EXPERIMENTS ON NONEQUILIBRIUM, NONSTATIONARY EXPANSION
OF WATER VAPOUR/CARRIER GAS MIXTURE IN A SHOCK TUBE

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S. P. Kalra

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Abstract

The investigation of condensation process in a nonstationary nonequilibrium expansion of water vapour/carrier gas (ultra pure Nz) mixture is presented here. The density variations, pressure variations and onset of condensation due to such expansion, are monitored at two fixed locations in the driver section of the shock tube by using a laser Fabry-Perot interferometer, a differential interferometer and piezotrons transducer devices. The effect of the cooling rate on the supersaturation at onset of condensation and its delay time (nonequilibrium zone) is determined. An empirical relation is derived between the supercooling and the rate of cooling at onset of condensation. A theoretical analysis is performed using this empirical relation in order to determine the location of the onset of condensation in such expansions. It is shown that the location of the onset is uniquely determined for a given initial driver condition, e.g., (i) relative humidity, $\varphi_4$, (ii) vapour mass fraction $\omega_4$, and (iii) temperature $T_4$. A parametric study, using these 3 parameters is also given. The experimental findings are in agreement with the theoretical prediction of onset.
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acknowledgement</td>
<td>ii</td>
</tr>
<tr>
<td>Abstract</td>
<td>iii</td>
</tr>
<tr>
<td>Table of Contents</td>
<td>iv</td>
</tr>
<tr>
<td>Symbols</td>
<td>v</td>
</tr>
<tr>
<td>1. INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>2. BASIC PHYSICS OF CONDENSATION PROCESS</td>
<td>2</td>
</tr>
<tr>
<td>2.1 Kinetics</td>
<td>2</td>
</tr>
<tr>
<td>2.2 Gas Dynamics</td>
<td>4</td>
</tr>
<tr>
<td>3. EXPERIMENTAL APPROACH</td>
<td>7</td>
</tr>
<tr>
<td>3.1 Condensation Shock Tube</td>
<td>7</td>
</tr>
<tr>
<td>3.2 Diagnostic Techniques</td>
<td>8</td>
</tr>
<tr>
<td>3.3 Experimental Results</td>
<td>9</td>
</tr>
<tr>
<td>4. ANALYSIS</td>
<td>11</td>
</tr>
<tr>
<td>5. DISCUSSION OF THE RESULTS</td>
<td>13</td>
</tr>
<tr>
<td>REFERENCES</td>
<td>15</td>
</tr>
<tr>
<td>FIGURES</td>
<td></td>
</tr>
</tbody>
</table>
SYMBOLS

a  sound speed
b  concentration
c  constant(s)
d  free energy
e  nucleation rate
f  Boltzmann constant
g  latent heat
h  Mach number
i  number of molecules per cluster
j  characteristics line slope
k  pressure
l  gas constant
m  supersaturation
n  time
o  temperature
p  particle velocity
q  condensation front velocity
r  specific volume
s  axial coordinates
t  specific heat ratio
u  density
v  surface tension
w  molecular weight
x  specific humidity
y  relative humidity
\( \varepsilon \) exponent

\( \Delta \) condensation delay \[ \Delta = \frac{T_s - T_c}{T_s} \]

\( \Delta G \) free energy change

\( \Delta T \) supercooling \[ \Delta T = (T_s - T_c) \]

Subscripts and Superscripts

\( c \) condensation state

\( s \) saturation state

\( v \) vapour state

\( l \) initial state in the low pressure section (channel) of the shock tube

\( 4 \) initial state in the high pressure section (chamber) of the shock tube

\( 4, l \) ratio of the specified property between chamber and channel (i.e., \( P_{4} = \frac{P_{4}}{P_{l}} \))

\( * \) critical state in the nucleation process
1. INTRODUCTION

In science and technology we find many problems of interest dealing with the condensation process from its supersaturated state viz: (i) the formation of aerosols, (ii) the application of the cloud chamber technology to nuclear and high energy physics, (iii) the expansion of steam in turbine nozzles, (iv) the expansion of vapour into high vacuum when spacecraft are vented, (v) the expansion of condensable vapour in propulsion devices, (vi) supersonic and hypersonic wind tunnel designs, (vii) two-phase-flows associated with water-cooled nuclear-power-reactor design and many others. Although condensation of vapour occurring from the supersaturation state instead of from its equilibrium saturated state has been observed for almost three-quarters of a century (Refs. 1 and 2), recently renewed efforts are being made to provide quantitative predictions of condensation rates, associated relaxation times and to perform reliable experiments, e.g., Hill (Ref. 3), Deych (Ref. 4), Barschdroff et al (Ref. 5).

Condensation of vapours may be initiated by either a heterogeneous nucleation or a homogeneous nucleation mechanism. In the former case the condensation of vapour takes place on foreign nuclei which act as centres for condensation. Their presence leads to condensation near equilibrium. Homogeneous nucleation results in the absence of foreign nuclei or in fast transient processes where condensation accumulation on foreign bodies remains negligibly small. In this case random collisions may lead to the agglomeration of small numbers of vapour molecules forming clusters. The theory of homogeneous nucleation initiated by Volmer and Weber (Ref. 6) which predicts the rate of formation of clusters of critical size, a size that permits unlimited growth of the cluster. These clusters are formed at a high supersaturated state. The condensation due to homogeneous nucleation may become much delayed with respect to the equilibrium state. In the present investigation, the mechanism of homogeneous nucleation results in a phase change process in a highly supersaturated state.

Various techniques have been used in the past to study both the kinetics and the gas dynamics of condensation reaction in gaseous vapours. From among these, the Wilson's cloud chamber and nozzle expansion, have been used extensively. Oriani and Sundquist (Ref. 7) have discussed cloud chamber measurements where Wegener and Mach (Ref. 8) gave a comprehensive review of condensation in nozzle flows.

