Analysis of the Heat and Vapor Propagation from the Walls of the Nolan, Pollak and Gardner Type Condensation Nucleus Counters

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ABSTRACT

The measurement of the temperature drop in a dry Pollak condensation nucleus counter by Israel and Nix has been interpreted as an indication that the expansion process does not yield as high a supersaturation as predicted by thermodynamics. An analysis of the heat and vapor diffusion from the walls of the chamber indicates that the counter does indeed develop the supersaturations predicted by thermodynamics in the absence of dropwise condensation and that the natural sensitive time is of the order of 0.3 sec. The measurements of Israel and Nix can be explained in terms of a thermal analysis of the thermocouple itself. The seemingly rapid response of the thermocouple is an indication of the attainment of the steady-state heat flow from the thermocouple and is not an indication that the thermocouple is reading the temperture of the ambient gas accurately. Moreover, the anomalous temperature drop observed about 1 sec after the expansion marks the point at which convection currents sweep away the heated gas which has accumulated immediately adjacent to the thermocouple.

1. Introduction

In the course of early meteorological investigations Coulier (1875), Kiessling (1884) and Aitken (1923) developed the expansion cloud chamber in an attempt to reproduce in the laboratory the conditions under which clouds are formed in the atmosphere. Air samples were sealed in vessels and saturated with water vapor. A sudden adiabatic expansion cooled the gas with the result that the system was supersaturated with respect to water vapor. At small supersaturations cloud formation is dependent upon the presence of condensation nuclei.

C. T. R. Wilson (1897) showed that after repeated expansions on the same gas sample, the chamber finally becomes free of particulate nuclei and larger supersaturations can be used without creating a cloud. Finally, at a supersaturation ratio of about 3.9, ions serve as centers for condensation. Wilson further developed the expansion chamber by adding an electrode to which a potential of several hundred volts could be applied. The purpose of the clearing field was to remove old ions which have diffused away from their original points of formation along the trajectories of high energy charged particles. In this way fresh new ion trajectories could be made visible using the expansion cloud chamber. The trails of droplets were extensively used in the study of the interactions and properties of fundamental particles in cosmic rays. As a result the expansion cloud chamber underwent extensive development as a tool for high energy nuclear physics. Das Gupta and Ghosh (1946) review this development. Relatively little effort has been devoted to developing cloud chambers which are specifically suited for nucleation and condensation experiments. The problems encountered in the study of condensation processes are somewhat different from those encountered in nuclear physics and a systematic effort to identify and solve these problems has not been fully accomplished. This paper attempts to describe the characteristics of a) the heat flow from the walls of long cylindrical cloud chambers commonly employed for measuring condensation nucleus concentrations, and b) the thermal characteristics of fine wire thermocouples used for measuring temperatures inside dry expansion chambers.

A brief review of the processes taking place in the cloud chamber is in order. Expansion chambers are designed most frequently for precise fast expansions from a well defined initial volume to a well defined final volume. The expansion cools the gas while the walls of the chamber remain at ambient temperature. The walls in turn communicate heat to the layer of gas adjacent to them by conduction. However, it would seem at first sight that in large chambers the gas in the center of the chamber would not experience an appreciable rise in temperature for a second or more after the expansion, owing to the relatively slow nature of the heat conduction process. Nevertheless, there is a mechanism which begins to increase the temperature of the center of the chamber immediately following a fast expansion. The layer of heated gas adjacent to the walls expands and compresses the entire chamber, resulting in a general increase in temperature throughout the chamber. This only occurs, of course, when the final volume of the system is fixed. If the chamber is open to the atmosphere, the compressive mechanism is absent but heat conduction from the walls still persists.

Heat conduction from the walls renders the cloud chamber volume heterogeneous in temperature immedi-

ately after the expansion. Since the whole volume is clearly not adiabatic, the frequently used procedure of employing volume expansion ratios in the calculation of final temperatures is a very poor procedure. An alternative procedure gives much better results.

If the chamber is sufficiently large so that heat conduction and vapor diffusion do not affect the central portions of the chamber, during the portion of the expansion which is critical to the experiment, it is irrelevant as to the mechanism by which the central portion is expanded or compressed; it still remains adiabatic. Moreover, Kassner and Schmitt (1966) have shown that the final temperature may be satisfactorily inferred from the following form of the adiabatic law:

$$T=T_1\left(\frac{P_1}{P}\right)^{(1-\gamma)/\gamma}, \tag{1}$$

where P, P_1, T and T_1 are the instantaneous and initial values of the pressure and temperature, respectively, and γ is the ratio of the specific heats or, more appropriately, an *effective adiabatic index*. They give a method for determining the effective adiabatic index. The pressure may be measured continuously throughout the expansion with a flush diaphragm, strain-gage type, pressure transducer and recorded on a light beam oscillograph.

