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ANALYSIS OF CONDENSATION IN THE CENTERED EXPANSION WAVE IN A SH--ETC(U)
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ANALYSIS OF CONDENSATION IN THE CENTERED EXPANSION WAVE IN A SHOCK TUBE

by

Benjamin J.C. Wu

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Report #35 prepared for Contract N00014-75-C-0263:"Experimental and Theoretical Study of Condensation by Homogeneous Nucleation", (Power Branch, Office of Naval Research, Washington, D.C.).
Submitted by Peter P. Wegener, Principal Investigator.

DEPARTMENT OF ENGINEERING
AND APPLIED SCIENCE

YALE UNIVERSITY

ABSTRACT

A method to determine the onset of vapor condensation by homogeneous nucleation in the unsteady flow in a centered expansion wave generated in a shock tube is described. The test gas, consisting of an inert carrier gas and a vapor, is taken to be an inviscid perfect gas, and the flow is assumed to remain isentropic up to the point of onset of condensation. The latter assumption is valid in experiments where onset is made to occur at the tail of the expansion. Under these conditions the condensation rate is found by combining gasdynamics and the kinetics of nucleation and droplet growth. The method discussed is not limited to a particular choice of the kinetic equations. Finally, we present as an illustration, a procedure with which the nucleation rate in an experiment may be deduced and compared with theory.

NOMENCLATURE

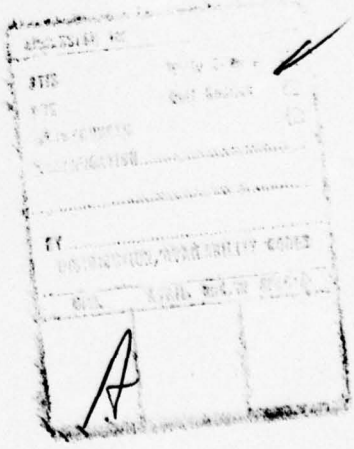
a	sound speed
c	wave velocity (= x/t).
g	mass fraction of condensate
J	nucleation rate
N	droplet concentration
p	pressure
p_∞	saturation vapor pressure over a flat surface.
r	radius
R	universal gas constant
t	time
T	temperature
u	flow velocity
x	distance from center of expansion wave
Y_1, Y_2, Y_3	— defined in Eqs. (13), (15), and (16).
γ	specific heat ratio
Γ	modification to the classical nucleation rate, Eq.(24).
μ	molecular weight
ρ	density
σ	surface tension
τ	temporal variable of integration
χ	spatial variable of integration

Subscript

c	condensate
$c\ell$	classical
o	initial state of expansion
obs	observation station
v	vapor

Superscript

$*$	critical cluster



ANALYSIS OF CONDENSATION IN THE CENTERED EXPANSION WAVE IN A SHOCK TUBE

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INTRODUCTION

The use of shock tubes in studies of vapor condensation began with the experiments of Wegener and Lundquist [1]* who employed the unsteady isentropic expansion of the driver gas resulting from diaphragm rupture to obtain the rapid cooling required in achieving condensation by homogeneous nucleation. Since then, this technique has been extended by a number of investigators: Glass and Patterson [2] obtained time-distance (t-x) records of condensation of water vapor in moist air with a streak camera and schlieren photography. Their results confirmed qualitatively the shape of the condensation onset curve to be expected on a t-x diagram as noted by Heybey and Reed [3]**. Figure 1 is a schematic t-x diagram of such a flow. In the absence of condensation, the characteristics, which are lines of constant thermodynamic state, for the ideal expansion are straight lines centered at the origin [4]. For a given initial vapor pressure, it has been found [5,6] that

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* Numbers in brackets designate References at the end of paper.

** They used the term "condensation shock" instead of the term onset of condensation commonly used today. This latter terminology implies the fact that a shock is often not present in condensing flows.

condensation onset occurs at a larger supercooling (i.e. lower temperature) when the cooling rate is higher, and vice versa. Hence, nearer to the origin, onset occurs closer to the tail of the expansion wave while infinitely far from the diaphragm, onset approaches the saturation point asymptotically. The onset curve terminates at the point K where it intersects the tail of the expansion.