Recently, shock tube techniques have been applied for condensation studies owing to its advantages of providing a wide spectrum of cooling rates for a given experimental run, although Wegener and Lundquist (Ref. 9) initiated condensation studies using this technique; it has been used since then by Glass, Patterson (Ref. 10), Homer (Ref. 11), Kung and Bauer (Ref. 12), Kawada and Mori (Ref. 13) in various forms. We are also reporting here an experimental shock tube investigation of water vapour condensation process. The emphasis is on the effect of rate of cooling on various parameters of interest viz condensation rates, condensation delay time*.

*Condensation delay time is defined as the delay in onset of condensation measured from the time of its equilibrium saturation state to the time of condensation (i.e., tc-t_s)
onset of condensation and corresponding supersaturation, etc. The experimental results are analyzed to give an empirical relation between rate of cooling and condensation delay* (from equilibrium temperature $T_s$), i.e.,

$$\Delta = \frac{(T_s - T_c)}{T_s}.$$ 

An important contribution from this study is the coupling of the above experimental results with a theoretical analysis, predicting the onset of condensation, condensation front velocity, and its local Mach number at any location in the rarefaction fan of the shock tube for a given initial condition in the expansion chamber (driver) of the shock tubes. The initial conditions are completely defined by specifying initial relative humidity, $\varphi_4$, vapour mass fraction $\omega_4$ and temperature $T_4$. The parametric study, showing the effect of initial parameters on the onset of condensation path in an x-t diagram has also been performed. Ultra-pure $N_2$ was used as a carrier gas in the present experiments.

2. BASIC PHYSICS OF CONDENSATION PROCESS

2.1 Kinetics

When a vapour is expanded adiabatically its vapour pressure decreases as temperature falls. Generally the vapour pressure falls faster than its pressure and therefore we expect the pressure temperature path of a flow particle (i.e., isentrope) will intersect an equilibrium saturation line, as shown in a P-T plot in Figure 1. If phase change occurs in thermodynamic equilibrium, it should follow a saturation line instead of the isentrope. This may happen if the system undergoes a slow change of state or if foreign nuclei are present in sufficient numbers to act as condensation centres. However, in practice adiabatic expansions are generally fast processes and an isentrope extends into the supersaturated state, until spontaneous condensation by homogeneous nucleation takes place. The condensate centres formed in the supersaturated state are a result of fluctuations in the vapour itself. Some of them grow into critical size clusters and these may grow further. At this point their further growth depends upon various physical processes, i.e., thermal accumulation, mass accumulation, heat conduction into the surrounding vapour and diffusion and related processes. Eventually, aggregation into larger clusters occurs and thermodynamic equilibrium is established and isentropes approach saturation lines.

In analogy with the "Boltzmann factor" for equilibrium distributions, Volmar and Weber wrote the following expression for the concentration of critical clusters:

$$C^* = C \exp \left( \frac{-\Delta G^*}{kT} \right)$$  \hspace{1cm} (1)

where $C$ is the concentration of monomers in the system and $\Delta G^*$ is the work associated with the formation of critical clusters.

The classical nucleation rate equation per unit volume per unit time can therefore be written as (Ref. 6):

*Please note that the condensation delay is defined here in terms of the equilibrium saturation temperature ($T_s$) and the onset of condensation temperature ($T_c$).
\[ I = K \exp \left(-\frac{\Delta G^*}{kT}\right) \tag{2} \]

where \( K \) is a factor and its magnitude depends upon the particular approximation used in deriving the rate equation. The quantity \( \Delta G^* \), which is essentially a free energy of formation of a critical size nucleus, contains the effects of: (a) the free energy change of the vapour molecule in reducing their pressure from \( p \) to flat film value \( p_s \), (b) the change in free energy due to transition from vapour to liquid film, and (c) the surface free energy for the creation of liquid clusters. On combining these contributions to \( \Delta G^* \), it is noted that it has a strong dependence on surface tension and is also a function of supersaturation ratio \( S \), temperature \( T \), and molecular volume \( V_C \).

The condition for thermodynamic equilibrium is

\[ \left( \frac{\partial \Delta G}{\partial n} \right)_{p_t, T} = 0 \tag{3} \]

which is at the maximum of the function \( \Delta G (\Delta G = \Delta G^*) \). Here \( n \) is the number of molecules per cluster. This equilibrium condition is unusual as it is associated with the maximum of \( \Delta G \). Therefore it is an unstable equilibrium, generally known as a metastable state. In analogy with other reactions (e.g., dissociation) an energy barrier exists and it must be overcome before further growth becomes possible.

Using Volmer's approach, Wegener (Ref. 14) has expressed nucleation rate, \( I \), in C.G.S. units

\[ I = 5.4 \times 10^{19} \left( \frac{P}{T} \right)^2 \left( \frac{\sigma}{\rho_c} \right)^{\frac{1}{2}} \exp \left[-17.6 \left( \frac{\sigma}{T} \right)^3 \left( \frac{\mu}{\rho_c} \right)^2 \frac{1}{\ln 2S} \right] \tag{4} \]

We may use Equation (4) to estimate nucleation rates for various supersaturation values. Wegener (Ref. 14) computed for the particular case of water vapour and results are shown in Fig. 2. As is obvious from these estimates shown in Fig. 2, the nucleation rate increases by several orders of magnitude for small change in the supersaturation \( S \). Therefore once appreciable supersaturation is reached, the condensation will be effectively instantaneous. This physical situation we technically call the onset of condensation.

In an expansion cooling process, we define theoretically the onset point as a point of maximum supersaturation. In nozzle and shock tube experiments, the corresponding expansion flows do not depart appreciably from the isentropic flow until the onset is reached. So a small deviation (arbitrary value, say, 1%) from the isentrope will locate the onset point.