The remaining problem is the determination of the rapidity with which heat propagates into the interior of the chamber. This determines how long after the expansion one can hope to obtain accurate estimates of the temperature from Eq. (1). Such considerations determine the natural sensitive time of the chamber. The sensitive time of the expansion cloud chamber has been investigated by Williams (1939), Hazen (1942), Endt (1948) and Kluyver and Endt (1950) for various modes of operation. However, these results do not relate appropriately to the situation in question in this paper.

If the pressure is monitored precisely throughout the expansion, the heat conduction from the walls may be regarded as a pure conduction problem to a good degree of approximation. The component of pressure increase due to the expansion of the heated boundary layers tends to compress the central portions of the chamber adiabatically. Since the pressure transducer reads the actual pressure, this factor is automatically taken into account. Thus, the diffusion of heat and vapor from the walls tends to destroy the adiabaticity of only those portions of the chamber into which the diffusion field has propagated in appreciable proportions, while those portions which have been only negligibly affected by diffusion remain essentially adiabatic.

In applying the expansion chamber to the measurement of condensation nucleus concentrations, measurements must be made in such a way that observations take place only in the central regions of the chamber and that the observations be concluded prior to the time that diffusion noticeably alters conditions in the observation region.

2. The theory of heat and vapor propagation from the walls of long cylindrical cloud chambers

In the case of the Pollak condensation nucleus counter, the effect of heat and vapor propagation from the walls on the theoretically attained supersaturation will be estimated. By theoretically attained supersaturation we mean that supersaturation calculated from Eq. (1) or an equivalent calculational procedure.

The supersaturation profiles can be obtained directly from the vapor and temperature diffusion profiles with the aid of equilibrium vapor pressure tables. The vapor and temperature profiles can be calculated approximately by neglecting the expansion of the heated gas and by treating the diffusion process as if it were taking place in a low density solid. The expansion of the boundary layers of a heated gas tends to extend the diffusion profiles farther into the central portions of the chamber but this same expansion process also tends to reduce the temperature everywhere along the diffusion profile. The most highly heated regions expand proportionately more than the not so highly heated regions. Moreover, the temperature gradient at the surface would be increased slightly by this effect so that the net propagation of heat from the walls in the actual situation of the nonuniform gas is somewhat greater than in the mathematical model employed where the gas is treated as a low density solid.

The low density solid model for the gas is also customarily employed in droplet growth theory and is dictated principally by the fact that the real problem is so complex that it cannot be handled by conventional methods. Morever, three additional assumptions are employed: 1) the heat capacity of the walls is infinite compared with that of the interior gas, 2) the length of the cylinder is sufficiently great compared to its diameter that it can be regarded as infinite, and 3) convection currents can be regarded as negligible throughout the time interval of interest. Assumption 1) is equivalent to saying that the temperature and vapor pressure at the walls remain constant throughout the expansion. The reduction in vapor density brought about by the expansion and the fact that walls remain at ambient temperature cause evaporation to occur at wet chamber surfaces so that both vapor and heat diffusion are in the same direction.

Using the above simplifications, the equations describing the uncoupled heat and vapor propagation into the chamber are

$$D\nabla^2\rho=\frac{\partial\rho}{\partial t}-k_\rho(t), \tag{2}$$

$$\kappa\nabla^2T=\frac{\partial T}{\partial t}-k_T(t), \tag{3}$$

where D and κ are the vapor and heat diffusion coefficients, respectively, and ∇^2 is in cylindrical coordinates.

The functions k_ρ and k_T are homogeneous *source* terms which account for the time dependent pressure expansion, i.e., the expansion decreases the vapor density as well as the temperature and this decrease can be described by means of a *source* (or rather a *sink*) term.

As the side wall is assumed to maintain thermal equilibrium at its surface, the vapor density there, ρ_1, is the equilibrium vapor density at the wall temperature T_1. Hence, the boundary and initial conditions are

$$T(R,t)=T(r,0)=T_1, \tag{4}$$

$$\rho(R,t)=\rho(r,0)=\rho_1, \tag{5}$$

where R is the radius of the cylinder and r is the variable.