First quantitative experiments on concensation in a shock tube were attempted by Courtney [7], who made static pressure and light scattering measurements at four observation stations along the shock tube driver. Three vapors, H_2O, CH_3OH and $CC\ell_4$, were studied. However, the uncertainties in his experiments precluded the determination of the thermodynamic state at the onset of condensation from his data. Kawada and Mori [8] first reported successful experiments with $CHC\ell_2F$, $CH_3C\ell$, $C_2C\ell_3F_3$, and CH_3OH in a shock tube. In addition to static pressure, they measured the gas density during the expansion using a laser Mach-Zehnder interferometer. The appearance of condensate droplets was detected by light extinction, and experimental condensation onset was defined by these authors as the point where the transmitted light has been attenuated by 5%. Using a similar setup, Kalra [9] did experiments on condensation of water vapor, and critical supersaturation data were obtained. Taking advantage of the relative ease with which the composition and initial temperature of the test gas may be changed in shock tube experiments, Barschdorff [10] studied at our laboratory the carrier gas effects on condensation of water vapor by homogeneous nucleation. Barschdorff's work is probably the first successful use of the shock tube to provide data on the homogeneous nucleation rate of vapor condensation.

It has been recognized that condensation in the expansion wave, where the flow is subsonic in the experiments cited, may cause an upstream influence due to the addition of latent heat to the flow. Thus the gasdynamic state at the observation station may be affected by prior condensation elsewhere in the expansion wave. For example, the state at point A in Figure 1 may have been altered by the condensation and heat addition having occurred between points A and K in Figure 1. Barschdorff noted that at point K, there could be no prior influence of the gasdynamic state since condensation could not have occurred before time t_k. Instead of moving the observation station x_{obs} to coincide with the point K, Barschdorff adjusted the initial pressure in the driven section to move the terminating characteristic so that it intersects with the onset curve at the fixed observation station. In this way* the uncertainty in the state of the gas arising from heat addition effects [11] may be avoided and the onset point may be determined with confidence. This technique has further been used by Lee [12] on H_2O and D_2O, and Wu and Belle [13] to study the binary nucleation of H_2O and C_2H_5OH mixtures.

Other regions of the shock tube flow have been used to produce the environment for test vapors to condense. The shock wave (or the reflected shock) in the driven section is often chosen to heat and vaporize [14] or to decompose [15-18] a substance to produce the test vapor. The temperature behind the shock causing the reaction may be below the

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* Termed "matched expansion in the driver section" by Barschdorff. This technique is especially valuable if one deals with a steel shock tube, where the observation station cannot be moved readily.

saturation temperature of the vapor and, in time, condensation may occur in the uniform flow region following the shock wave [15,17,18]. On the other hand, if the shock temperature is high enough to produce a superheated vapor, the vapor may then be cooled in an expansion wave formed by rupturing a secondary diaphragm [14] or by the interaction of the contact surface with the shock wave reflected from the end plate of the driven section [16]. However,the difficulties in the accurate gasdynamic characteristics of these complex flows and in the determination of the onset of condensation in the relatively gradual condensation process in a uniform flow have made it impossible to date to obtain reliable nucleation rate data from these experiments.

ANALYSIS OF THE CONDENSATION PROCESS IN A CENTERED EXPANSION WAVE.

The one-dimensional flow in a centered expansion fan differs from that in the well-known isentropic expansion occurring in a nozzle mainly in that it is unsteady. Thus, an observer located at a fixed position x_{obs} sees, as time goes on, different fluid elements which have undergone different flow histories. The variation of a flow variable with time as recorded at a fixed position is in fact a succession of instantaneous values taken on by different fluid elements as they flow past this location. To compute the total mass fraction $g(x_{obs}, t)$ of condensate at a given time t at the observation station x_{obs} it is necessary to trace the fluid element back to its initial position and integrate the amount condensed along the particle path.*

Consider a fluid element at (x,t). A droplet of condensate contained in this fluid element must be formed at a

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* The approach here is analogous to Oswatitsch's original treatment [5] of condensation process in steady nozzle flow, discussed by many authors, e.g., Wu [19].