Although in presenting the analysis of the experimental results, we need not take into account the above described kinetics explicitly, its consideration helps in understanding the condensation process and for an initial estimation of the onset point for various \( S \) values within the constraints of the developed expansion fan. Representative values calculated by various workers are given in the table below (see Ref. 14):
TABLE

Computed and Observed Critical Supersaturation in Water Vapour for \( J = 1 \) cm\(^{-3}\) sec\(^{-1}\) at \( T = 261^\circ\)K

<table>
<thead>
<tr>
<th>Reference</th>
<th>( S = \frac{P}{P_s} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Theory</td>
<td></td>
</tr>
<tr>
<td>Becker and Doring (1935)</td>
<td>5.28</td>
</tr>
<tr>
<td>Volmer (1939), Frenkel (1946)</td>
<td>5.10</td>
</tr>
<tr>
<td>Sander and Damköhler (1943)</td>
<td>4.44</td>
</tr>
<tr>
<td>Barnard (1953)</td>
<td>4.72</td>
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<tr>
<td>Yang (1963)</td>
<td>4.48</td>
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<tr>
<td>Experiment</td>
<td></td>
</tr>
<tr>
<td>Volmer and Flood (1943)</td>
<td>5.03</td>
</tr>
<tr>
<td>Sander and Damköhler</td>
<td>4.39</td>
</tr>
<tr>
<td>Madonna et al (1961)</td>
<td>4.22</td>
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The empirical results derived in this report from our shock tube experiments implicitly take into account the kinetics of the process and may be obtained by joining the basic kinetics with the fundamental conservation equation of gasdynamics, equation of state and the rate equations. A rigorous theoretical approach on these lines has been adopted by one of the members of our group (J. P. Sislian) whose work will be reported in a separate UTIAS report and as a Ph.D. thesis of this University.

2.2 Gas Dynamics

The rarefaction waves are produced in the driver section of the shock tube due to expansion of gas into a low pressure channel. Any fluid particle in the rarefaction zone experiences a cooling effect whose rate depends upon the location from the origin (i.e., diaphragm). Due to the cooling effect, the saturation ratio of vapour \( S = \frac{P}{P_s} \) increases with time (initially vapour is unsaturated). In this nonstationary expansion fan, homogeneous nucleation induces the phase change mechanism and therefore it does not take place at equilibrium saturation ratio \( S = 1 \). The vapour becomes supersaturated and enters into nonequilibrium state. The nucleation rate also increases with increasing supersaturation \( S \), until it achieves its critical peak value. At this point onset takes place and both the supersaturation, \( S \), and nucleation rate drop very rapidly. The condensation reaction adds heat into the flow field thus an increase in pressure and temperature at the onset point occurs. Until this point in x-t plane, the flow is assumed as an isentropic flow. This is a fair assumption, especially for low mass fractions of water vapour in a mixture.

The shock tube flow is treated as a one-dimensional, nonstationary flow and is illustrated on an x-t diagram for three different cases in Fig. 3: (i) frozen flow; no phase change occurs, (ii) equilibrium flow; phase change takes place at equilibrium saturation characteristics, and
(iii) nonequilibrium flow; phase change occurs in the supersaturation state. The condensation front is shown in the rarefaction fan by a thick line. We may note that the heat addition due to the condensation reaction tends to curve the straight characteristics lines such that their slope changes in the direction of increasing pressure. This is because of the compression effect caused by the additional heat of the condensation reaction. Also it results in an increase in the shock wave strength as shown qualitatively in Fig. 3. The cases of frozen flow and the equilibrium flow are the two limiting cases.

The x-t diagram presented here in Fig. 3 is an ideal case of an instant burst of a diaphragm. In reality there is always a finite opening time for a diaphragm. The appropriate corrections applicable to shock tube flows for the finite opening time of a diaphragm are discussed by Hall et al (Ref. 17) and can easily be applied.

If we assume that the diaphragm is removed instantaneously, the rarefaction head will move at the sound speed corresponding to the initial state of the water vapour-gas mixture in the high pressure chamber. The position of the tail depends on the initial state and the pressure ratio between driver and driven sections. For such an ideal, isentropic expansion, the pressure ratio, density ratio and temperature ratio in the rarefaction fan are given by (Ref. 18):

\[
\frac{P}{P_4} = \left[1 - \frac{\gamma_4-1}{\gamma_4+1} \left(\frac{x}{a_4 t} + 1\right)\right]^{\frac{2\gamma_4}{\gamma_4-1}}
\]

\[
\frac{P}{P_4} = \left(\frac{P}{P_4}\right)^{1/\gamma_4}
\]

\[
\frac{T}{T_4} = \left(\frac{P}{P_4}\right)^{\frac{\gamma_4-1}{\gamma_4}}
\]

where \(\gamma_4\) is the specific heat ratio for the gas (or mixture) in the driver section.

The equilibrium vapour pressure is given by the Clausius Clapeyron equation:

\[
\frac{dP_s}{dT} = \frac{L(T)}{(V_v - V_c)T}
\]

For a thermally perfect gas we have

\[
FV = (R/\mu)T
\]
Since $V_v \gg V_c$ (i.e., specific volume of vapour $\gg$ specific volume of condensates) and assuming latent heat 'L' independent of temperature T, Eqs. 6 and 7 yield

$$\ln P_s = -\frac{L}{RT} + \text{const.} \quad (8)$$

By fitting the data on vapour pressure from standard handbooks, Wegener (Ref. 14) defined the vapour pressure as follows:

$$P_s = 10^{\left(-\frac{A}{T} + B\right)} \quad (9)$$

where

$$A = 2263; B = 6.064 \text{ for } 273^\circ K < T < 395^\circ K; \text{ liquid}$$

and

$$A = 2676; B = 7.582 \text{ for } 175^\circ K < T < 273^\circ K; \text{ solid}$$

By knowing the pressure time history at a given location in the driver section of the shock tube and for a known value of $\gamma_4$, we can calculate the corresponding density and temperature histories at the locations by using Eq. 5. The temperature and pressure profiles, so determined, can now be used to evaluate the supersaturation, $S$, history with the help of Eq. 9 at the corresponding location.