Using straightforward Laplace transform techniques, the solutions may be expressed as convolutions of the separate inverses (Scott, 1955), i.e.,

$$T(r,t)=T_1+2\sum_{n=1}^{\infty}\frac{J_0(r\alpha_n/R)}{\alpha_n J_1(\alpha_n)}k_T(t)^*\exp\left(-\frac{\kappa\alpha_n^2}{R^2}t\right), \tag{6}$$

$$\rho(r,t)=\rho_1+2\sum_{n=1}^{\infty}\frac{J_0(r\alpha_n/R)}{\alpha_n J_1(\alpha_n)}k_\rho(t)^*\exp\left(-\frac{D\alpha_n^2}{R^2}t\right), \tag{7}$$

where the asterisk dentotes convolution.

The simplest case consists of an instantaneous expansion to the final values ρ_0 and T_0 at time zero, where these values are determined by assuming chamber *adiabaticity*. In this case the *source* terms are given as pulsed functions of the form

$$k_\rho(t)=(\rho_1-\rho_0)\delta(t), \tag{8}$$

where $\delta(t)$ is the Dirac delta function. This particular solution, while crude in its approximation, does have the merit that it can be rendered dimensionless and thus applicable to any gas-vapor combination in any tube of any diameter.[1]

3. The dimensionless solution

The dimensionless solution is

$$\frac{\mu(r,t)-\mu_0}{\mu_1-\mu_0}=1-2\sum_{n-1}^{\infty}\frac{J_0(r'\alpha_n)}{\alpha_n J_1(\alpha_n)}\exp(-t'\alpha_n^2). \tag{9}$$

Here the scaled variables are $r'=r/R$ and $t'=c/R^2t$. The character μ represents the diffusing quantity since Eq. (9) is valid for both heat and vapor diffusion; thus, $\mu=T$ when $c=\kappa$ and $\mu=\rho$ when $c=D$. Plots of the function $f(r',t')$ are shown in Fig. 1 for various *scaled* times t', where

$$f(r',t')=1-2\sum_{n=1}^{\infty}\frac{J_0(r'\alpha_n)}{\alpha_n J_1(\alpha_n)}\exp(-\alpha_n^2 t'). \tag{10}$$

Fig. 1 provides sufficient data to determine the heat and vapor diffusion profiles at different times for an expansion chamber of any dimensions containing any gas-vapor combination. In applying these data the *real time* can be obtained by multiplying the scaled time t' by R^2/κ for the temperature profile and by R^2/D for the vapor profile. The true radius can be obtained by multiplying the *scaled radius* r' by the actual radius R of the expansion chamber.

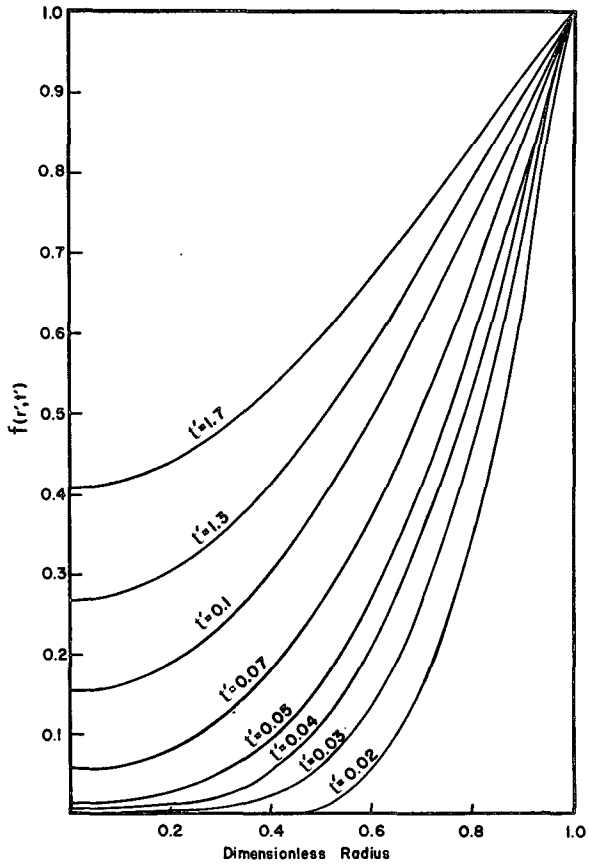

FIG. 1. Generalized diagram of heat and vapor diffusion from the wall of an infinite cylinder for an abrupt temperature change.