point, say (χ,τ), upstream on the same particle path with an initial radius $r_o(\chi,\tau)$. The droplet will grow to its present size $r(\chi,\tau;\ x,\ t)$ by condensation of vapor molecules on its surface, i.e.,

$$r(\chi,\tau;\ x,t) = r_o(\chi,\tau) + \int_{\tau}^{t} \frac{\partial\ r(\chi,\tau;\ x,t)}{\partial\ t}\ dt. \quad (1)$$

Here, the integration is to be carried out along the particle path passing through (x,t). The integrand $\partial r(\chi,\tau;\ x,t)/\partial t$ may be identified as the droplet growth rate which in turn is a function of the local states of the gas and the droplet.

Let $J(\chi,\tau)$ be the nucleation rate, i.e., the rate of formation of new nuclei of criticsl size per unit volume at (χ,τ); then $J(\chi,\tau)/\rho(\chi,\tau)$ is the number of nuclei formed at (χ,τ) per unit mass of the fluid per unit time, and the mass contained in these droplets is

$$\frac{4\pi}{3}\ r^3(\chi,\tau;\ x,t)\ \ \frac{J(\chi,\tau)}{\rho(\chi,\tau)}\ \rho_c(x,t)\ .$$

Consequently, the mass fraction g of condensate in the fluid element at (x,t) is the integral of all droplets formed before (x,t). Thus,

$$g(x,t) = \frac{\text{mass of condensate}}{\text{total mass of fluid}} = \frac{4\pi}{3}\ \rho_c(x,t) \int_{t_o}^{t} r^3(\chi,\tau;x,t)\ \frac{J(\chi,\tau)}{\rho(\chi,\tau)} d\tau\ . \quad (2)$$

Again, the integration is to be performed along the particle

path. The nucleation rate J and gas density ρ are both functions of the thermodynamic state at (χ,τ), which may be found from gasdynamic considerations. We shall postpone the discussion of the kinetic equations of droplet growth and nucleation to the next section and review briefly the flow in an ideal centered expansion fan without condensation.

The gasdynamic analysis of the expansion flow is substantially simplified if we assume that the flow along the particle path under consideration remains isentropic until the actual onset of condensation. This assumption is valid in experiments done with "matched expansion" technique discussed before. Moreover, it is assumed that the flow is inviscid* and the mixture of vapor and carrier gas remains a thermally and calorically perfect gas. For many vapors, e.g. H_2O, the perfect gas assumption is valid for the relatively low vapor pressures (<0.1% of the critical pressure) typically found in the experiments. If a polyatomic vapor is used it may be necessary to relax the assumption of caloric ideality**

The solution for the centered expansion of a perfect gas is well known [4]. It can be shown that lines of constant (x/t) are the characteristics of this problem on which fluid velocity, temperature, pressure, density, etc., are constant. The local fluid velocity u and speed of sound a are related to the speed of sound a_o in the undisturbed fluid ahead of the expansion wave by (see Fig. 1)

$$u = (c + a_o) \frac{2}{\gamma + 1} . \qquad (3)$$

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* This is an excellent assumption considering the typical Reynolds numbers of 5×10^5.

** This is found to be the case for SF_6 vapor, in which the vibrational modes contribute appreciably to its specific heats even at 200 K.

and

$$a = a_o - \frac{\gamma-1}{2} u \ , \tag{4}$$

where $c = x/t$ and

$$a_o = \sqrt{\gamma \frac{R}{\mu_o} T_o} \ . \tag{5}$$

The other properties of interest are given by the usual isentropic relations,

$$\frac{T}{T_o} = \left(\frac{a}{a_o}\right)^2 = \left(\frac{2}{\gamma+1} - \frac{\gamma-1}{\gamma+1} \frac{c}{a_o}\right)^2 = \left(\frac{p}{p_o}\right)^{\frac{\gamma-1}{\gamma}} = \left(\frac{\rho}{\rho_o}\right)^{\gamma-1} . \tag{6}$$

