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AN EXPERIMENTAL DETERMINATION OF THE HOMOGENEOUS NUCLEATION RATE OF WATER VAPOR

IN ARGON AND HELIUM

by

LOUIS BENTON ALLEN, JR.

A DISSERTATION

Presented to the Faculty of the Graduate School of the UNIVERSITY OF MISSOURI AT ROLLA

In Partial Fulfillment of the Requirements for the Degree
DOCTOR OF PHILOSOPHY

in

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AN EXPERIMENTAL DETERMINATION OF THE HOMOGENEOUS NUCLEATION RATE OF WATER VAPOR

IN ARGON AND HELIUM

An Abstract of a Dissertation Presented to the Faculty of the Graduate School University of Missouri at Rolla

In Partial Fulfillment of the Requirements for the Degree Doctor of Philosophy

by

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May 1968

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Abstract

An expansion type cloud chamber was used to measure the nucleation rate of water vapor in an atmosphere of helium and argon. A careful study was made of the thermodynamic characteristics during the expansion so that the nucleation data could be interpreted with reasonable accuracy and consistency.

A fine wire thermocouple was used to measure the gas temperature during the course of an isentropic expansion in the dry chamber. When the finite heat capacity of the thermocouple is accounted for, it is found that there is almost perfect agreement with the temperature calculated from the equation of state and the pressure measurement. This establishes the expansion cloud chamber as the instrument with the most accurately known thermodynamic characteristics and the one where the supersaturation may be calculated with the greatest precision.

The homogeneous nucleation rate of water vapor in a helium atmosphere was measured as a function of temperature, supersaturation and sensitive time. It was found that there exists a form of heterogeneous nucleation occurring above the *ion limit* at about the *critical supersaturation* predicted by the classical Becker-Doring theory for homogeneous nucleation. This form of heterogeneous nucleation appears to occur upon chemically bonded centers whose concentration is very low and depends upon the vapor pressure before the expansion. The consistency of the number of these nucleating centers indicates that they may be a neutral product of the action of natural radioactivity and cosmic rays.

A semiphenomenological theory was developed along the lines of the classical theory but which includes the chemical bond energy of the heterogeneous nucleating center. The *theory* predicts a different temperature dependence for the heterogeneous and homogeneous nucleation rates and at least qualitatively explains the essential features of the experimental data.

A considerable disparity in the temperature dependence of the critical supersaturation limit has existed for many years. The variation in the temperature dependence with nucleation rate as determined by the author's data shows: (a) that a large part of the disparity is due mainly to the interpretation of the experiments and (b) that the different temperature dependence of the heterogeneous and homogeneous nucleation rates is responsible for the different temperature dependences reported by the various experimenters.

It was definitely established that the nucleation rate of water vapor is higher in an argon atmosphere than in a helium atmosphere. This may be due to a disruption factor related to the higher velocity of the *light* helium atoms. It is, however, more likely due to the hydration of the argon atom into the critical cluster with a resultant increased stability in the critical clusters.

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James L. Kassner, Jr., Dissertation Supervisor

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CHAPTER I

STATEMENT OF THE PROBLEM

Man has long been studying the atmosphere, yet it is only in the last few years that technology has advanced to the point where a true understanding of atmospheric processes is possible. This understanding must be turned to useful ends if we are ever to forget the fear of storms, make the deserts bloom or to continue to have clean air to breathe. Of all the problems facing atmospheric scientists, perhaps the most fascinating is that of the action of water vapor in the atmosphere. Water vapor content determines the stability of atmospheric layers and exerts control on the energy balance in the atmosphere¹. The details of the processes by which the size distribution of droplets in a cloud changes with time is not well understood.²,³

Before an understanding of complex processes may be attained, the most elementary and basic processes must be comprehended. Nucleation, the formation of new droplets, is not the simple process that it was once thought to be. Nucleation on particulate matter is a form of heterogeneous nucleation. Atmospheric nuclei serve to lower the energy barrier for condensation. Ions also serve to reduce the free energy of formation of clusters. In the absence of condensation nuclei and ions, droplets begin to form as a result of chance fluctuations at a supersaturation of 4.8 and higher. The latter has been termed homogeneous nucleation. Early experimental results,^{4,5,6,7,8} observing the so-called critical limit for heterogeneous nucleation on smoke particles, dust particles, ions and homogeneous nucleation, showed that at least a qualitative explanation was provided by classical theory⁹. Classical theory is based on the idea that a barrier to nucleation exists and that statistical fluctuations carry the embryos over the barrier. A critical size of embryo is associated with the peak height of the free energy barrier. It now appears that a full understanding of homogeneous nucleation is required before the various forms of heterogeneous nucleation may be comprehended. Heterogeneous nucleation, which is the dominant form of nucleation found in nature, encompasses the features of homogeneous nucleation with the addition of extra interfacial energies which greatly complicate the problem.¹⁰