4. A more exact solution taking into account the actual expansion

However, the case of immediate concern is the *Pollak condensation nucleus counter* (Metnieks and Pollak, 1959), where a more realistic form for $k_\rho(t)$ and $k_T(t)$ can be provided since we know the manner of operation of the counter. The expansion of the Pollak type counter closely resembles the discharge of a capacitor through a resistance. A rough approximation to the density variation with time during the pressure expansion is given by the exponential form

$$\rho'(t)=\rho_0'+(\rho_1'-\rho_0')\exp(-t/\tau),$$

where ρ' represents the total gas density at any time, ρ_1' the initial gas density, ρ_0' the final gas density and τ is an appropriate relaxation time.

[1] The solution is valid so long as the tube can be regarded as infinitely long, i.e., we are not particularly interested in what happens near the ends of the cylinder.

Simple kinetic theory arguments lead to an *order of magnitude* value for τ of

$$\tau=\frac{2V_0}{A\langle v_0\rangle(\gamma^{\alpha'-1})}, \tag{11}$$

where V_0 is the volume of the cylinder, A the area of the expansion orifice, $\langle v_0\rangle$ the average speed of the outside air molecules (assuming that the inside and outside temperatures are initially the same), γ the adiabatic gas index, and $\alpha'=1/(\alpha^{\gamma-1})$ where α is the density ratio ρ_1'/ρ_0'. For typical Pollak counter conditions, the relaxation time constant is $\tau=0.055$ sec. This gives fair agreement with the pressure curves of Israel and Nix (1966).

The time variation of the temperature is inferred from the density variation using the adiabatic law:

$$T(t)=T_1\left(\frac{\rho'(t)}{\rho_1'}\right)^{\gamma-1}$$

$$=T_1\left(\frac{\rho_0'}{\rho_1'}\right)^{\gamma-1}\sum_{n=0}^{\infty}C_n'\exp(-t_n/\tau), \tag{12}$$

where

$$C_n'=\left(\frac{\rho_1'}{\rho_0'}-1\right)^n\frac{(\gamma-1)!}{n!(\gamma-n-1)!}.$$

Assuming Dalton's law holds, the vapor density ρ follows the same law as the gas density ρ'. Thus,

$$\rho=\rho_0+(\rho_1-\rho_0)\exp(-t/\tau). \tag{13}$$

The source terms can now be written as

$$k_\rho(t)=\frac{d\rho'(t)}{dt}=-\left(\frac{\rho_1-\rho_0}{\tau}\right)$$

$$\times\exp(-t/\tau)=-k_0\rho\exp(-t/\tau),$$

$$k_T(t)=-T\left(\frac{\rho_0'}{\rho_1'}\right)^{\gamma-1}\sum_{n=1}^{\infty}C_n\exp(-t_n/\tau),$$

where

$$C_n=\left(\frac{\rho_1'}{\rho_0'}-1\right)^n\frac{(\gamma-1)!}{T(n-1)!(\gamma-n-1)!}=\frac{C_n'n}{\tau}.$$

Using these source terms in (6) and (7), and making use of the identity

$$\exp(-AT)*\exp(-Bt)=\frac{\exp(-At)-\exp(-Bt)}{B-A},$$

where A and B are positive, real constants, we finally obtain

$$\rho(r,t)=\rho_1-2k_{\rho 0}\sum_{i=1}^{\infty}\frac{J_0\left(r\frac{\alpha_i}{R}\right)}{\alpha_iJ_i(\alpha_i)}\frac{\exp(\beta_it)-\exp(-t/\tau)}{\left(\frac{1}{\tau}-\beta_i\right)}, \tag{14}$$