The particle path of a fluid element may be obtained by integrating the *local fluid velocity*, since by definition,

$$u = \frac{dx}{dt} \Big) \quad \text{along particle path.} \tag{7}$$

Together with Eq.(3), this equation may be integrated to give the required equation of the particle path in the x-t plane,

$$x = + \frac{a_o t}{\gamma-1} \left[2 - (\gamma+1) \left(\frac{t_o}{t}\right)^{\frac{\gamma-1}{\gamma+1}} \right] . \tag{8}$$

The initial conditions chosen are the initial position of the particle x_o and the time, t_o, when the head of the expansion fan reaches x_o (Fig. 1). The head of the expansion fan is also the leading characteristic which propagates into the undisturbed fluid with speed a_o. Therefore, $x_o = - a_o t_o$.

Using the particle path, $x = x(t)$ and $\chi = \chi(\tau)$, one may express the condensate mass fraction g for a particular fluid element identified by its initial position (x_o, t_o) in terms of the single variable t,

$$g(t) = \frac{4\pi\rho_c(t)}{3}\int_{t_o}^{t} r^3(\tau,t)\ \frac{J(\tau)}{\rho(\tau)}\,d\tau\ , \qquad (9)$$

where ρ_c, r, J and ρ are evaluated along this particle path. Likewise, Eq. (1) becomes.

$$r(\tau,t) = r_o(\tau) + \int_{\tau}^{t} \frac{\partial r(\tau,t)}{\partial t}\ dt\ . \qquad (10)$$

Analogous to the treatment [19] of condensing steady nozzle flow, a set of four simultaneous first order differential equations may be obtained by successive differentiation of Eq. (9). Thus,

$$\frac{dg(t)}{dt} = \frac{4\pi\rho_c(t)}{3}\left\{ 3\int_{t_o}^{t} \frac{J(\tau)r^2(\tau,t)}{\rho(\tau)}\ \frac{\partial r(\tau,t)}{\partial t}\,d\tau + \frac{J(t)r^3(\tau,t)}{\rho(t)}\right]_{\tau=t}\right\}. \qquad (11)$$

A significant simplification of Eq.(11) can be made if the drop-growth rate $\partial r/\partial t$ may be taken out of the integral. This is permissible when a suitable average growth rate $\dot{r}(t)$ is assigned to all droplets present in this fluid element, as is commonly done in the solution of condensation in steady nozzle flows [19]. Under this assumption, Eq. (11) becomes

$$\frac{dg(t)}{dt} = 4\pi\ \rho_c\ \dot{r}(t)\int_{t_o}^{t} \frac{J(\tau)r^2(\tau,t)}{\rho(\tau)}\,d\tau + \frac{4\pi\rho_c}{3}\ \frac{J(t)r_o^{\,3}(t)}{\rho(t)}\ , \tag{12}$$

where the quantity $r(\tau,t)$ evaluated at $\tau=t$ has been replaced by the initial radius of the nuclei formed at t. Here, the integral on the right hand side is the total exposed area of all the droplets, hence, the first term on the right hand side is the condensation rate due to growth of all droplets. The second term is that due to the formation of new droplets at t. Letting

$$Y_1(t) \equiv \int_{t_o}^{t} \frac{J(\tau)r^2(\tau,t)}{\rho(\tau)}\ d\tau\ , \tag{13}$$

one gets

$$\frac{dg(t)}{dt} = 4\pi\rho_c\ Y_1(t)\ \dot{r}(t) + \frac{4\pi}{3}\ \rho_c\ \frac{J(t)r_o^{\,3}(t)}{\rho(t)}\ . \tag{14}$$