We define the temperature, $T_s$, at which the vapour is saturated and the temperature, $T_c$, at which the onset of condensation occurs. Both these temperatures can be determined from known (experimental) pressure/density profiles. The dimensionless quality, $\Delta$:

$$\Delta = \frac{T_s - T_c}{T_s} \quad (10)$$

is termed the 'condensation delay' because of the fact that there will always be a delay for the onset of condensation unless condensation occurs at equilibrium saturation (i.e., $T_s = T_c$, therefore $\Delta = 0$; no delay). At a finite rate of expansion, condensation sets in the supersaturation state ($T_c < T_s$) so that $\Delta$ is a positive fraction. From the physics of the process we know that the supersaturation will increase in magnitude for higher rates of cooling. Therefore it is a reasonable hypothesis that the condensation delay, $\Delta$, is a monotonically increasing function of rate of cooling $\left(-\frac{dT}{dt}\right)$ (Ref. 19). Therefore

$$\Delta = c \left(-\frac{dT}{dt}\right)^{\epsilon} \quad (11)$$

where the minus sign indicates that temperature decreases with time.
We can determine the constant $C$ and exponent $\varepsilon$ from experimental fitting of the data. Since we can also define $\Delta$ and $\frac{\partial t}{\partial t}$ from the shock tube flow equations in the rarefaction fan as functions of $x$ and $t$, which leads to an equation of motion for the propagation of the 'onset of condensation front' in an $x$-$t$ plane. More details for the formulation of above equation are given later in this report in a section for the "Analysis of Experimental Results".

3. EXPERIMENTAL APPROACH

Extensive experimental research has been done on condensation effects in steady expansions which includes the cloud chamber work of Wilson (Ref. 1), Powell (Ref. 20), Pound et al (Ref. 21) and Allen and Kassner (Ref. 22); nozzle expansions by Stodola (Ref. 2), Oswatitsch (Ref. 23), Stein and Wegener (Ref. 24), Dawanson, Wilson, Hill and Russel (Ref. 25), and Kurshakov, Talmanov and Tkalenko (Ref. 26). Their investigations were primarily directed towards condensation kinetics.

Worthy of detailed study are the explosive expansion of steam/water vapour in water-cooled nuclear power reactors resulting from an accidental break in a large pipe (similar to the one-dimensional blast situation) and the blast arising from sudden fuel rod excursions (similar to a spherical explosion). Very little theoretical and experimental work has been done to date on these types of nonstationary flow problems. Wegener and Lundquist (Ref. 9) did some light-scattering experiments to observe the condensed phase behind a contact surface in a shock tube. Recently, Kawada and Mori (Ref. 13) investigated the condensation kinetics of refrigerant vapours using a shock tube.

3.1 Condensation Shock Tube

A shock tube facility was built in order to investigate nonstationary, nonequilibrium two-phase flows. The shock tube is made of square stainless-steel tubing (1" x 1" cross-section). The test section is approximately 10" long and 1" wide and has high optical quality glass windows coated with a nonreflecting coating for He-Ne laser light at a wavelength of 6328Å. Two techniques are used to inject water vapour into the system: (i) after evacuating the system to a desired degree, water is injected into a vertical Pyrex glass cup which can also be heated externally, the water surface thus exposed to the vacuum evaporates and the vapour pressure starts rising in the system. When a desired vapour pressure is reached, a valve which isolates both systems is closed. At this point a carrier gas (ultra pure, nitrogen/air) is mixed with water vapour to obtain the desired pressure in the driver section of the shock tube; (ii) the alternative is to bubble the carrier gas through ultra-pure water until the desired relative humidity is obtained in the system. A separate regulator is provided for inserting the carrier gas if more pressure is desired in the driver section. The high-pressure (driver) and low-pressure (driven) sections are separated by a cellulophane diaphragm.

A good vacuum is achieved using a diffusion pump before each experimental run in order to have a high degree of purity for a test mixture. A schematic diagram and a photograph of the facility are shown in Figs. 4 and 5, respectively.
3.2 Diagnostic Techniques

Measurements are performed at two locations in the driver section of the shock tube. The parameters monitored are density, density gradient, pressure and onset of condensation. Two laser interferometers and two piezotron-transducers are used for this purpose. A brief description of these techniques is outlined below.

An external Fabry-Perot (F-P) cavity is formed using two optical semi-transparent flats (flatness $\approx \lambda/50$). Two first-surface mirrors fold the laser beam within the cavity such that the optical length of the cavity is an integer multiple of the laser cavity. This helps to reduce the effect of transverse-mode interference. Folding of the beam within the cavity also increases the sensitivity for measurements by threefold as the beam will make three passes through the test section instead of one. A schematic of the F-P arrangement is shown in Fig. 6a and a photograph in Fig. 6b.

The transmitted intensity $I_T$ of the laser light through the external F-P cavity is a function of the losses within the cavity and can be expressed as follows:

$$
\frac{I_T}{I_0} = \frac{1}{1 + F \sin^2(\frac{\delta}{2})}
$$

where $I_0$ is the input intensity of the laser radiation and $F$ is defined by

$$
F = \frac{4 R_{ef}}{(1 - R_{ef})^2}
$$

and is determined by losses within the cavity as defined by the effective reflectivity, $R_{ef}$, of the cavity. The value of $F$ is constant for a given Fabry-Perot cavity. Normalizing the above, one can write (Kalra, Ref. 27)

$$
\tilde{I}_s = \frac{\cos^2(\frac{\delta}{2})}{1 + F \sin^2(\frac{\delta}{2})}
$$

The value of $\tilde{I}_s$ varies from 1 to 0 as $\delta$ varies from 0 to $\pi$ (corresponding to half a fringe shift). The intensity $\tilde{I}_s$ is modulated with time in accordance with the phase $\delta$, which changes linearly with density, such that:

$$
\delta(t) = \frac{6\pi KL \rho(t)}{\lambda}
$$

where $K$ is the Gladstone-Dale constant, $L$ is the width of the test section, $\lambda$ is the He-Ne laser wavelength (6328 Å) and $\rho(t)$ is the density at each instant of time. Therefore, measurements of the modulation of intensity within the F-P cavity can be used to determine the density profiles through a rarefaction wave. These intensity modulations are measured by a photomultiplier tube (EMI 9558B).
To align and calibrate the Fabry-Perot cavity, one of the flat plates of the cavity is rigidly attached with a piezoelectric crystal tube. An oscillating electrical signal is applied to the crystal which induces translational motion to the flat, thus introducing a path change within the cavity. This path change (or phase change) modulates the output intensity. These modulations are observable only if good alignment is achieved. This piezoelectric crystal oscillator also serves the purpose of calibrating for fractional fringe measurement (Kalra, Ref. 27). Figure 7 shows an output from the oscillator (upper beam) and the modulated intensity (lower beam). The calibration curve and reflectivity effects are shown in Fig. 8a, 8b. Application of this technique for density measurements in strong shock waves in helium has been established by Kalra (Ref. 28).