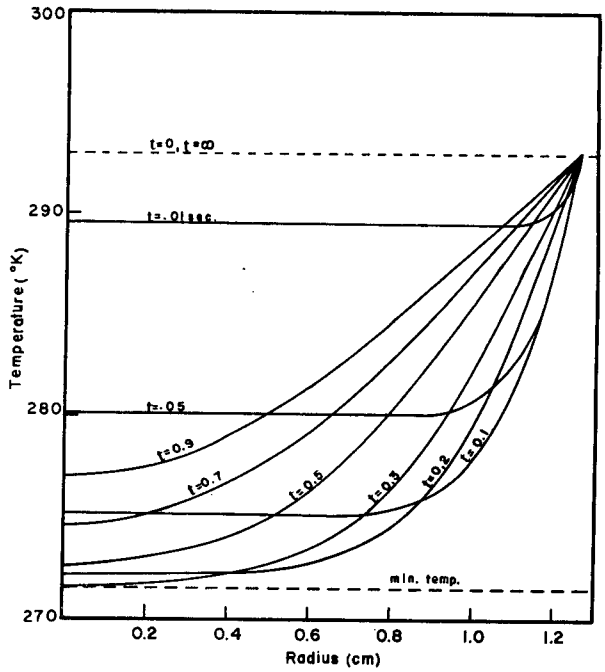

FIG. 2. Temperature diffusion profiles for the Pollak counter for a normal expansion speed.

$$T(r,t)=T_1-2\sum_{i=1}^{\infty}\frac{J_0(r\alpha_i/R)}{\alpha_iJ_i(\alpha_i)}$$

$$\times\sum_{n=1}^{\infty}C_n\left[\frac{\exp(-\beta_i't)-\exp(-t_n/\tau)}{\left(\frac{n}{\tau}-\beta_i'\right)}\right], \tag{15}$$

where $\beta_i=D\alpha_i^2/R^2$ and $\beta_i'=k\alpha_i^2R^2$. Temperature and vapor denstiy profiles are exhibited in Figs. 2 and 3.

5. Supersaturation decrement

Using the vapor density and temperature profiles given by Eqs. (14) and (15), it is possible to calculate the per cent decrease in the supersaturation due to heat and vapor diffusion from the walls, a quantity we call the *supersaturation decrement* for the standard Pollak counter. The supersaturation decrement is given by

$$\eta=\frac{|S(r,t)-S_0(t)|}{S_0(t)}100. \tag{16}$$

Here we take the *pre-expansion* and *post-expansion* total pressures as 1.0 and 0.83 atm, respectively, and the initial and final temperatures as 293K and 271.5K, respectively, leading to a supersaturation ratio of roughly 3.56. In Eq. (16), $S(r,t)$ is the supersaturation obtained by taking the ratio of the actual vapor density, $\rho(r,t)$, to that of the vapor density at equilibrium with the temperature, $T(r,t)$. $S_0(t)$ is the supersaturation obtained assuming the absence of heat and vapor flow from the

walls. Fig. 4 shows the supersaturation decrement as a function of radius for the Pollak counter at different times.

This analysis shows that the central portion of the chamber remains unaffected by wall effects for about 0.3 sec. This is roughly the time required to exhaust the chamber to its minimum value of vapor density, so that, at least in the center of the chamber, the assumption of adiabaticity should lead to correct results. Clearly, any expansion requiring longer than 0.3 sec will never reach its maximum supersaturation as inferred from adiabatic calculations, since beyond this time the wall effects have been transmitted all the way to the center of the chamber. Although the Pollak counter is not designed with a large margin of safety in this regard, the above calculations certainly vindicate it from the charge that the wall effect is responsible for the very low supersaturations which the results of Israel and Nix would indicate are established in lieu of the supersaturations predicted by the adiabatic law. The rate at which heat is transmitted from the walls of the expansion chamber towards the center has been verified by careful fine wire thermocouple measurements in a dry chamber in this laboratory. Calculations and experiment agree well within 20% which gives us confidence in this type of analysis.

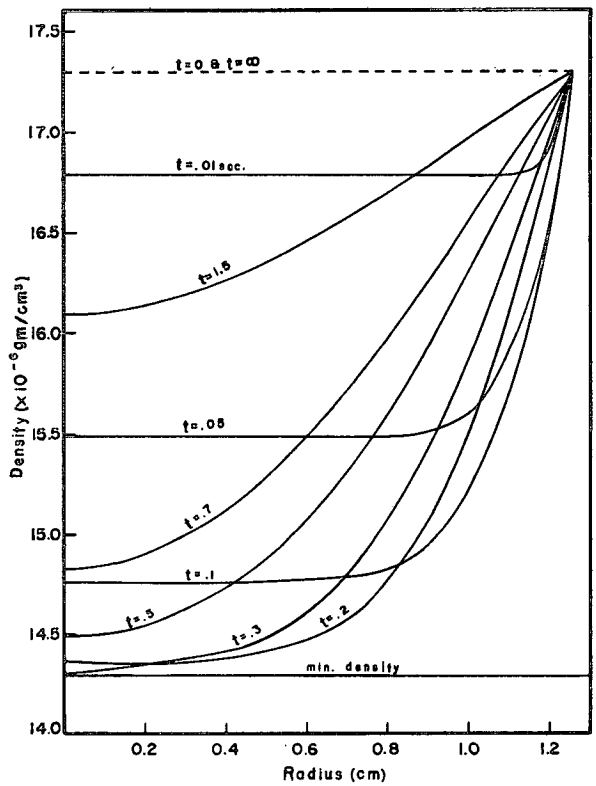

FIG. 3. Vapor density diffusion profiles for the Pollak counter for normal expansion speed.