The semiphenomenological classical theory, developed by Farkas¹¹, Becker and Doring¹², Zeldovich¹³, Frenkel¹⁴ and others for the homogeneous nucleation of liquid drops, seemed at first to predict nucleation rates which showed good agreement with experiment. More definitive experimental data on the nucleation of water drops from the vapor by various groups, namely Volmer and Flood¹⁵ and Powell¹⁶, exhibit self consistency within themselves but display considerable disparity when intercompared. Attempts to compare results comprehensively^{17,18,19,20} have only emphasized the pecular nature of homogeneous nucleation in water vapor.

The extent of renewed interest in nucleation phenomena is evidenced by the amount of recent theoretical activity²¹⁻³⁵. This renewed interest also indicates a general lack of confidence in the classical nucleation theory. Moreover, there has been a resurgence of interest in the experimental measurement of homogeneous nucleation rates.^{19,20,36-40} Most experimental studies have observed the critical supersaturation limit only. The critical supersaturation limit is usually taken as that point where noticeable droplet formation occurs in the expansion chamber. This may be for nucleation rates of from one to one million droplets per cubic centimeter per second, depending upon the details of the observation system and the sensitive time of the apparatus. Due to the nature of most of the experiments where sensitive times and droplet densities are estimated only to an order of magnitude, a given experiment may be intrepreted differently by different authors.^{20,36} Since both the estimates of sensitive times and drop densities are usually consistent for a given investigator, the temperature dependence is adequately determined but the nucleation rate to which it belongs is somewhat ambiguous.

Definitive experimental work must be done so that a comprehensive picture of the homogeneous nucleation process may be constructed for comparison with theory. It is the purpose of this study to provide a set of data overlapping that of several other experimenters and to provide the most extensive measurements possible, utilizing the capabilities of the highly automated and instrumented cloud chamber of this laboratory. The investigation undertaken by the author will experimentally determine the homogeneous nucleation rate as a function of sensitive time, supersaturation and temperature.

<u>1-1</u>. Instruments for measuring the nucleation rate. Nucleation theory specifically predicts the rate of formation of droplets as a function of supersaturation and temperature. It is the function of experimentation to verify the essential features of the theory. The nucleation rate is such an exceedingly steep function of increasing supersaturation

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that, in a rather crude manner of speaking, it exhibits a supersaturation at which the nucleation rate first becomes observable. This has become known as the *critical supersaturation*. Early work directed toward the confirmation of nucleation theory was done with the expansion cloud chamber which was used to measure the aforementioned critical supersaturation. This technique involved an estimation of droplet concentration. Moreover, it was necessary to estimate the nucleation period or sensitive time of the cloud chamber in order to convert the observations to a nucleation rate. Little advance in technique has been reported until recent years. It is improbable that any major developments in nucleation theory will evolve until more definitive experimental measurements are forthcoming. It is to this end that work in this Jaboratory has been directed.

Several experimental techniques are useful for studying nucleation phenomena. Each possesses its own characteristic advantages and disadvantages. All achieve a state of supersaturation either by adiabatic cooling or by nonisothermal vapor diffusion. The expansion cloud chamber employs the nonisothermal diffusion of vapor to produce supersaturation.

Nucleation phenomena have been studied by means of expansion nozzle techniques by Ruedy⁴¹, Wegener⁴² and Pouring⁴³. The nozzle method has the advantage of providing a steady state observation. Pressure is measured as a function of position in the flow stream. Deviations from the characteristic isentropic flow indicate the presence of condensation. The complete removal of dust and ions is impractical, but under conditions of rapid production of the condition of supersaturation such high nucleation rates are achieved that any contribution due to heterogeneous nucleation is obscured. Because very high nucleation rates

are associated with very small critical cluster sizes, it is improbable that nozzle experiments will be of value in evaluating a nucleation theory which is essentially a precatastropic theory. Moreover, it is likely that the liquid drop model breaks down for such small cluster sizes.

