

may be also easily determined and plotted. The integral may be determined by the area under the curve $F$, and at any point, $x$:

$$\delta^2 = K_1 m^{-1} \int_{x_0}^{x} F \, dx + \frac{m}{m} \delta^2_0$$

For $\delta_0 = 0$ at $x = 0$:

$$\delta^2 = K_1 m^{-1} \int_{x_0}^{x} F \, dx$$  \hspace{1cm} (E-9a)

where $K_1 = \frac{C_2}{\rho^* U^*}$

Equation (E-6) was applied to a nozzle of known geometry, using the assumption:

$$\frac{u}{U} = \frac{2y}{\delta} - \left(\frac{y}{\delta}\right)^2$$

for which $C_1 = 9$ and $C_2 = 30$

At first glance, the assumption of a laminar boundary layer may seem unjustified. However, Launder (R1) has shown that turbulent boundary layers under sufficiently negative pressure gradients will revert to a definite laminar form. Also, for the conditions of this calculation the observed boundary layer at the nozzle throat was such that the boundary layer Reynolds Number was about 2000 at the throat. This is less than the transition value and would indicate that a reversion to a laminar form could have taken place (R1). Hence, the laminar assumption is not unreasonable.
Calculations were made according to Table I for CO₂ at stagnation conditions of 84 psia and -100°F, according to the corresponding values.

\[ F = (A/A*)^{-8} (p/p_o)^{-7} \quad p_o = 84 \text{ psia} \]

\[ K_1 = \frac{30\mu}{\rho*U*} \quad \text{for CO}_2 \text{ at } T_o = -100°F \]

Results of calculations are shown in Figure E1.

At the conditions noted above and for the nozzle in question, limited interferometer data provided visual means of measuring the boundary layer thickness, and four values are shown in Figure E1. These observations are estimated to be accurate ±15%.

Also shown in Figure 1, to assist in estimating the relative size of the boundary layer, are those values of uniform flow flat plate laminar boundary layers calculated for the densities and velocities at the entrance and throat of the nozzle, respectively.

It is seen that the limited experimental data seems to agree well with the approximate calculation of boundary layer thickness. More experimental data is necessary for a more thorough evaluation.

The extremely small boundary layers resulting in the pressure gradients which occur when compressibility becomes important make it appear that further interferometer observations would be an advisable method of gaining such data if it is not readily available in the literature.

The fact that the past "history" of the boundary layer has a greatly decreased effect on the value of boundary layer thickness in the region of rapid acceleration is quite apparent both from the curve in Figure 1 and from inspection of the equations. Equation (E-9) shows that the value of \( \delta^2 \), which exists at the beginning of integration must be multiplied by the ratio \( m_{xo}/m_x \). Hence, if the value of \( m \) increases appreciably, as it does when flow area \( A \) and density \( \rho \) decrease, then the term \( (m_{xo}/m_x)\delta^2 \) contributes
little when added to the integral term.

The fact that one is limited to an assumed boundary layer velocity profile during an integration does not restrict the possibility of breaking the flow into regions, and applying Equation (E-9) to different velocity profiles (i.e. different constants $C_1$ and $C_2$) over the various intervals of $x$, with relative ease.

**Turbulent Boundary Layer**

It is apparent that Equation (E-7) is readily applicable to turbulent boundary layers in its present form, merely by selecting the expressions desired for Equations (E-2a) and (E-2b), that is, the velocity profile and wall shear dependence.

However, since most persons are accustomed to referring to turbulent boundary layer behavior in terms of momentum thickness $\theta$, and shape factor $H = \delta^*/\theta$, it is worthwhile to re-derive Equation (E-7) in terms of $\theta$ and $H$.

$$\frac{d}{dx} \rho \int_0^{\delta} (U - u)dy + \rho \frac{dU}{dx} \int_0^{\delta} (U - u)dy = \tau_o$$

$$\frac{d}{dx} (\rho U^2 \theta) + \rho \delta^* U \frac{dU}{dx} = \tau_0 = a_1 \frac{U^{1/n}}{U^2} \rho U^2$$

This can be written as:

$$\rho U^2 \frac{d\theta}{dx} + \theta \frac{d}{dx} (\rho U^2) + \rho H \theta U \frac{dU}{dx} = a_1 \frac{U^{1/n}}{U\theta} \rho U^2$$

$$\frac{d\theta}{dx} + \theta \left[ \frac{1}{\rho U^2} \frac{d(\rho U^2)}{dx} + H \frac{1}{U} \frac{dU}{dx} \right] = a_1 \frac{U^{1/n}}{U\theta}$$

Again:

$$\frac{d}{dx} (m\theta) = mQ$$

where $Q = a_1 \frac{U^{1/n}}{U\theta}$

and $m = e^\int pdx$

$$pdx = \left[ \frac{d(\rho U^2)}{\rho U^2} + H \frac{dU}{U} \right]$$
\[ m = \left( \frac{\rho U^2}{\rho U_o^2} \right) (\frac{U}{U_o})^H \quad \text{or} \quad \rho U^{H+2} \]

Hence, it is seen that:

\[ (m\delta)^{1/n} \frac{d}{dx} (m\delta) = \frac{n+1}{n} \alpha (\frac{\nu}{U})^{1/n} \]

Integrating yields:

\[ \left[ (m\delta)^{\frac{n+1}{n}} \right]_{x_o}^x = \alpha (\frac{n+1}{n}) \int_{x_o}^x (\frac{\nu}{U})^{1/n} dx \]  \hspace{1cm} (E-7b)

Inspection will also show that Equation (7b) could have been obtained directly from Equation (7), using the relationship between the two integrating factors, and between \( \delta \) and \( \theta \). Note that \( C_1 = 2(H + 2) \).

Substituting for \( m \) in Equation (7b) yields:

\[ (\rho U^{H+2} \delta) \frac{n+1}{n} = \frac{n+1}{n} \alpha (\frac{\nu}{U})^{1/n} \int_{x_o}^x (\frac{\rho U^{H+2} \delta}{U^{1/n}}) \frac{n+1}{n} \left( \frac{\rho U^{H+2} \delta}{U^{1/n}} \right)^{n+1} U^n dx + (\rho U_o^{H+2} \delta) \frac{n+1}{n} \]

This can be further simplified to either of two forms:

\[ \frac{n+1}{n} \theta^{1/n} = (\rho U^{H+2}) \frac{n+1}{n} \left[ \frac{n+1}{n} \alpha (\frac{\nu}{U})^{1/n} \int_{x_o}^x \rho \frac{n+1}{n} U^n dx + C_0 \right] \]  \hspace{1cm} (E-11)

or

\[ \theta^{1/n} = \rho \frac{n+1}{n} U^{-n} \left[ \frac{n+1}{n} \alpha (\frac{\nu}{U})^{1/n} \int_{x_o}^x \rho \frac{n+1}{n} U^n dx + C_0 \right] \]  \hspace{1cm} (E-12)

where

\[ n = (H + 2) \frac{(n+1)}{n} = \frac{1}{n} \]

\[ C_0 = (\rho U^{H+2} \delta) \frac{n+1}{n} \theta_{x = x_o} \]

\[ C_0' = C_0 / \nu^{1/2} \]

For the case of negligible density gradient Equation (E12) reduces to Equation (22.6) in Schlichting