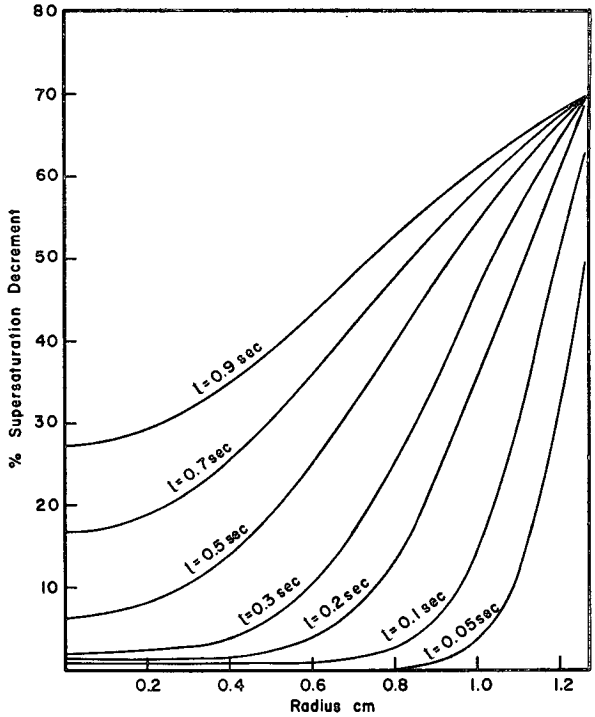

FIG. 4. Supersaturation decrement profiles for the Pollak counter for a normal expansion speed.

In order to explain the results of Israel and Nix we must turn our attention to other possible phenomena.

6. Measurements of temperature with fine wire thermocouples

Israel and Nix (1966) have investigated the thermodynamic processes taking place in the Pollak condensation nucleus counter and have concluded that the supersaturation developed by the expansion remains much lower than that predicted by thermodynamics. These investigators measured the temperature drop during an expansion with dry air and found it to be much smaller than predicted by the adiabatic law.

Measurements of gas temperature with fine wire thermocouples are not as simple as they appear to be. Some measurements have been made in this laboratory in an expansion chamber filled with dry gas in which both temperature and pressure were recorded on a light beam oscillograph with high accuracy, utilizing expanded scale techniques. From the equation of state for the gas the temperature can be calculated accurately by assuming that the expansion is isentropic. It was noted that the thermocouple began to read a temperatrue drop which was too low by about 1.5C almost from the beginning of the expansion. After the end of the expansion the calculated and measured temperatures come into agreement once again after several tenths of a second. This phenomena caused us to examine more carefully the problem of the heat flow from the thermocouple.