A differential interferometer is designed by splitting the laser beam into two closely-spaced parallel beams (separation \( \approx 0.1 \text{ cm} \)) and then recombining them with another similar splitter. This system is similar to the Mach-Zehnder interferometer system and registers the density change occurring within 1 mm distance and also a sharp gradient in density. Figure 9 gives the schematic of the differential interferometer.

In addition to the measurements of density and density gradients, these optical devices give very accurate records of the onset of condensation (spatial resolution \( \approx 50 \text{ nsec}, \text{i.e., limits of the recording device} \)).

Two low-pressure piezotron transducers (Kistler Model 206) are used at the same two locations to monitor pressure profiles. These special transducers have very high sensitivities (\( \approx 0.5 \text{ mm Hg/mV} \)) and frequency response (rise time \( \approx 3 \mu \text{sec} \)) but with very low output impedances. Therefore, piezotron couplers (Model 504D) are used to convert the low output impedances into high-output impedances for recording purposes.

The opening time of a diaphragm is also monitored optically by having the laser light fall on a photo diode as soon as the diaphragm bursts. The photodiode is hooked to a device which triggers the oscilloscopes, thus giving an accurate zero time reference for the traces. A typical experimental arrangement including oscilloscope traces are shown in Fig. 10.

3.3 Experimental Results

In the rarefaction fan, the expansion is shown by a decrease in pressure with time from the head of the wave H to the onset of condensation F (Fig. 11a). The sudden release of latent heat due to the phase transformation from vapour/steam to water increases the pressure as depicted by the sudden jump to S, typical of a shock front. The traces, a and c are the output of the two piezotrons transducers located at 17.1 cm and 35.1 cm from the diaphragm, respectively. The pressure jump at the onset of condensation (F to S) at the 17.1 cm location is relatively sharp and more in magnitude compared to the jump at 35.1 cm. This is what is expected as the cooling rates are higher at locations closer to the diaphragm. The differential interferometer (registers density gradients) traces, show a smooth variation near the head of the rarefaction wave and an instant variation at the onset because of the formation of the condensed vapour at the onset point, which induces a large density gradient. The density
history is recorded by F-P laser interferometer. This interferometer records density change of the order of $1.6432 \times 10^{-5}$ gm/cc per fringe shift. It is evident from the traces that initially there are larger density variations for a given time and the change slows down as time progresses. A unique feature of the interferometer is a detection of the onset point shown as a very sharp streak on the oscilloscope tracer. This is due to the fact that a large density gradient at the onset location throws the beam out of alignment temporarily because of the schlieren effect. This again helps us to locate very accurately the onset location. Some additional representative oscilloscope traces are shown in Fig. 11b and 11c. Figure 11d is the oscilloscope recording in dry N2.

It was desired later to monitor onset location for an additional point within the test section in order to have an idea regarding the curvature of the locus of the onset front in x-t plane. For this purpose, the laser beam acting as an input to the differential interferometer was split in two forming an additional 2-beam interferometer. Therefore the experimental traces shown in Figs. 12a, 12b, 12c, 12d and 12e at location 17.1 cm have 3 tracers instead of two. The third tracer is at 26.1 cm and is obtained by chopping the lower trace of the dual beam oscilloscope. All the traces have a time base of 200 μsec/per large division. There is a time delay (500 μsec) between the start of trace at 35.1 cm with respect to the trace at 17.1 cm.

As explained earlier at the onset point we expect an appreciable departure from the isentropic flow (i.e., appreciable heat is added at this point in the flow field). Therefore, the experimental curve departs from the theoretical equilibrium isentropes as expected and as shown in Fig. 13. Initially, there is no appreciable variation in the experimental and theoretical density profiles. This is because of the very low initial mass fraction of the water vapour ($< 2\%$). However, once condensation occurs the deviation becomes quite marked. Figure 14 shows a density profile.

Two representative runs are plotted in Fig. 15. The onset of the condensation locations are shown on the pressure-temperature diagram along with the equilibrium curve $P_s(T)$ defined by Clausius-Clapeyron equation. It is readily seen that the onset of condensation does not occur at the intersection of the isentrope with the equilibrium $P_s(T)$ curve, but at a supersaturated state.

The temperature and the corresponding supersaturation variation at two locations in an expansion fan are shown in Fig. 16a. The location closer to the diaphragm has a larger rate of cooling and hence large supersaturation is expected. A few more results are also shown in Fig. 16b. The condensation delay time, also called $t_{\text{relax}}$, must depend upon the rate of cooling. To provide a quantitative insight, we did measurements at two locations in the rarefaction zone of the shock tube with different rates. The dispersion of the experimental results exhibiting this effect is shown in Fig. 17. A mean condensation time is thus estimated and is indicated in this plot. The condensation delay times have a strong dependence on the rate of cooling. That is, at the station near the diaphragm the delay is about half of what it is at the station which is twice as far and is independent of the diaphragm pressure ratio $P_4$, as also expected from the analysis discussed in the next section.
From the experimental results, an empirical relation between adiabatic supercooling, $\Delta$, and the rate of cooling $\frac{dT}{dt}$, is derived viz:

$$\Delta = \frac{T_s - T_c}{T_s} = 1.73 \times 10^{-3} \left(- \frac{dT}{dt}\right)^{0.387} \tag{16}$$

where $T_s$ is equilibrium saturation temperature and $T_c$ is the temperature at the onset of condensation. The empirical equation and the experimental results, another relation between supercooling $\Delta T (\Delta T = T_s - T_c)$ and supersaturation $S$ is derived for prediction of the onset. It is given by $\Delta T = 18.321 (S-1)^{0.311}$. Therefore for a given supersaturation, $S$, one can estimate supercooling required to initiate onset within the experimental constraints. As one expects at $S = 1$, i.e., condensation at saturation time will yield $\Delta T = 0$. The experimental and theoretical prediction shown in the above equation are shown in Fig. 19.