References


R2) Schlichting, H. - Boundary Layer Theory - McGraw-Hill
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<td>.172</td>
<td>.0091</td>
<td></td>
</tr>
<tr>
<td>1.8</td>
<td>1.170</td>
<td>.447</td>
<td>1/3.5</td>
<td>1/.00358</td>
<td>79.8</td>
<td>13.59</td>
<td>22.87</td>
<td>.0147</td>
<td>.336</td>
<td>.0127</td>
<td></td>
</tr>
<tr>
<td>2.0</td>
<td>1.321</td>
<td>.360</td>
<td>1/9.29</td>
<td>1/7.84x10^-4</td>
<td>137.2</td>
<td>21.70</td>
<td>44.57</td>
<td>.00963</td>
<td>.429</td>
<td>.0143</td>
<td></td>
</tr>
<tr>
<td>2.2</td>
<td>1.565</td>
<td>.2835</td>
<td>1/36</td>
<td>6800</td>
<td>189.0</td>
<td>32.62</td>
<td>77.19</td>
<td>.00827</td>
<td>.638</td>
<td>.0175</td>
<td></td>
</tr>
</tbody>
</table>

\[ \delta^2 = \frac{30u}{p\lambda*} K_o = 4.8 \times 10^{-4} K_o \text{ in}^2 \]

\[ \delta = 0.019 \sqrt{K_o} \text{ in} \]

\( \delta^† \)
APPENDIX F

ESTIMATED EFFECT OF DEPARTURE FROM 1-DIMENSIONAL FLOW ON

NOZZLE THROAT PRESSURE MEASUREMENTS

An observed discrepancy between measured density and pressure ratios occurred in the throat region, as mentioned in the text. Measured pressures at the throat tap were about 1.8" Hg higher than the value predicted from density measurements.

The boundary layer estimation presented in Appendix E indicates a relatively rapidly changing boundary layer thickness in the region of the throat. An order of magnitude analysis indicates that this departure from one-dimensionality, coupled with the sonic velocity of the mainstream, causes a considerable lateral pressure gradient, perpendicular to the flat side walls, of a magnitude which might account for the discrepancy.

The estimation of Appendix E yields a boundary layer with the following geometry at the throat

\[ \delta = 0.012" \quad 0.007" \quad 0.013" \]
\[ 0.02" \quad 0.02" \]

Approximating the curve as a segment of circular arc, and approximating displacement thickness by \( \delta^* = 1/3 \delta \) yields

\[ \delta^* = 0.02" \quad 0.02" \]

The radius of displacement thickness curvature is approximately

\[ R \delta = 0.4 = R \left( \frac{0.02}{0.4} \right) \]
\[ R = 8" \]

The transverse pressure gradient across the streamlines is given by

\[ \frac{1}{\rho} \frac{dP}{dn} = \frac{V^2}{R} \]
for which
\[ V = 820 \text{ ft/sec.} \]
\[ \rho = .25 \text{ lbm/ft}^3 \]
\[ R = .67 \text{ ft} \]

Hence, the estimated pressure gradient over the top of the boundary layer at the throat is
\[ \frac{dp}{dn} = \rho \frac{V^2}{R} = 7500 \frac{\text{lbf}}{\text{ft}^3} = 4.33 \frac{\text{psi}}{\text{in}} \]

The side walls are approximately 1" apart. At a point midway between them, the pressure gradient is zero. Assuming the pressure gradient is linear,
\[ \frac{dp}{dz} = 4.33 \]
\[ z = 0 \]
\[ z = 1.0" \]
\[ \frac{dp}{dz} = -4.33 + 8.66z \]

And:
\[ p = p_w - 4.33z + 4.33z^2 \]
\[ \bar{p} = \frac{p_wz - \frac{4.33z^2}{2} + \frac{4.33z^3}{3}}{1} \]
\[ = p_w - .71 \]

Hence:
\[ p_w - \bar{p} = .71 \text{ psi} \]

This is of sufficient magnitude to offer a possible explanation of the less than 1 psi difference between average mainstream and measured wall pressure at the throat.

It is noted that greater rates of curvature of the boundary layer occur upstream of the throat at relatively negligible velocities. Downstream, where velocities continue to increase the boundary layer has very little curvature.
APPENDIX G

DISCUSSION OF INTERFEROMETRY APPLICATION TO TWO-DIMENSIONAL NOZZLE FLOW

Extensive discussions of the theory behind interferometry techniques are numerous. References (14) and (36) are included for the reader who may desire additional detail.

Basically, an interferometer consists of a monochromatic source of light which is focused into a beam of nearly parallel rays. The beam is then split into two beams traveling separate paths of identical distance after which the beams are rejoined and focused on a viewing, or photographic, plate. An interference pattern of parallel light and dark bands, as seen in Fig. 12 is formed when the two beams are rejoined due to the fact that the various rays of each beam travel slightly different distances depending on the geometry of the mirror arrangement.

One beam is passed perpendicularly through the test section. To compensate for the change in optical path length caused by the glass side walls, "compensating" glasses of exactly the same thickness, must be placed in the path of the other beam. During testing, the only change which occurs is the density of the vapor in the test section which influences the velocity of light passing through it according to the relation, for two-dimensional flow:

\[
\frac{S\lambda_0}{\ell} = n_1 - n_2
\]

where \( n \) is the index of refraction of the gas in the test section, \( \ell \) is the width of test section through which light passes, \( \lambda_0 \) is the wave length of light being used, and \( S \) is the number of fringe shifts which occur at the point in question due to a change in the index from \( n_1 \) to \( n_2 \).

The index of refraction is related to the density by the relationship

\[
G_0 = n - 1
\]

where \( G \) is the Gladstone-Dale constant, fixed for any gas.
The technique used in this investigation was to first photograph the interference fringe pattern for no flow and then take another photograph under test conditions. The fringes of each were then numbered from a common point which effectively represented stagnation conditions. For each distance $x$ it was then a simple, though time consuming, matter to determine the fringe shift $S$ and thence to calculate the density ratio. Photos used in calculating the density curves in Fig. 15(a), (b) and Fig. 16(a), (b) are shown in Figs. 11 and 12, respectively. A graphical illustration of the determination of $S$ for test 1 (Fig. 15) is given in Fig. 13. Test 2 was done more precisely by measuring the distances to each fringe in the no-flow picture as measurements in the flow picture proceeded, since it was detected that slight departure from perfectly even spacing of the no-flow fringes occurs, probably due to minute differences in thickness of the optical glasses. (Note differences in Tables 3 and 4.)

Although interferometric methods did not succeed in producing measured effects of condensation, as mentioned in the text, the results obtained for non-condensing flow does offer a recommendation for possible further application in this direction. One important unfavorable aspect of it, however, is the extreme amount of time and effort required to reduce the photographs to meaningful data. Each of the two tests presented here require alignment of the interferometer with the nozzle (a process which may take as long as two days, after which, however, it is permanent), photography, development of negatives and prints (another day), measurements from prints, and calculations based on the measurements. If pressure measurements are taken, coordinating the location of pressure and density measurements also requires painstaking accuracy.