Similarly, with

$$Y_2(t) \equiv \int_{t_o}^{t} \frac{J(\tau)r(\tau,t)}{\rho(\tau)}\ d\tau, \tag{15}$$

and

$$Y_3(t) \equiv \int_{t_o}^{t} \frac{J(\tau)}{\rho(\tau)}\ d\tau, \tag{16}$$

one gets, by successive differentiation,

$$\frac{dY_1(t)}{dt} = 2\, Y_2(t)\, \dot{r}(t) + \frac{J(t) r_o^2(t)}{\rho(t)} \quad , \tag{17}$$

$$\frac{dY_2(t)}{dt} = Y_3(t)\, \dot{r}(t) + \frac{J(t) r_o(t)}{\rho(t)} \quad , \tag{18}$$

and

$$\frac{dY_3(t)}{dt} = \frac{J(t)}{\rho(t)} \quad . \tag{19}$$

Certain other quantities, e.g., the droplet concentration N, and various averages of the droplets radii, may easily be calculated from the Y's. Thus,

$$N(t) = \rho(t)\, Y_3(t) \quad , \tag{20}$$

the arithmetic mean radius of the droplets is

$$\bar{r}_1(t) = \frac{Y_2(t)}{Y_3(t)} \quad , \tag{21}$$

and the mass mean radius is

$$\bar{r}_3(t) = \left[\frac{3g(t)}{4\pi \rho_c(t)\, Y_3(t)} \right]^{1/3} \quad . \tag{22}$$

METHOD OF SOLUTION

Provided the droplet growth and nucleation rates $\dot{r}$ and J are known as functions of the instantaneous thermodynamic state of the gas and droplets, Eqs. (14), (17), (18), and (19) may be integrated numerically as an initial

value problem yielding g(t) along a particular particle path. It should be noted that these equations are not limited to any particular choice of the kinetic equations. In fact any equations for droplet growth and nucleation (not necessarily homogeneous nucleation) may be used. We shall, however, discuss as an example how these equations have been used by us in evaluating our experiments [6].

When condensation by homogeneous nucleation is considered, the initial radius r_o may be taken [19] to be that of a critical cluster r^{*o} given by the Gibbs-Thomson equation

$$r^* = \frac{2\,\sigma\,\mu_v}{RT\,\rho_c\,\ell n(p_v/p_\infty)} \,. \tag{23}$$

The droplets in a condensate aerosol in the onset zone are found to be very small ($r \lesssim 0.01$ μm) in typical shock tube experiments where the mean free path is of the order of 0.1 μm. Their growth process is hence governed by free molecular flow conditions. Droplet growth laws applicable here are available [6]. In contrast to the droplet growth, there is serious disagreement over the homogeneous nucleation rate equations applicable to condensation of vapors. Here, we shall adopt the approach [6] used in evaluating nozzle data and assume

$$J = \Gamma\, J_{c\ell}. \tag{24}$$

Here, $J_{c\ell}$ is the rate given by the so-called "classical theory" of homogeneous nucleation, and Γ represents the modification to classical theory resulting from statistical mechanics considerations of the nucleation process. The value of Γ is believed to vary only gradually in the condensation zone [6].

To date, a theoretical solution to Γ is not available. Using an experimentally found nucleation rate J_{exp}, we can then determine Γ when the classical rate is calculated. In practice, Γ is found in an inverse fashion by iteration. Thus, constant trial values of Γ are used in Eq. (24) which is in turn substituted into Eqs. (14), (17-19) to calculate g(t). If this calculated g shows an onset of condensation which agrees with experimental observations, the assumed value of Γ is taken to be the solution. Otherwise the procedure is repeated with a different trial value of Γ. This method has been used successfully in evaluating shock tube experiments on condensation of H_2O [10,12], D_2O [12], and $CC\ell_4$ and $CHC\ell_3$ [6,20]. The values of Γ determined in these experiments agree well with those of nozzle experiments.

SUMMARY

In conclusion, we found that using the "matched expansion" technique, the shock tube may be applied to the study of condensation by homogeneous nucleation. The state at the onset of condensation may be determined free from gasdynamic uncertainties due to heat addition affects. A method of analyzing the condensation process in such unsteady expansion flows using any desired droplet growth and nucleation rate equations has been developed and found successful in evaluating experiments with several vapors.

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Stimulating discussions with P.P. Wegener of Yale University are gratefully acknowledged.