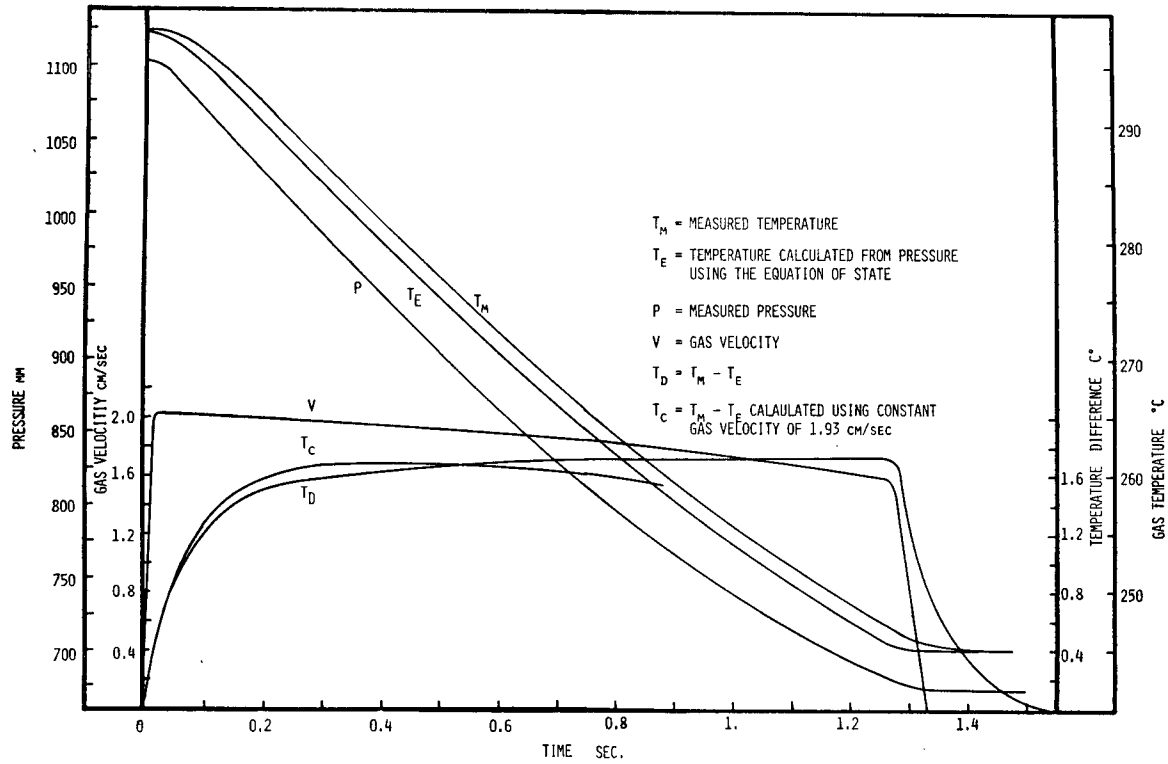

FIG. 5. Measurement of gas temperatures with fine wire thermocouples.

Let us look briefly at the physical situation. The thermocouple is initially in thermal equilibrium with the gas. Suddenly the gas temperature begins to decrease. The thermocouple wire has a finite thermal capacity and must communicate its excess heat to the surrounding gas by diffusion.

If the expansion proceeds at a constant rate, the temperature of the surrounding medium drops nearly linearly and the thermocouple becomes a steady source of heat just as surely as if it were being heated with an electrical current. Under these circumstances the rate of diffusion of heat away from the thermocouple will adjust itself so that a steady state condition exists, i.e., heat diffuses away from the thermocouple just as fast as it is being developed in the thermocouple (to use the electrical analogy). Since the establishment of the steady state only requires a time of the order of 10^{-4} sec, we see a very fast response to sudden environmental changes. However, the temperature being indicated by the thermocopule is not the true temperature of the gas.

The question then arises, how far off are the temperature readings? This point cannot be resolved by experiment alone. First, let us determine the speed with which temperature equilibrium is attained within the thermocouple wire itself. The solution to this problem is given by Churchill (1958), i.e.,

$$T(r,t)=1-2\sum_{n=1}^{\infty}\frac{J_0(\alpha_n r/r_0)}{\alpha_n J_1(\alpha_n)}\exp\left(-\alpha_n{}^2\frac{\kappa t}{C_p r_0{}^2}\right),$$

where κ is the thermal conductivity of the wire, C_p the heat capacity, r_0 the radius of the thermocouple, and the α_n are the zeros of the Bessel functions: $\alpha_1=2.405$, $\alpha_2=5.520$, $\alpha_3=8.654$ and $\alpha_4=11.79$. We used chromel-alumel thermocouples made from 0.0005 inch (12 μ) diameter wire. It is seen that a perturbation on the outside of the thermocouple is felt at the center with a half life of 3.0 μsec. Therefore, the relaxation time of the thermocouple itself is completely negligible. This is, of course, one necessary ingredient for fast response.

The calculation of the heat flow from the thermocouple surface out through the gas is much more difficult because the radial symmetry is lost when the gas begins to move past the thermocouple as it does in expansion cloud chambers. However, in this case the gas velocity is small and laminar flow may be assumed (Adams, 1954; Schlicting, 1960).