Such a laborious process in reducing the data is still predicated upon a two-dimensional nozzle and becomes prohibitively difficult for three-
dimensional flow. Reference 36 involves the analysis of one three-dimensional nozzle photograph.
APPENDIX H

COMPUTATIONAL PROGRAM

Part 1: Comparison of Equations of State
Part 2: Isentrope and Enthalpy Calculations
Part 3: Normal Shock Calculations
Part 4: Nucleation Contours Calculations
Part 5: Condensation Calculations
PART 1

* M3851-3374, FMS, DEBUG, 1.1.1.14
* XEQ
* C
* COMPARISON OF EQUATIONS OF STATE

READ 4, T, V

C

CALCULATION USING BENEDICT WEBB KUBIN EQUATION OF STATE

R = 0.08207
V = V0*44*16.08
T = T0/1.8
AO = 2.62392
BO = 0.054074
C0 = 0.133670
A = 0.13333
B = 0.0039667
C = 14814.8
ALPHA = 0.00000890
GAMMA = 0.0000557
PBWR = R*T/V + (BO*V*T - A*T - C0/((T*T)))/(V*V) + (5*V*T - A)/(V**3) + A*A
1LPHA/(V**2) + (C*(1.*GAMMA*(V**2)/((T*T*(V**3)))*EXP((1/(-GAMMA)/((V**2)))))
PBWR = PBWR*14.696
PRINT 1, PBWR

C

CALCULATION USING MARTIN AND HOU EQUATION OF STATE

V = V0
T = T0
BV = 0.007495

ATWO = 8.9273631
BTWO = 0.00262476
CTWO = 1.0504757
ATHRE = 1.6907819
BTHR = 0.00704617
CTHR = 0.052424
AFOUR = 0.002112459
BFIVE = 1.9365593*10**(-4)
TC = 1.575

R = 2.24381

PMH = R*T/(V-BV) + (ATWO + BTWO*T + CTWO*EXP(-3.475*T/TC))/(V-BV)**2
1 + (ATHRE + BTHR*T + CTHR*EXP(5.475*T/TC))/(V-BV)**3 + AFOUR
2/(1/(V-BV)**4) + BFIVE*T/(1/(V-BV)**5)
PRINT 2, PMH

C

CALCULATION USING PLANK EQUATION OF STATE

C

FIND LOCATION OF ZERO REMAINDER

R = 1.9644
T = T0/1.8
J = 1
K = 1
PP = PBWR/14.696
DELP = 1*PP

11 VP = R*T/PP + (0.0825*0.012657*PP)*1000*(0.01*T)**3
VP = VP/62.43
IF (VP>V) 9.15, 25
5 IF (1.0001*VP-V) 6.6, 25
6 IF (J-2) 7.8, 8
7 J = 2
8 IF (K-Z) 10, 9, 9
9 DELP = 5*DELP
10 PP = 1./DELP
GO TO 11
25 IF (*9999*VP-V) 15,26,26
26 IF (K-2) 27,28,28
27 K=2
28 IF (J-2) 30,29,29
29 Delp=.5*Delp
30 Pp=Pp+Delp
GO TO 11
15 Pp=14.696*Pp
PRINT 3, Pp
1 FORMAT (15H PRESSURE(WR) =, F12.7)
2 FORMAT (15H PRESSURE(MH) =, F14.7)
3 FORMAT (15H PRESSURE(P) =, F14.7)
4 FORMAT (F5.1, F5.3)
END
PART 2

C MAIN FOR ISENTROPE DECREASING P
DIMENSION MARK(4), XOUT(200), Y(1), TOL(1), YM(1)

1 *DY(1)
COMMON PZERO, HZERO, TZERO
CALL ENT (P + T + H, 4)

100 READ4, PZERO, TZERO, DELTAP, MP

4 FORMAT (3F12.0, 14)

X = 0.
CALL ENT (PZERO, TZERO, Y(1), 3)

H = 0.001
YMIN(1) = 10.

H = DELTAP / 3.
XOUT(1) = X
DO 2 I = 2, MP

2 XOUT(I) = XOUT(I-1) + DELTAP
MARK(I) = 1
MARK(2) = MP
MARK(4) = 0
CALL RUNGE (I, X, Y, TOL, YM, H, XOUT, MARK)
GO TO 100
END

TOTAL 0022

* LABEL
C DIFFEQ FOR ISENTROPE CALCULATION
SUBROUTINE DIFFEQ (N, X, Y, DY)
DIMENSION Y(1), DY(I)
COMMON PZERO

P = PZERO - X
IF (P) 1 = 2

1 CALL EXIT

2 CALL ENT (P + T + Y(1), 2)
CALL FOR RHO (P + T + RHO)
DY(1) = -1. / RHO
RETURN
END

TOTAL 0013
* LABEL
SUBROUTINE PRINT (N,XOUT,Y,DY,J)
C PRINT FOR ISENTROPE CALCULATION
DIMENSION XOUT(200),Y(1),DY(1)
COMMON PZERO,HZERO,TZERO
P=PZERO-XOUT(J)
P=RATIO=P/PZERO
CALL ENT (P,T,Y(1),2)
T=RATIO=T/TZERO
CALL FORRHO (P,T,RHO)
CALL FOR RHO (PZERO,TZERO,RHOZERO)
Z=P/(T*RHO*1.8644)
RATIO=RHO/RHOZERO
G=(P-P SAVE)/(T+T SAVE)/(T-T SAVE)/(1+P SAVE)
GAMMA=G/(G-1)
G=Z*GAMMA/(1-(1-Z)*GAMMA)
VEL=0
W=2*(HZERO-Y(1))*1033*980.7
IF(W) 6 6*7
7 VEL=SQR(F(W)
6 H=Y(1)*.0242048+.142*.60
P SAVE=P
T SAVE=T
IF(J=1) 2 2*3
2 PRINT 5,RATIO,TRATIO,Ratio,P,T,GAMMA,GR,H,Z,VEL
RETURN
3 PRINT 1,RATIO,TRATIO,Ratio,P,T,GAMMA,GR,H,Z,VEL
5 FORMAT (120H1) P/P0 T/T0 RHO/RHO0 P
1 T GAMMA EFF GAMMA H Z VEL
2 ./36X,84H ATM DEG K
3 CAL/GM CM/SEC +/3F12.4+F12.3+F12.1*2F12.4+F12.2*
4 *F12.4+F12.1)
1 FORMAT (3F12.4+F12.3+F12.1*2F12.4+F12.2*2F12.4+F12.1)
RETURN
END

* LABEL
SUBROUTINE FORRHO (P,T,RHO)
R=1.8644
V=R*T/P-(82.5+1.2657*P)/(01*T)**3.333
RHO=1./V
RETURN
END

TOTAL 0035

TOTAL 0007
P IN ATM T IN DEG K H IN ATM-CC/GM
SUBROUTINE ENT (P,T,H,L)
DIMENSION HZERO(130),TH(130)
HOFF(P,T) = 4.333*(82.6*P+1.265*P**1.01*T)**1.333
GO TO (1,2,3,4,5,6)
4 J = 0
7 J = J + 1
C HZERO IN BTU/LB MOLE TH IN DEG R
READS*,HZERO(J),TH(J)
5 FORMAT (2F10.0)
TH(J) = TH(J)/1.8
HZERO(J) = HZERO(J)*52081
IF(TH(J))6,6,7
C NOW HZERO IS IN ATM-CC/GM AND T IS IN DEG K
C ZERO H AT 0 DEG GASEOUS
6 JEND = J - 1
1 RETURN
3 DO 33 J = 1,JEND
IF(T-TH(J))32,33,33
33 CONTINUE
GO TO 100
32 IB = J - 1
H = HZERO(IB)+(HZERO(IB+1)-HZERO(IB))/(TH(IB+1)-TH(IB))*(T-TH(IB))
1 - HOFF(P,T)
RETURN
2 DO 23 J = 1,JEND-5
HJ = HZERO(J) - HOFF(P,TH(J))
IF(HJ-H)23,25,24
23 CONTINUE
J = JEND
24 JH = J - 5
IF(JH)100,100,29
DO 27 K = JH,J
HAK = HZERO(K) - HOFF(P,TH(K))
IF(HAK-H)27,28,28
27 HB = HA
GO TO 100
28 IB = K - 1
T = TH(IB)+(TH(IB+1)-TH(IB))*(H-HB)/(HB-HB)
RETURN
25 T = TH(J)
RETURN
100 PRINT101,P,T,TH
101 FORMAT (/42H T OR H IS OUT OF THE RANGE OF HZERO INPUT
1 */ 12X,2H P,12X,2H T,12X,2H H,/*3F14.2)
CALL EXIT
END