4. ANALYSIS

The empirical equation (Eq. 16) expressing the relationship between the adiabatic supercooling of the water-vapour/nitrogen mixture and the cooling rate in the nonstationary rarefaction wave in a shock tube, may be rearranged into an equation for the location of the condensation front in such a rarefaction wave. The flow being isentropic up to the onset of condensation, the cooling rate a particle of the gas mixture would experience on passing through this wave is (Ref. 18):

$$\frac{1}{T_4} \frac{dT}{dt} = 2 \left(\frac{\gamma_4 - 1}{\gamma_4 + 1}\right) \left[1 - \frac{\gamma_4 - 1}{\gamma_4 + 1} \left(\frac{x}{a_4 t} + 1\right)\right] \left[\frac{x}{a_4 t} - \frac{2}{\gamma_4 + 1} \left(\frac{x}{a_4 t} + 1\right)\right] \frac{1}{t} \tag{17}$$

where $T_4$ and $a_4$ denote, respectively, the temperature and the speed of sound of the mixture in the driver section, and $\gamma_4$ is the ratio of specific heats of the mixture and is equal to

$$\gamma_4 = \frac{C_{p1}(1 - \omega_4) + C_{pv} \omega_4}{C_{v1}(1 - \omega_4) + C_{vv} \omega_4} \tag{18}$$

$C_{p1}$, $C_{pv}$, $C_{v1}$, $C_{vv}$ being the specific heats for constant pressure and constant volume of nitrogen and water vapour respectively; $\omega_4$ is the specific humidity in the driver. It can be seen from Eq. 17 that the greatest cooling rate occurs at the head of the wave, where $x/a_4 t = -1$ and becomes unlimited near the origin ($t \rightarrow 0$).

In addition, from Ref. 18

$$\frac{T}{T_4} = \left(\frac{a}{a_4}\right)^2 \quad \text{and} \quad \frac{a}{a_4} = \frac{\gamma_4 - 1}{\gamma_4 + 1} \left(\frac{2}{\gamma_4 - 1} - \frac{x}{a_4 t}\right) \tag{19}$$

hence

$$\Delta = 1 - \frac{T_c}{T_s} = 1 - \frac{\frac{T_c}{T_4}}{\frac{T_s}{T_4}} = 1 - \frac{1}{\left(\frac{2}{\gamma_4 - 1 - \frac{x}{a_4 t}}\right)^2} \tag{20}$$
where \( x_s/a_4 t_s \) is the nondimensional slope of the characteristic along which saturation occurs. The equation of the condensation front is then

\[
1 - \left( \frac{2}{\gamma_4 - 1} \cdot \frac{x}{a_4 t} \right)^2 = 1.73 \times 10^{-3} \left( \frac{dt}{dt} \right) \quad (21)
\]

and \( \frac{dt}{dt} \) can be found from Eq. 17. From the condition that \( \frac{dt}{dt} \to \infty \) as \( t \to \infty \), it follows that the condensation front will tend asymptotically to the characteristic \( x_s/a_4 t_s \) along which equilibrium condensation occurs.

On the other hand, the lowest temperature, the particles of the gas mixture can attain, is reached at the tail of the wave, and the condensation front will begin at some point on the line because of the rarefaction mechanism ceases afterwards. The velocity of the front is obtained from Eq. 21 and is

\[
\frac{dx}{dt} = U = \frac{x}{t} - \frac{0.387}{\gamma_4 - 1} \left[ 1 - \left( \frac{2}{\gamma_4 - 1} \cdot \frac{x}{a_4 t} \right)^2 \right] \quad (22)
\]

\[
0.387 \left\{ 2 \cdot \frac{\epsilon_4}{a_4} \cdot \frac{\epsilon_4}{x t} - \frac{\beta_4}{\gamma_4 - 1} - \frac{x}{a_4 t} + \frac{\beta_4}{\gamma_4 - 1} + \frac{x}{a_4 t} + 1 \right\} + \frac{2}{\gamma_4 - 1} \cdot \frac{x}{a_4 t} \quad (23)
\]

where \( \epsilon_4 = (\gamma_4 - 1)/(\gamma_4 + 1) \) and \( \beta_4 = 2/(\gamma_4 + 1) \). The Mach number relative to the front is

\[
M = \frac{u - U}{a} \quad (23)
\]

u is the particle velocity, and a the speed of sound at the point considered.

Equation (21) shows that the location of the condensation front depends on \( \omega_4 \) (through \( \gamma_4 \) and \( a_4 \)), \( T_4 \) and \( x_s/t_s \). For fixed values of \( T_4 \) and \( \omega_4 \) in the driver section, the ratio \( x_s/a_4 t_s = N_s \) depends only on \( \varphi_4 \), the relative humidity of the water vapour in the driver, in the following way

\[
\varphi_4 = \frac{10}{\gamma_4} \left( 1 - \frac{1}{n} \right) \quad ; \quad \frac{T_s}{T_4} = \frac{T_s}{T_4} = \left[ 1 + \frac{1}{\gamma_4 - 1} (N_s + 1) \right]^2 \quad (24)
\]

the numerator in the expressions for \( \varphi_4 \) being the nondimensional water vapour saturation pressure. Hence the location of the condensation front
depends on $T_4$, $\omega_4$ and $\varphi_4$, i.e., on the parameters which determine the condition of the mixture in the driver section.

The onset equation (21) is numerically solved using Eq. 17 and Newton's iteration technique for the pair of values of $x$, $t$, satisfying Eq. 21. The velocity and Mach number for the onset front are also computed simultaneously from Eqs. 22 and 23. For these computations, $T_4$, $\omega_4$ and $\varphi_4$ are used as input parameters and the propagation of onset of condensation front in x-t plane is determined for a wide range of these parameters. These parameters essentially define the initial state of mixture in the driver section.