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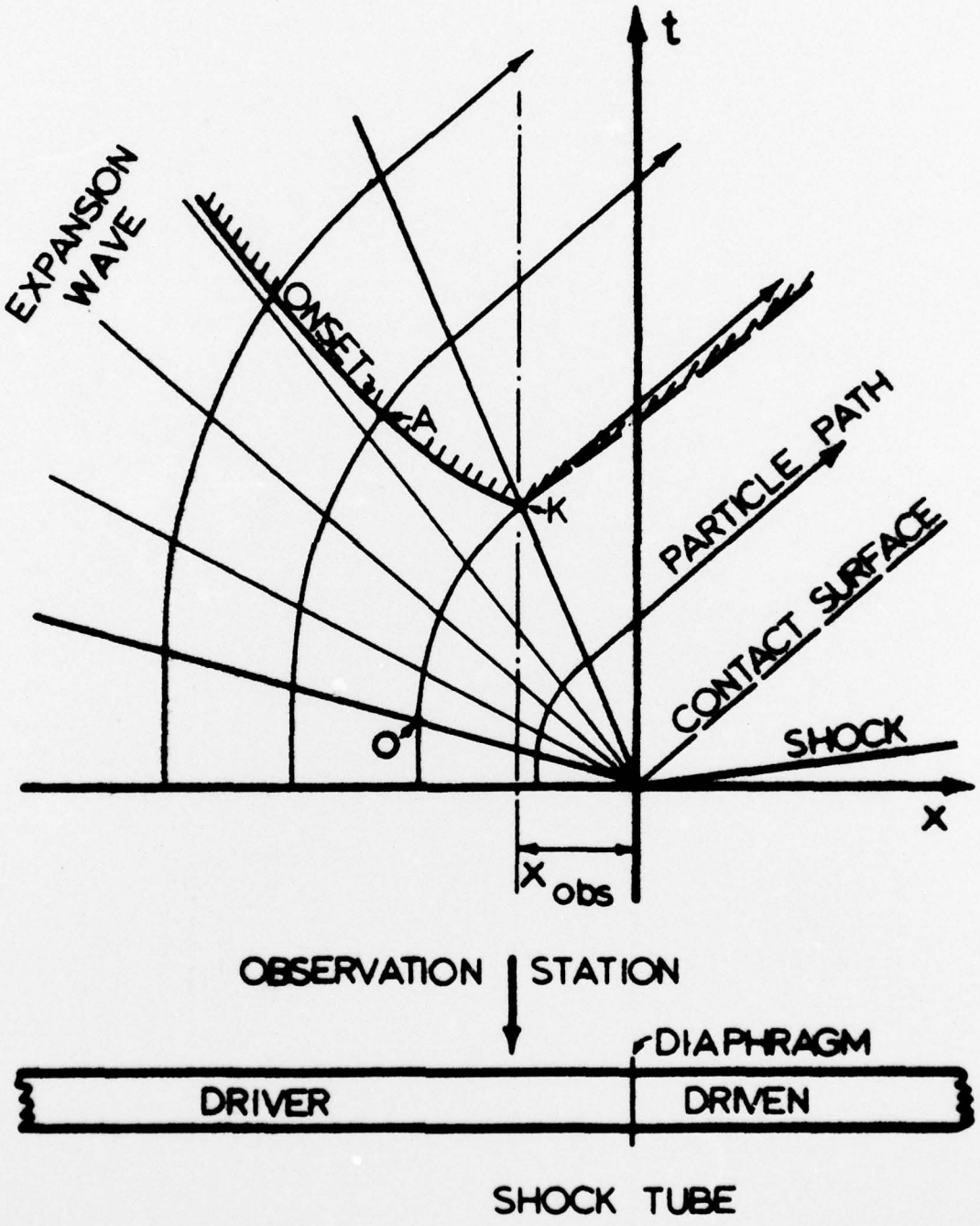
FIGURE CAPTION

Figure 1: Schematic t-x diagram of a centered expansion wave with condensation formed in a shock tube. Point 0 represents (x_o, t_o), and point K, (x_{obs}, t_k).

Figure 1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of the expansion. Under these conditions the condensation rate is found by combining gasdynamics and the kinetics of nucleation and droplet growth. The method discussed is not limited to a particular choice of the kinetic equations. Finally, we present as an illustration, a procedure with which the nucleation rate in an experiment may be deduced and compared with theory.