TOTAL 0047*
C     MAIN TO PUNCH ENTHALPY TABLES
DIMENSION HH(6)
CALL ENT(P,T,HH,4)
    T1=T1+60.
        T2=T1+10.
    PUNCH7,T2
1 FORMAT (F17.1)
P=P+5.
    T=T1
7 FORMAT (F16.5)
CALL ENT (P,T,HH(K),3)
    HH(K)=HH(K)*.0242048+.142.60
C     H TO BE PUNCHED TO AGREE WITH DIN I E IC CAL/GM O H AT O SOLID
2 PUNCH3,P,HH
3 FORMAT (F6.1,F11.2)
    CALL EXIT.
END

      0022
TOTAL  0022*
PART 3

* LABEL
SUBROUTINE DIFEQ (N,X,Y,DY)
DIMENSION Y(2),DY(2)
COMMON PZERO
S=1.
P=X
GO TO (1,2)*N
1 P=PZERO-X
S=-1.
2 CALL ENT (P,T,Y(1),Z)
CALL FOR RHO (P,T,RHO)
DY(1)=S/RHO
DY(2)=1.
RETURN
END

TOTAL 0015*

C MAIN FOR SHOCK CALCULATIONS
CALL ENT (P,T,H+4)
PRINT1
1 FORMAT (120H,TOLL,P01,T01,P1,T1)
2 CALL START(1)
END

TOTAL 0008*

* LABEL
SUBROUTINE EQ1(V,REM)
COMMON PZERO,TZERO,HZERO,TOLL,P2,T2,P1,T1,H1
CALL FOR RHO (P1,T1,R1)
U1=SQRT(1.0*(HZERO-H1))
U2=U1*R1/V
P2=P1+R1*U1-V*U2*U2
H2=HZERO-U2*U2*5
CALL ENT(P2,T2,H2,Z)
CALL FOR RHO (P2,T2,R2)
REM=R2-V
RETURN
END

TOTAL 0013*
* LABEL
SUBROUTINE ENT (P,T,H,L)
C P IN ATM T IN DEG K H IN ATM-CC/GM
DIMENSION HZERO(130),TH(130)
HOPFP(P,T)=4.333*(82.6*P+1.265*P*P)/(0.01*T)**3*333
GO TO (1,2,3,4,1,6)
1 J=0
7 J=J+1
C HZERO IN BTU/LB MOLE TH IN DEG R
READS,HZERO(J),TH(J)
5 FORMAT (2F10,0)
TH(J)=TH(J)/1.8
HZERO(J)=HZERO(J)*52081
IF(TH(J)<6.67)
3 C NOW HZERO IS IN ATM-CC/GM AND T IS IN DEG K
C ZERO H AT 0 DEG GASEOUS
6 JFND,J=1
1 RETURN
3 DO 33 J=1,JFND
IF(TH(J)32,33,33
33 CONTINUE
GO TO 100
32 IF=J=1
1 H=HZERO(IB)+(HZERO(IB+1)-HZERO(IB))/(TH(IB+1)-TH(IB))*(T-TH(IB))
1 -HOPFP(P,T)
RETURN
2 DO 23 J=1,JFND,5
H1=HZERO(J)-HOPFP(P,TH(J))
IF(H1-H)23,25,24
23 CONTINUE
J=JFND
24 IFJH)100,100,29
29 DO 27 K=JH,J
HA=HZERO(K)-HOPFP(P,TH(K))
IF(HA-H)27,28,28
27 HB=HA
GO TO 100
28 IFK=1
1 T=TH(IB)+(TH(IB+1)-TH(IB))*(H-HB)/(HA-HB)
RETURN
25 T=TH(J)
RETURN
100 PRINT101,P,T,H
101 FORMAT (/42,H,T OR H IS OUT OF THE RANGE OF HZERO INPUT
1 / 12X*2H P,12X*2H T,12X*2H H,3F14,2)
CALL EXIT
END
TOTAL 0048
SUBROUTINE PRINT (NXOUT, Y, DX, J)
DIMENSION Y(2), DX(2), XOUT(1), R(3), REM(3)
COMMON PZERO, ZERO, HZERO, TOLL, P2, T2, P1, T1, H1
GO TO (3,4), N
3 P1=PZERO-XOUT(J)
CALL ENT (P1, T1, Y(1), 2)
H1=Y(1)
C
FIND FIRST GUESS AT RHO2
CALL FOR RHO(P1, T1, R(1))
CALL FOR RHO(PZERO, TZERO, RZERO)
R(1)=1*01*R(1)
R(2) = R(1)
   DR=R1*(RZERO-R(1))
CALL E01 (R(1), REM(1))
11 R(2) = R(2) + DR
CALL E01 (R(2), REM(2))
IF(REM(1)*REM(2)) 10, 10, 11
10 R(3)=(R(1)+R(2))*5
CALL E01 (R(3), REM(3))
L=1
IF (REM(3)*REM(1)) 12, 13, 13
12 L=2
13 R(L)=R(3)
IF ((R(2)-R(1))*2/R(1)-TOLL) 14, 14, 10
14 CONTINUE
CALL START(2)
4 IF (Y(1)-HzERO) 5, 6, 6
5 XB=Y(2)
HB=Y(1)
RETURN
6 P02=Y(2)-(Y(2)-XB)*(Y(1)-HzERO)/(Y(1)-HB)
CALL ENT (PO2, T02, HZERO, 2)
POUT=PO2/PZERO
POUTA=PI/P02
PRINT7 TOLL, PZERO, TZERO, P1, T1, P2, T2, PO2, T02, POUT, POUTA
7 FORMAT (F10.6*4(F10.3*F10.1*2F10.5))
CALL START(1)
END
SUBROUTINE START(N)
DIMENSION MARK(4), XOUT(200), Y(2), TOL(2), YMIN(2), DY(2)
COMMON PZERO, TZERO, HZERO, TOLL, P2, T2

H= .05
DO 1 J=1,2
MARK(J)=1
1 YMIN(J)=20.
GO TO(100, 200), N

100 READ*, PZERO, TZERO, P1, TOLL
C SET CONSTANTS FOR RUNGE INTEGRATION DOWN THE ISENTROPE X=0.
TOL(1)=TOLL
CALL ENT (PZERO, TZERO, Y(1), 3)
HZERO=Y(1)
MARK(4)=0