Figures 20, 21 and 22 show the location of the condensation front when two of the three parameters $T_4$, $\omega_4$ and $\varphi_4$ are kept fixed and the third is varied. The supersaturation attained in the condensation front and Mach number are shown in Fig. 20 for one particular case. The experimental point for the onset of condensation is also shown for a typical experimental run. The results from this parametric study are indicated in the following section.

5. DISCUSSION OF THE RESULTS

The present investigation of homogeneous (nonequilibrium) condensation of water vapour/nitrogen mixtures in nonstationary rarefaction waves in a shock tube show the following facts:

1. The expansion is isentropic up to the onset of condensation, i.e., within the experimental accuracy the results show no significant departure from well known isentropic flows in a shock tube expansion fan until the onset is reached.

2. In the narrow condensation zone a spontaneous change of state in the gas mixture occurs giving rise to condensation front, after which flow rapidly approaches the equilibrium expansion conditions.

3. Two empirical relations viz (a) between the nondimensional adiabatic supercooling and the cooling rate ($\Delta = \frac{T_S - T_C}{T_S} = 1.73 \times 10^{-3}(\frac{dT}{dt})^{0.387}$) and (b) the supercooling and the supersaturation ($\Delta T = T_S - T_C = 18.321^{0.311}$) are derived from the experimental results.

4. The empirical relation (a) above is inserted in the theoretical expression for the cooling rate in an isentropic expansion in a shock tube; in order to formulate a general analytical expression for the propagation of the onset of condensation front in an x-t plane.

5. The onset of condensation front propagates in x-t plane such that its slope with respect to x-axis decreases until it approaches asymptotically parallel to the saturation characteristics line.

6. The onset of condensation is most sensitive to relative humidity variations in the driver section. An increase in the relative humidity
in the driver section (keeping $T_4$ and $ω_4$ constant) results in a shorter condensation delay time at a given location in the shock tube expansion fan.

7. An increase in the specific humidity $ω_4$ (keeping $T_4$ and $φ_4$ constant) displaces the condensation front such that condensation occurs at lower temperature zone of the rarefaction fan.

8. The variation of $T_4$ alone (while $ω_4$ and $φ_4$ are fixed) has little effect on the location condensation front.

9. The diaphragm pressure ratio, $P_{41}$, across the driver and the channel section of the shock tube has no effect on the location of the condensation front as it does not change the rate of cooling. By increasing $P_{41}$, the amplitude of the rarefaction fan increases and the condensation front line propagates further towards the tail of the fan.

10. The validity of the kinetics of the nucleation process can be examined knowing the functional dependence of critical supersaturation versus temperature, and it should be approximately linear between $Ln s$ and $T^{-3/2}$ (Ref. 14). This dependence is also examined in the present investigation and shown in Fig. 23. This shows a very good agreement for the classical nucleation theory developed by Volmer and Weber (Refs. 6, 16).

11. We find that the shock tube results (nonstationary flow) show a good qualitative agreement with the nozzle flow (steady flow) experiments. But these results extend quantitatively in providing condensation from higher supersaturation state of vapour and varying cooling rates for an experiment of specified initial conditions. As a matter of fact, this extra degree of freedom (i.e., varying rates of cooling) obtained in shock tube flows is because of the additional dimension of time '$t$' which is absent in nozzle flows.
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FIG. 1 A TYPICAL VAPOUR PRESSURE CURVE AND ISENTROPE WITH DEFINITIONS

FIG. 2 NUCLEATION RATES OF LIQUID DROPLETS IN WATER VAPOUR

(REF. 14)
FIG. 3 x-t DIAGRAM FOR A SHOCK TUBE FLOW

(i) WITHOUT CONDENSATION (---)
(ii) WITH NONEQUILIBRIUM CONDENSATION (-----)
(iii) EQUILIBRIUM CONDENSATION WILL OCCUR ALONG Teq CHARACTERISTICS.

SHOCK WAVE AND CONTACT SURFACE ARE SHOWN BY ————.
VACUUM & PRESSURE GAUGES

LASER

WATER VAPOUR INJECTION SYSTEM

HEATER

DIAPHRAGM

MECHANICAL DIAPHRAGM BURSTING DEVICE

GLASS WINDOW

TEST SECTION

STAINLESS STEEL TUBE

PRESSURE SENSORS

GAS CYLINDER

TO VACUUM PUMP

PHOTodiode

FIG. 4 SCHEMATIC OF THE CONDENSATION SHOCK TUBE FACILITY
FIG. 6a  SCHEMATIC OF MULTIPASS FABRY-PEROT INTERFEROMETER
FIG. 6b MULTIPASS FABRY PEROT INTERFEROMETER
UPPER TRACE OSCILLATING MIRROR SIGNAL
LOWER TRACE MODULATED INTENSITY SIGNAL

BOTH TRACES REPRESENT MODULATION OF INTENSITY SIGNAL

FIG. 7 FABRY PEROT FRINGES WITH OSCILLATING MIRROR.
FIG. 8 A  CALIBRATION CURVE

FIG. 8 B  FABRY PEROT NORMALIZED INTENSITY PROFILES FOR VARIOUS EFFECTIVE REFLECTIVITIES OF F.P. CAVITY.
FIG. 9 SCHEMATIC OF THE DIFFERENTIAL LASER INTERFEROMETER
FIG. 10 BASIC ARRANGEMENT FOR STUDYING TWO-PHASE-FLOWS IN A CONDENSATION SHOCK TUBE
FIG. 11a  TYPICAL EXPERIMENTAL RESULTS

\( P_4 = 680 \text{Torr}, \quad R = 8.4 \text{Torr} \)
\( \eta_4 = 92\% \quad T_4 = 20^\circ \text{C} \)

\( P_4 = 680 \text{Torr}, \quad R = 0.25 \text{Torr} \)
\( \eta_4 = 61.7\% \quad T_4 = 20^\circ \text{C} \)

a. PIEZOTRON PRESSURE TRACE AT \( X_1 \)
b. DIFFERENTIAL LASER INTERFEROMETER OUTPUT
c. PIEZOTRON PRESSURE TRACE AT \( X_2 \)
d. FABRY PEROT LASER INTERFEROMETER OUTPUT
FIG. 11 b