Since there is no variation in the direction of the axis of the *stretched out* thermocouple wire, the problem reduces to a two-dimensional heat flow problem,

$$C_p\frac{\partial T}{\partial t}=\kappa_g\frac{\partial^2 T}{\partial x^2}+\frac{\partial^2 T}{\partial y^2}+f(x,y,x,t),$$

where C_p is the heat capacity of the gas at constant pressure, κ_g the thermal conductivity of the gas and $f(x,y,x,t)$ a source function which allows for varying expansion speeds, i.e., it allows for the motion of the gas

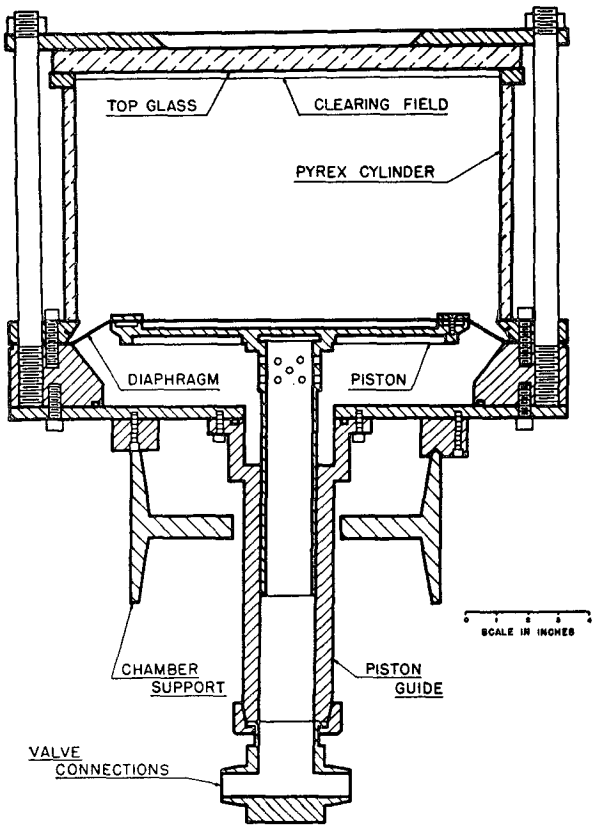

FIG. 6. The cloud chamber used for measuring gas temperature with a thermocouple.

over the thermocouple. Only numerical solutions of this problem were attempted.

Fig. 5 shows the results for an expansion in dry argon. The cloud chamber used for this study is shown in Fig. 6. The thermocouple was located 8 cm from the top glass. The evacuated chamber was filled with tank argon which was passed through a liquid nitrogen cold trap to insure its dryness. Note that Fig. 5 indicates close agreement between the theoretically predicted thermocouple temperature and the measured thermocouple temperature. The difference is about 1.5C after a short time for a gas velocity of 2 cm sec^{-1}. Clearly, the fast response of the thermocouple is no indication of the accuracy with which it reads the gas temperature.

A simple calculation shows that the heat capacity of the wire is sufficient to cause a 0.1C rise in temperature in a cylinder of gas with a radius of 0.3 cm. This is misleading, however, since the small gradients make the dispersal of the evolved heat very slow and the temperature rise close to the wire is much higher.

Israel and Nix (1966) investigated the thermodynamic processes in the Pollak counter by inserting a fine wire thermocouple into the dry chamber. They reported only a fraction of the temperature change expected from a calculation of the temperature drop by means of the adiabatic law. This result is exactly what one would expect from the foregoing analysis.

Moreover, their Fig. 2 showed a peculiar anomaly at a time of 1.5 sec. One can explain this feature as follows. At the end of the expansion the thermocouple still has a heated gas mass surrounding it and so it reads a temperature which is too high. The heating of the gas adjacent to the walls excites convection currents which take a moment to get started. At 1.5 sec after the expansion the convective motion sweeps the heated gas surrounding the thermocouple away, allowing it to read a temperature which more closely approximates the true temperature. The temperature readings from about 2 sec on should more closely approximate true values and an extrapolation of this part of the curve back to the time immediately after the expansion gives more nearly the temperature drop brought about by the expansion.

7. Conclusions

In conclusion we might say that thermocouple measurements of gas temperature present a degree of complexity which has not been generally recognized. The actual response of the fine wire thermocouple (as opposed to the speed with which it responds to a sudden change in its environment) is very slow. The disconcerting results of Israel and Nix can be adequately explained in terms of actual thermocouple response. Their work is useful in that it locates the point in time when the Pollak counter becomes subject to turbulent convection currents. However, the Pollak counter's adiabaticity cannot be questioned on the basis of these measurements. In all probability it is sufficiently adiabatic for the purpose for which it was intended, provided all measurements are completed before the convective motion takes place. Prior to this time the adiabaticity of the center of the chamber is undisturbed by heat flow from the walls and convection.

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