4 FORMAT (4F12.0)
XOUT(1)=PZERO=P1
CALL RUNGE (1*X, Y, TOL, YMIN, H, XOUT, MARK)
CALL EXIT

200 X=P2
CALL ENT (P2, T2, Y(1), 3)
MARK(2)=(PZERO-P2)*20 + 20.
TOL(2)=1*
Y(2)=X
XOUT(1)=P2
MAR=MARK(2)

7 DO 7 K=2, MAR
XOUT(K)=XOUT(K-1)+ .05
CALL RUNGE (2*X, Y, TOL, YMIN, H, XOUT, MARK)
CALL EXIT
END

SUBROUTINE FORRHO (P, T, RHO)
R=1.8644
V=R*T/P-(82.5+1.2657*P)/(.01*T)**3.333
RHO=1./V
RETURN
END

TOTAL 0031*

0031

TOTAL 0006*

0006
* LABEL
C PROGRAM FOR RATE CONTOURS
C READ SECTION
DIMENSION R(5), PP(5), FP(3), P(3)
101 READ1, RHOA*RHOB*SURFA*SURFB*WMOL
1 FORMAT (5F12.0)
READ1  TSTART*TEND*DELTAT
READ40,R
40 FORMAT (5E12.2)
PRINT22, RHOA*RHOB*SURFA*SURFB*WMOL*R
22 FORMAT (14H1RATE CONTOURS,/
1 6H RHO=*F8.3, 2H -, F9.6*10H (T DEG C),/
2 8H SIGMA *=F8.3, 2H -, F9.6*10H (T DEG C),/
3 7H WMOL *=F6.1,/
4 24H TABLE CONTAINS P IN ATM,/
5 19H*6H RATE*=E12.3, 4E18.3*10H P INF ,/
6 9H T DEG K,/
7 5H =* )
M=(TEND-TSTART)/DELTAT
T=TSTART+DELTAT
C COMPUTATION
RHOA=RHOA-RHOB*(T-273/16)
SIGMA= SURFA-SURFB*(T-273/16)
CALL PINFIN (T,PINF)
IF (PINF) 20, 20, 74
74 C1=-1.013256E+1.0E16/(1.38*T) *PINF
C1=LOGF(C1)
C1=LOGF(KSORT(FWOMOL*2.0SIGMA/(1.1416*6.025E23))/RHOA+2.0*C1
C2=2.0SIGMA*WOMOL/(82.062*1.013256E*RHOB*T)
C2=C2*C2*1.88*SIGMA*1.0E16/(1.38*T)
DO 19 1=1,5
19 IF(R(J)) 19, 19, 21
21 A=LOGF(R(J))-C1
B=-2.0
C=C2
C CHOP FOR P
P(3)=0.0
FP(1)=C
11 P(3)=P(3)+3.0
IF(P(3)=55.0) 66, 66, 19
66 IF(FP(1)*(P(3)*P(3)*(A+B*P(3))+C))12, 12, 11
2 P(1)=P(3)-3.0
P(2)=P(3)
6 IF(P(1)+P(2))=4.5
IF (P(2)-P(1)=0.05*P(2)) 10, 10, 9
9 L=1
FP(3)=P(3)*P(3)*(A+B*P(3))+C
IF (FP(1)*FP(3)) 4, 4, 5
4 L=2
6 P(L)=P(3)
GO TO 6
10 IF (P(3)=-70.0) 18, 18, 19
18 PP(J)=EXPFP(P(3))*PINF
19 CONTINUE
20 PRINT 33, T, PP, P1NF
33 FORMAT ( F10.2, 9X, 5E18.3, E11.3 )
GO TO 101
END

TOTAL  0060*
C MAIN FOR CONDENSATION

DIMENSION Y(10), TOL(10), YMIN(10), YOUT(100), MARK(5)
COMMON PZERO, TZERO, WMOL, GAMMA, RHOL,
HFG, SURFA, SURFB, XI, ALPHA, AMACH, X, AREA,
ZDADX, P, T, RSTAR, TO, AMASS, AVG, ASTAR,
3 SSAT, INTERP, Z, YDROP, YRATE, GAMMA, GAMB, Y, NPART
4 RTS, INTERP
INTERP=1
CALL AREA(CX, AREA, DADY)

READ 1, PZERO
READ 1, TZERO
READ 1, WMOL
READ 1, Z
READ 1, GAMMA
READ 1, GAMMB
READ 1, RHOL
READ 1, HFG
READ 1, SURFA
READ 1, SURFB
READ 1, XI
READ 1, ALPHA
READ 1, RTM, E
READ 1, TOL(4)
READ 1, TOLL
READ 1, AMACH
READ 3, CNTAM
READ 3, RZERO

1 FORMAT (F 20, 0)
3 FORMAT (E11, 3)
4 PRINT2, PZERO, TZERO, WMOL, Z, GAMMA, GAMMB, RHOL, HFG, SURFA
1 SURFB, XI, ALPHA, RTM, E, TOLL(4), TOLL, AMACH, CNTAM, RZERO
4B H ALPHA=, T14.51//11H RATE=10.**, T16.2//9H TOL(4)=, T18.6//TH TOLL
5=T18.6//9H AMACH1=, T10.2//8H CNTAM=, T12.3//8H RZERO=, T12.3//8H)
15 PZERO=2115.5, PZERO
TZERO=1.8*(TZERO+273, 16)
INTERP=2
XOUT(1)=1
XOUT(2)=AMACH * AMACH
XOUT(3)=2 * XOUT(2)
H=0.01
MARK(1)=1
MARK(2)=30
MARK(4)=0
DO 24 I=5, 9
24 TOL(1)=TOLL
DO 81 I=1, 3
81 TOL(1)=0.1
DO 25 I=1, 9
Y(I)=0
25 YMIN(I)=0.00001

PART 5

H14.
WMOL=WMOL/2
Y(1)=25.1327*62.4*CNTAM*
Y(2)=Y(1)*PZERO
Y(3)=Y(2)*PZERO/2.
Y(5)=1.
Y(6)=1.
PZERO=PZERO/2115.
TZERO=TZERO/1.8-273.16
NPART=1
X=0.
N=9
CALL RUNGE(N, X, Y, TOL, YMIN, H, XOUT, MARK)
READ 77, L
77 FORMAT(12)
IF(L) 101,101,100
101 CALL EXIT
END

* LABEL
C SUBROUTINE TO COMPUTE NUCLEATION RATE
SUBROUTINE RATEC(PRESS, TEMP, SIGMA, SSAT, RATE)
DIMENSION Y(10)
COMMON PZERO, TZERO, WMOL, GAMMA, RHOL,
1HFC, SURFA, SURFB, XI, ALPHA, AMACH, X, AREA,
2DADX, P, T, RSTAR, TD, AMASS, AVGR, ASTAR, SSAT
3 INTERP, Z, YDROP, YRATE, GAMMA, GAMB, Y, NPART, RTIMES
A=RSTAR
B=10.**16/(1.380*TEMP)
C=-4.1887*SIGMA*A*A*B
13 D=PRESS*1.01325E6*B
14 E=WMOL/(RHOL*6.025E23)
EE=2.*SIGMA/(3.*1416*WMOL)*6.025E23
AA=2.*LOGF(D)+LOGF(E)+5*LOGF(EE)+C
AA=AA+2.*303.16*
IF(AA) 16,16,17
16 RATE=0.0
GO TO 12
17 CONTINUE
IF(AA=85.0) 18,18,19
19 AA=85.0
18 RATE=EXPF(AA)
12 RETURN
END