Scale 200 μSec/Cm

\[ P_4 = 680 \text{ TORR} \]
\[ y_4 = 84\% \]
\[ T_4 = 292.2 \text{ °K} \]

\[ P_1 = 0.2 \text{ TORR} \]

FIG. 11c

Scale 200 μSec/Cm

\[ P_4 = 580 \text{ TORR} \]
\[ y_4 = 58\% \]
\[ T_4 = 296 \text{ °K} \]

\[ P_1 = 0.15 \text{ TORR} \]

FIG. 11d

Scale 200 μSec/Cm

\[ P_4 = 680 \text{ TORR} \]
\[ y_4 = 0\% \]
\[ T_4 = 294 \text{ °K} \]

\[ P_1 = 0.3 \text{ TORR} \]

DRY NITROGEN RUN

FIG. 11d
$X = 17.1 \text{ Cm}$

$X = 26.1 \text{ Cm}$

$X = 35.1 \text{ Cm}$

**FIG. 12d**

$P_4 = 600 \text{ TORR}$

$y_4 = 88.4 \%$

$T_4 = 297.75 \text{ °K}$

$P_1 = 100 \text{ TORR}$

**Scale 200 \(\mu\text{Sec}/\text{Cm}\)**

---

$X = 17.1 \text{ Cm}$

$X = 26.1 \text{ Cm}$

$X = 35.1 \text{ Cm}$

**FIG. 12e**

$P_4 = 440 \text{ TORR}$

$y_4 = 91.60 \%$

$T_4 = 295.35 \text{ °K}$

$P_1 = 100 \text{ TORR}$

**Scale 200 \(\mu\text{Sec}/\text{Cm}\)**
\( P_0 = 680 \text{ Torr} \quad T_0 = 293.5^\circ \text{K} \)

\( \phi = 61.7\% \)

**Theoretical Isentropes**

- Experimental Points at Location \( X_1 \)
- Experimental Points at Location \( X_2 \)

---

Pressure bump due to condensation and hence departure from isentrope

**FIG. 13** The Theoretical (Equilibrium) Isentropes and Experimental Results

(Pressure History at 2 Locations)
$P_0 = 600 \text{ Torr}$, $T_0 = 293.59 \text{ K}$.
$
u_0 = 61.75$

Experimental points at location X₂.
FIG. 16a TEMPERATURE AND SUPERSATURATION HISTORY OF WATER VAPOUR/N<sub>2</sub> MIXTURE AT TWO LOCATIONS IN A SHOCK TUBE
FIG. 17 DISPERSION OF EXPERIMENTAL RESULTS SHOWING DEPENDENCE OF CONDENSATION DELAY TIME ON THE RATE OF COOLING (I.E., LOCATION IN THE SHOCK TUBE RAREFACTION ZONE)
FIG. 18 ADIABATIC SUPERCOOLING RESULTING CONDENSATION DELAY, Δ, VERSUS THE RATE OF COOLING \( \frac{dT}{dt} \) IN NONSTATIONARY EXPANSION OF WATER VAPOUR/N₂ MIXTURE IN A SHOCK TUBE

Theory: \( \Delta t = 18.32 (S-1)^{0.31} \)

Experiments: ▲

FIG. 19 IMPERICAL RELATION BETWEEN SUPERCOOLING AND SUPERSATURATION IN A NONSTATIONARY EXPANSION OF WATER VAPOUR/N₂ MIXTURE IN A SHOCK TUBE FLOW
$T_4 = 295.3^\circ K$

$\omega_4 = 0.0177$

'1' $\Phi_4 = 97.3\%$

'2' $\Phi_4 = 70.0\%$

'3' $\Phi_4 = 40.0\%$

EXPERIMENTAL ONSET POINT AT SUPERSATURATION=13.75

FIG. 20 x-t DIAGRAM FOR CONDENSATION FRONT SHOWING THE EFFECT OF VARIATION OF RELATIVE HUMIDITY, $\Phi_4$ WITH TEMPERATURE $T_4$ SPECIFIC HUMIDITY, $\omega_4$ ARE KEPT CONSTANT, MACH NUMBER SUPERSATURATION AND AN EXPERIMENTAL ONSET POINT ARE ALSO SHOWN FOR A TYPICAL CASE

FIG. 21 x-t DIAGRAM FOR A CONDENSATION FRONT WHERE $\Phi_4$ AND $T_4$ ARE KEPT CONSTANT AND $\omega_4$ IS VARIED
\( \phi_4 = 97.3\% \)
\( \omega_4 = 0.0177 \)

- \( T_4 = 295.3 \text{°K} \)
- \( T_4 = 320 \text{°K} \)
- \( T_4 = 400 \text{°K} \)

**FIG. 22** x-t DIAGRAM FOR CONDENSATION FRONT WHERE \( \phi_4 \) AND \( \omega_4 \) ARE KEPT CONSTANT AND \( T_4 \) IS VARIED

**FIG. 23** DEPENDENCE OF CRITICAL SUPERSATURATION ON TEMPERATURE
In the investigation of condensation process in a nonstationary nonequilibrium expansion of water vapour/carrier gas (ultra pure N\textsubscript{2}) mixture is presented here. The density variations, pressure variations and onset of condensation due to such expansion, are monitored at two fixed locations in the driver section of the shock tube by using a laser Fabry-Perot interferometer, a differential interferometer and piezotrons transducer devices. The effect of the cooling rate on the supersaturation at onset of condensation and its delay time (nonequilibrium zone) is determined. An empirical relation is derived between the supercooling and the rate of cooling at onset of condensation. A theoretical analysis is performed using this empirical relation in order to determine the location of the onset of condensation in such expansions. It is shown that the location of the onset is uniquely determined for a given initial driver condition, e.g., (i) relative humidity, $q_a$, (ii) vapour mass fraction, $w_a$, and (iii) temperature $T_a$. A parametric study, using these 3 parameters is also given. The experimental findings are in agreement with the theoretical prediction of onset.