TOTAL 0072

TOTAL 0025
* LABEL
 C SUBROUTINE TO COMPUTE DERIVATIVES
 SUBROUTINE DIFEQ (N, X, Y, DY)
 DIMENSION Y(10),DY(10)
 COMMON PZERO, TZERO, WMOL, GAMMA, RHOL, HFG, SURFA, SURFB, XI, ALPH,
 AMACH, X, AREA, 2DAX, P, T, RSTAR, DD, AMASS, AVGR, HSTAR,
 3 SSAT, INTERPZ, YDROP, YRATE, GAMA, GAMB, Y, NPART
 55 TEMP=Y(6)*(TZERO+273.16)
 3 R=0.0478/WMOL
 GAMMA=GAMA-GAMB*(TEMP-273.16)
 FAC=1./(1.+(GAMMA-1.)*(1.-Z))
 GAMMA=GAMA*FAC
 GO TO (1*2,NPART)
 1 DO 3 I=1,9
 3 DY(1)=0.
 DY(5)=-Y(5)/(2.*GAMMA*2*(GAMMA-1.)/GAMMA)
 DY(8)=R*TEMP*DY(5)/Y(5)
 DY(6)=Y(6)*(GAMMA-1.)*DY(5)/(GAMMA*Y(5))
 RETURN
 2 CALL PINFIN (TEMP, PINF)
 PRESS=Y(5)*PZERO
 SSAT=PRESS/PINF
 IF (SSAT=1.) 30, 30, 31
 30 RATE=0
 RSTAR=0
 DROPM=0
 GO TO 32
 31 SIGMA=1. + SURFA-SURFB*(TEMP-273.16)
 RSTAR=2.*SIGMA*/WMOL/(182.062*1.01325E6*RHOL*TEMP*LOG(1.315))
 IF (Y(1)-25.1327) 65, 65, 66
 65 AVG=1.3*RSTAR
 GO TO 67
 66 AVG=SORTF(Z.*Y(3)/Y(1))
 67 CALL RATEC(PRESS, TEMP, SIGMA, SSAT, RATE)
 CALL DROPGC(PRESS, TEMP, SSAT, AVG, DROPG)
 6 DROP=DROPG(Y(7)*SQRTEMP/GAMMA**8.*314E7/WMOL*TEMP)
 VEL=SORTF(-2.*Y(8)*980.7E6)
 DY(9)=1./VEL
 32 CALL AREAC(X, AREA, DADX)
 CONLAB=GAMA-1.0*/GAMMA**HFG*/WMOL/(1544.*1.8*TEMP)**7.78
 D(1)=25.*1327.*RATE*AREA
 D(2)=Y(1)*DROPG+DY(1)*RSTAR
 D(3)=Y(2)*DROPG+12.*56637.*RATE*AREA*RSTAR*RSTAR
 YDROP=RHOL/AMASS*Y(3)*DROPG
 YRATE=RHOL/AMASS**4.*1888*RATE*AREA*RSTAR**3
 DY(4)=YDROP+YRATE
 GO TO 444
 444 I=I+1
 D(1)=Y(5)*(((CONLAB-1.)/(1.-Y(4)))*DY(4)-DADX)/(1.+1.-Y(4))
 1. ((GAMMA-1.)/GAMMA**7.)/(GAMMA*Y(7)/Y(7))
 D(6)=Y(6)*((CONLAB*DY(4)+(GAMMA-1.)/GAMMA*1.-Y(4)))*DY(5)/Y(5)
 D(7)=Y(7)*(-(1.-Y(4))*DY(5)/(GAMMA*Y(7)/Y(7)/Y(5))-1.)
 5*Y(6)*DY(6).
PINFIN FOR CO2 0 TO 240 DEG K

BELOW 93 PINF = C*EXP(-HFG/R/T) WAS USED AND PINF AT 93 USED TO GET C

ABOVE 93 AN EQUATION OF THE FORM P = C*EXP(A/T) WAS USED TO INTERPOLATE

BETWEEN DATA POINTS. DATA TAKEN FROM HANDBOOK OF PHYSICS AND

CHEMISTRY, INTERNATIONAL CRITICAL TABLES, AND MATHESON GAS DATA

SUBROUTINE PINFIN (T,PINF)

IF (T-93.11.1+2

1 A=6.033
B=-1263.
GO TO 10

2 IF (T-140.13.1+4

3 A=7.492
B=-1430.
GO TO 10

4 IF (T-160.15.1+6

5 A=7.0851
B=-1373.
GO TO 10

6 IF (T-195.17.1+8

7 A=6.8355
B=-1333.
GO TO 10

8 IF (T-216.69.1+11

9 A=7.028
B=-1367.
GO TO 10

10 PINF = 10***(A+B/T)
RETURN
END
* LABEL
C SUBROUTINE TO PRINT
SUBROUTINE PRINT(N, XOUT, YOUT, DY, J)
DIMENSION XOUT(1), YOUT(J), Y(10), DY(J)
COMMON PZERO, TZERO, WMOL, GAMMA, RHOL,
  HFG, SURFA, SURFB, XI, ALPHA, AMACH, X, AREA,
  2DADP, P, T, RSTAR, TD, AMASS, AVGRI, ASTAR
3 SSAT, INTERPZ, YDROP, YRAT, GAMA, GAMB, Y*NPART
GAMMA=GAMA/GAMB*YOUT(6)*(TZERO+273.16)
GAMMA=GAMMA/(1.0+(GAMMA-1.0)*(1.0-2.0))
IF (J-2) 21,22,32
21 AMASS=ASTAR*PZERO*YOUT(5)*215.4*SQRTF(GAMMA*WMOL/(32.2*1544.1))
     *(TZERO+273.16)*YOUT(6)*1.081)
AMASS=32.2*654.6*AMASS/(30.48*30.48)
PRINT 10 PSTAR=YOUT(5)
TSTAR=YOUT(6)
GSTAR=GAMMA
RETURN
22 NPART=2
DO 26 I=1,9
26 Y(I)=YOUT(I)
DO 27 I=1,3
27 Y(1)=Y(1)*AMASS
Y(7)=AMACH
A=STAR*Y(5)*AMACH*SqrtF(Y(6)*GSTAR/(TSTAR*GAMMA))
INTERP=3
AREA=A*STAR
CALL AREAC(X, AREA, DADP)
INTERP=2
XOUT(1)=X
DO 25 I=3,63
25 XOUT(I)=XOUT(I)+2
32 CALL AREAC(XOUT(J), AREA, DADP)
AREA=AREA/ASTAR
TEMP=YOUT(6)*(TZERO+273.16)
CALL PINFIN(TEMP, PINF)
PRESS=YOUT(5)*PZERO
SSAT=PRESS/PINF
SICMA=SUMA-SURFS*(TEMP-273.16)
IF(SSAT=1.0) 69,69,67
67 CALL RATEC(PRESS, TEMP, SIGMA, SSAT, RATE)
IF(YOUT(1)-25.1327) 69,69,68
68 AVGR= SQRTF(2.0*YOUT(1)/YOUT(1))
CALL DROPC(PRESS, TEMP, SSAT, AVGR, DROPG)
DTD=TOUT-YOUT(6)*(TZERO+273.16)
PPSF=215.4*PZERO
TDEGR=1.8*(TZERO+273.16)
TOTALN=YOUT(1)*WMOL/PPSF/(25.1327*62.4*AMASS*1544.1*TDEGR)
69 PRINT 11,XOUT(J),AREA,YOUT(5),YOUT(6),SSAT,YOUT(7),YOUT(4),
1 YDROP, YRAT, DTD, AVGRI, RSTAR, RATE, TOTALN
RETURN
10 FORMAT (120H X A/A* P/PO T/TO S/SAT M/MACH MOIST BY GROWTH
1Y GROWTH, NEW PART TD-T AVGR RSTAR NRATE TOTALN
2 */)

H18.
* LABEL
  SUBROUTINE DROPGC(PRESS, TEMP, SSAT, AVGR, DROPG)
  COMMON ZERO, ZER0, WMOL, GAMMAV, RHOL,
  HFG, SURFA, SURFB, XI, ALPHA, AMACH, X, AREA,
  2DAX, P, T, RSTAR, TD, AMASS, AVCR, ASTAR,
  3SSAT, INTERP
  J=1
  P=PRESS*1.01325E6
  T=TEMP
  R=AVGR
  L=0
  D(1)=0
  D(2)=0
  TD=T-1.
  2
  TD=TD+1.
  5
  CALL PINFIN(TD, PINF)
  PD=1.01325E6*PINF*EXPF(RSTAR*T/(TD*R)*LOGF(SSAT))
  GASCON=8.3147/WMOL
  CONLAM=(GAMMA-1.)/GAMMA*HFG*778./(1544.45*1.8*TMP)*WMOL
  BETA=P/SQRTF(6.2832*GASCON*T)
  BETAD = PD/SQRTF(6.2832*GASCON*TD)
  DROPG=X/RHOL*(BETA-BETAD)
  AA = 1. - BETAD*TD/(BETA*T)
  AAA=5*(CONLAM-1.)*GAMMA/(GAMMA-1.)*(1.-BETAD/BETA)
  A = AA+AAA - (1./XI - 1.) *ALPHA*(TD/T-1.)
  GO TO (8,9), J
  8
  J=2
  IF(A) 10,10,9
  10 DROPG=0.
  PRINT1, PD,P,T,A,AA,AAA,AAA),(4)
  11 FORMAT(8E12.3)
  RETURN
  9
  I=1
  IF(A) 2,2,3
  2
  I=2
  D(1)=TD
  D3=D(1)+D(2)
  IF(D3) 4,1,4
  4
  TD=5*(D(1)+D(2))
  L=L+1
  IF(L-20) 5,5,60
  60 RETURN
  6
  RETURN
  END
* 
C 
LABEL 
C 
FIRST ORDER DIFF. EQ. ROUTINE--ADJUSTS STEP SIZE 
C 
SUBROUTINE RUNGE(N, X, Y, TOL, YMIN, H, XOUT, MARK) 
DIMENSION Y(1), YMIN(1), TOL(1), SUB(50), XOUT(1), MARK(1) 
DIMENSION DY(50), YA(50), FA(50), FB(50), FC(50), YKEEP(50) 
KBTHW = 1 
KBIG = 1 
KLOW = 1 
NCOUNT = 15 
J = MARK(1) 
MAX = MARK(2) 
L = MARK(4) 
IF (L) 210, 210, 220 
210 
LTEST = 1 
GO TO 30 
220 
LTEST = 2 
NUM = L 
230 
DO 250 I = 1, N 
250 
SUB(I) = TOL(I)/32*O 
10 
IF (MAX - J) 20, 30, 30 
20 
RETURN 
30 
A = XOUT(J) - X 
B = ABSF(1.E-7 * X) 
IF (A + B) 40, 35, 35 
IF (A - B) 50, 50, 60 
40 
J = J + 1 
GO TO 10 
50 
CALL PRINT(N, XOUT, Y, DY, J) 
J = J + 1 
GO TO 10 
60 
IF (A - 1.5*H) 70, 70, 80 
70 
H = A 
L = L + 1 
GO TO 1000 
80 
IF (A - 3.*H) 90, 1000, 1000 
90 
H = .5*A 
C 
DO RUNGE-KUTTE-MERSON INTEGRATION 
C 
1000 
XA = X + H/3. 
XB = X + .5*H 
CALL DIFFEQ(N, X, Y, DY) 
X = X + H 
DO 1030 I = 1, N 
YKEEP(I) = Y(I) 
FA(I) = H*DY(I) 
1030 
YA(I) = Y(I) + FA(I)/3. 
CALL DIFFEQ(N, XA, YA, DY) 
DO 1040 I = 1, N 
1040 
YA(I) = Y(I) + FA(I)/6. + H*DY(I)/6. 
CALL DIFFEQ(N, XA, YA, DY) 
DO 1050 I = 1, N 
FB(I) = H*DY(I) 
C
1050 YA(I) = Y(I) + .125*FA(I) + .375*FB(I)
CALL DIFFEQ(N, XH, YA, DY)
DO 1060 I = 1, N
FC(I) = H*DY(I)
1060 YI(I) = Y(I) + 1.5*FA(I) - 1.5*Fd(I) + 2.5*FC(I)
CALL DIFFEQ(N, X, YA, DY)
DO 1130 I = 1, N
Y(I) = Y(I) + FA(I)/6. + 666666667*FC(I) + 4*DY(I)/6.
J = Y(I)
IF (ABSFU(I) - YM(I)) = 1130, 1090, 1090
1090 KLOW = 2
E = .2*ABSF(U - YA(I))
IF (E - ABSHI(I)*U) = 1110, 1100, 1100
1100 KBIG = 2
GO TO 1130
1110 IF (E - ABSF(SUB(I)*U) = 1130, 1120, 1120
1120 KBTUN = 2
1130 CONTINUE
GO TO (100, 1135, KLOW
1135 GO TO (1180, 1140, KBIG
1140 NCOUNT = NCOUNT - 1
IF (*NCOUNT = 1150, 1150, 1170
1150 PRINT 1160, X, H
PRINT 1165, (I, Y(I), DY(I), I = 1, N)
RETURN
1160 FORMAT (5XH4,STEP SIZE HALVED 15 TIMES CONSECUTIVELY SINCE LAST PRI
1NT = /29H PROGRAM TERMINATED AT X = , E16,8, 8H, H = , E16,8,
2/3H I = 13X, 4HY(I) , 16X, 5HDY(I) )
1165 FORMAT (13, 7X, 2(E16,8, X))
1170 KBIG = 1
IF (H - B) = 1176, 1172, 1172
1172 X = X - H
H = .5*H
DO 1174 I = 1, N
1174 Y(I) = YKEEP(I)
KBTUN = 1
KLOW = 1
GO TO 1000
1176 M = 15 - NCOUNT
PRINT 1178, M, X, H
PRINT 1165, (I, Y(I), DY(I), I = 1, N)
RETURN
1178 FORMAT (4H14,STEP SIZE BECAME TOO SMALL FOR COMPUTER, /20H IT HAS BE
1EN HALVED , 12, 21H TIMES CONSECUTIVELY, /29H PROGRAM TERMINATED AT
2X = , E16,8, 8H, H = , E16,8, /3H I = 13X, 4HY(I) , 16X,
5HDY(I) )
1180 NCOUNT = 15
GO TO (1190, 1200, KBTUN
1190 H = 2.*H
1200 KBTUN = 1
KLOW = 1
C CHECK FOR INTERMEDIATE PRINT OUT
C
100 GO TO (10, 110), LTEST
110 IF (L) 120, 120, 150
120 PRINT 130, X, H
PRINT 140, (I, Y(I), DY(I), I = 1, N)
L = NUM
GO TO 10
130 FORMAT (5H X = E16.8, 4X, 4HH = E16.8, 11X, 1H1, 13X, 4HY(I),
116X, 5HDY(I))/
140 FORMAT (55X, 13, 7X, 2(E16.8, 4X))
150 L = L - 1
GO TO 10
END
* LABEL
C INTERPOLATION ROUTINE FOR COMPUTING AREA
C DADX IS CONTINUOUS
SUBROUTINE AREAC(XX,AREA,DADX)
COMMON PZERO, TZERO, WMOL, GAMMA, RHOL,
1HFG, SURFA, SURFB, XI, ALPHA, AMACH,XX, AREA,
2DADX,P,T,RSTAR,TD,AMASS,AVGR,ASTAR,
3SSAT, I ITLRP
DIMENSION DD(50),X(50),A(50),D(50),DA(50)
C
C AREAC PART 1
C
C INTERP=1, READ AREA X AND ASTAR
C SET X*DADX TO CORRESPOND TO ASTAR
C ASTAR MUST BE IN AREA DATA LIST
GO TO (1,2,3,4), INTERP
1 I=0
10 I=I+1
READ10, X(I),A(I)
10 FORMAT(2F15.0)
12 READ10, XX,ASTAR
IEND=I-2
C IEND = LAST VALUE OF I USED MINUS 1
AREA=ASTAR
DADX=0.
C FIND IZERO
I=0
13 I=I+1
IF(X(I)-XX) 13,14,14
14 IZERO=I
D(IZERO)=0.
DO 15 I=IZERO,IEND
D(I+1) = 2.*(A(I+1)-A(I))/(X(I+1)-X(I))-D(I)
J=IZERO+I+1
15 D(J)=2.*(A(J+1)-A(J))/(X(J+1)-X(J))-D(J+1)
DO 17 I=2*IEND
17 DD(I) = (D(I)-D(I-1))/(X(I)-X(I-1))
C PLUG X INTO DD
RETURN
C
C AREAC PART 2
C
C GIVEN X OUTPUT IS AREA AND DADX
2 IF(XX-X(I)) 20,23,22
20 I=I-1
21 IF(XX-X(I)) 20,23,23
23 IB=I
19 I=I+1
GO TO 24

22  I=I+1
23  IF(XX-X(I))  25,23,22
25  IA=I
26  IB=I-1
27  C  FIND AREA AND DADX
28  AREA=A(IB) + (XX-X(IB))*(DD(IA)
29    *(XX-X(IB))*5 + D(IB))
30  DADX=(DD(IA)* (XX-X(IB))* D(IB))/AREA
31  RETURN
32  C  AREA  PART 3
33  C  GIVEN  AREA  FIND  X  AND  DADX
34  C  IF  MACH  IS  LESS  THAN  1  LOOK  BEFORE  THROAT
36  I=IZERO
37  IF(MACH-1.0)  30,31,31
39  I=I-1
40  IF (AREA-A(I))  32,32,30
42  IB=I
43  IA=I+1
45  GO TO 34
46  I=I+1
47  IF(AREA-A(I))  35,32,31
49  IA=I
50  IB=I-1
52  C  FIND X AND DADX
54  IF( D(IA)-D(1B))  36,37,35
56  SIGN=1
57  XX= (-D(1B)+SIGN* SQRT(D(1B)*D(1B) -2*DD(IA)*(A(IB)-AREA)))
58      /DD(IA) + X(IB)
59  IF(XX-X(IB))  38,39,39
61  SIGN=-1
62  GO TO 301
64  IF(XX-X(IA))  302,302,38
66  XX=(AREA-A(IB))/D(IB) + X(IB)
68  DADX=D(IB)/AREA
70  RETURN
71  END
SUBROUTINE AREAC (X, AREA, DAX)
COMMON PZERO, TZERO, WMOL, GAMMA, RHOL, HFG, SURFA, SURFB, XI, AL, AM, XX, AREA
1A, DAX, P, T, RSTAR, TD, AMASS, AVGR, ASTAR, SSAT, INTERP
GO TO (12, 2, 31) * INTERP
1 CONTINUE
ASTAR = 1
X = .55
RETURN
C NOZZLE TWO B
2 IF (X - 1.45) 10, 11, 11
10 AREA = 1.4 + .00889 * (X - .55)
DAX = .00889 / AREA
RETURN
11 IF (X - .45) 12, 13, 13
12 AREA = 1.008 + .0218 * (X - 1.45)
DAX = .0218 / AREA
RETURN
13 AREA = 1.082 + .0133 * (X - .45)
DAX = .0133 / AREA
RETURN
3 IF (AREA - 1.008) 30, 30, 31
30 X = (AREA - 1.0) * 112.5 + .55
DAX = .00889 / AREA
RETURN
31 X = (AREA - 1.008) / .0218 + 1.45
DAX = .0218 / AREA
RETURN
END

0028
TOTAL 0028
APPENDIX I

ESTIMATION OF EXPERIMENTAL ERROR

TEMPERATURE MEASUREMENTS

Estimated error (difference between measured and actual temperature) on temperature readings except where otherwise noted is + 0.50, - 1.50°F. This was determined after the following measurements and observations.

1) Temperature of various portions of the ice bath of the reference junction were found to vary by as much as 1°F from the freezing point, 32.2°F, despite its containment in an insulated thermos.

2) A calibration of the potentiometer (Wheatstone bridge type) used for measurements, compared exactly with those readings of the standard copper-constantan conversion tables at each end (32°F and 212°F) of the calibration and showed a maximum variation of + 1.0°F between.

3) The time constant of the thermocouple was measured with the aid of a Sanborn Recorder capable of recording a 50 cps signal with no distortion. Subjecting the thermocouple to various step changes in temperature, it was found that for still water \( \tau = .15 \) sec, for still air, \( \tau = 1.4 \) sec, and for response to the variation of human breath, \( \tau = .6 \) sec. The measured time constant of the potentiometer was \( .9 \) sec \( \pm .2 \) sec.

Based on these observations, a reference temperature of 32.2°F was assumed for all measurements and the variation in ice bath temperature allowed for in the estimated larger possible negative error.

Assuming a linear temperature transient over a 1 minute period, a variation from 190°F to - 50°F would result in a rate change of - 4.0°F/sec. Hence, for the non-condensing curves, this effect alone could contribute an error of about 5°F, in the thermocouple (\( \tau = .6 \)) and potentiometer circuit (\( \tau = .9 \) sec). Hence, in the non-condensing curves, an error of + 4.5°F, - 1.5°F is estimated. In measurements involving condensation, care was taken to let temperature readings reach a relatively constant level before data (photos) were taken. No transient effects are estimated for those cases.
PRESSURE MEASUREMENTS

Pressure readings taken by the mercury manometer board are estimated accurate within ±.1" Hg, this would amount to ± 1.25% error in the pressure ratios near station 18 on the figures for Nozzle I.

Minimum pressure readings for Nozzle II were in the vicinity of 28" Hg, well beyond the region of condensation. Hence, pressure ratios in this vicinity are estimated accurate to within ±0.5%.

In the region of condensation (station 10) pressure levels range near 80" Hg. Hence, error in pressure ratios in this region is estimated at ±0.125%, two orders of magnitude less than the pressure variation caused by condensation at this point.

Error has been introduced into the values of stagnation pressure wherever they have not been directly measured by the manometer board owing to the high pressure level, or the stagnation tank pressure gauge, as occurred several times for Nozzle I. General observations of the behavior of the pressure ratio at the throat have shown that although its value remained remarkably close to .535 for all pressure levels, it had a slight tendency to decrease with increasing pressure. Based on these observations (and others with air, where average p/p₀ measured .523 and the same tendency was noted) it is estimated that the pressure ratio at the throat could have been lowered to a minimum value of .525 at the highest pressure levels. Hence, calculated stagnation pressures are estimated to have an error of ± 1%, ± 2%.

Measurements in Nozzle II used the stagnation pressure gauge for all determinations of stagnation pressure after the gauge was calibrated and found to be accurate to within 1/2 psi at all pressures within the range of experimentation. Consequently, all stagnation pressure readings for Nozzle II are estimated accurate to within 1/2 psi and static pressure readings within ±.1" Hg.
DENSITY

Distance between optical glass side walls in the interferometric measurements was .956 inches. At the wall the boundary layer was about .006" at the throat and somewhat larger downstream (See Figure 12). Within the boundary layer densities are lower than in the mainstream, due to temperature recovery effects. This will cause a small error in the measured interferometric fringe shift data when applied to determination of density in the mainstream. An upper limit on error in density measurements was estimated by assuming a thickness of the order of the displacement thickness (also given an upper limit of \( \delta^* = 1/3 \delta \)) to be of temperature equal to the stagnation temperature.

Near the throat, assuming \( \delta^* = .002" \)

\[
2\delta^* = .004" \quad \quad \quad \quad \quad \quad l = .956" \approx 1.0"
\]

\[
\frac{T_{stream}}{T_{\delta}} = .8 \quad \quad \quad \quad \quad \quad \rho_{\delta} = .8
\]

Hence,

\[
\rho_s - \rho_\delta = .2\rho_s \quad \quad \quad \quad \quad \quad \rho_o - \rho_s = .6\rho_s
\]

The fringe shift due to difference in density over length \( l \) is

\[
S = \frac{8G}{\lambda} (\rho_1 - \rho_2)
\]

Hence,

\[
\frac{S_{\delta}}{S} = \frac{\delta^* (\rho_s - \rho_\delta)}{l (\rho_o - \rho_s)} = \frac{.004 (.2\rho_s)}{.96 (.6\rho_s)} = .13%
\]

Similar estimates further downstream indicated that maximum error in density measurements probably do not exceed .2%.