The diffusion chamber was invented in 1939 by Langsdorf⁴⁴, but it was not fully developed until the early 1950's.^{45,46,47} The gas in the upper part of the chamber must be less dense than the gas in the lower part of the chamber in order to prevent convection currents. Most thermal diffusion chambers operate with the upper plate at the higher temperature. Franc and Herz⁴⁸ first described an inverted diffusion cloud chamber, a chamber with water vapor diffusing up from the bottom. A very light gas such as hydrogen or helium must be used for the atmosphere in order to maintain the stability of the system. Katz and Ostermier³⁹ have used the inverted diffusion chamber to measure the temperature dependence of the critical supersaturation ratio for a number of vapors. Since the determination of the supersaturation is dependent upon an accurate knowledge of the diffusion coefficients for the vapor through the non-condensible gas, they have resorted to measuring the diffusion coefficients over a wide range of temperatures themselves.

The primary advantage of the thermal diffusion chamber lies in the fact that it is a steady state device. However, the fact that the thermodynamic coordinates are changing continuously as a function of position between the parallel chamber plates presents a problem in the determination of the state of supersaturation. Moreover, only a thin layer of the chamber exists at a state of high supersaturation. The thermal diffusion chamber suffers from the disadvantage that it can

measure only very small nucleation rates or the so-called critical supersaturation. It is most useful for measuring the temperature dependence of the critical supersaturation.

The expansion cloud chamber is the oldest device used for measur, ing nucleation rates. It was highly developed by nuclear physicists for the detection of ionizing particles. Moreover, it has undergone additional development in this laboratory as a tool for investigating nucleation and condensation phenomena.⁴⁷⁻⁵² Extensive studies of the thermodynamic characteristics of the expansion cloud chamber in this laboratory and in other laboratories make it the best understood of all devices available for studying nucleation phenomena. It can yield more information than any of the above mentioned instruments.

Expansion cloud chamber experiments have been customarily designed to yield only a measurement of the critical supersaturation, this is the simplest experiment which can be performed. The definition of the critical supersaturation, as it is used by a given investigator, is obviously influenced by the geometry of the observation system. Droplet densities have either been estimated visually or determined by light scattering techniques. Nucleation rates are estimated by making an educated guess for the sensitive time of the chamber, i.e. that time during which the bulk of the nucleation occurs.

The major advances made in this laboratory are the continuous measurement of the pressure throughout the expansion and the development of techniques for maintaining almost complete control over the expansion cycle. With proper instrumentation and automation, the expansion cloud chamber has an overwhelming advantage over other experimental methods in that nucleation measurements may be undertaken as a function of supersaturation, temperature and sensitive time. No other instrument has been used to measure the dependence of the nucleation rate upon either supersaturation or time. Such diversity of information makes it possible to distinguish more clearly between different types of nucleation schemes. For this reason, the expansion cloud chamber has been selected for this work.

CHAPTER II

EXPERIMENTAL TECHNIQUE

The cloud chamber used in this work was first put into operation in March, 1962 and has undergone continuous improvement since that time, Fig. 1. Great strides were made in regulating the temperature of the cloud chamber. A servo pressure regulation control system was developed which made possible more accurate control over the starting pressure, thereby insuring accurate saturation at the starting temperature. A mechanical brake was added to the piston guide for damping out piston oscillations during the expansion. A program board was installed to facilitate the interconnection of the programming circuitry to the operating devices such as valves, lights, recorder, etc. These improvements were initiated in whole or in part by the author and contributed substantially to the success of the measurements made during the course of this work.

2-1. <u>Temperature control</u>. Allard⁴⁹ used a top to bottom gradient of one Centigrade degree in order to eliminate condensation on the top glass and side walls of the cloud chamber. It was felt that such a large gradient should not be necessary and that a gradient of several hundreths of a degree should suffice. Packwood⁵⁰, Schmitt⁵¹, Dawbarn⁵², Smith⁵³ and White⁵⁴ all attempted to reduce the magnitude of the graient without much success. Before the problem could be properly assessed, a set of thermocouples was constructed and mounted inside the cloud chamber. They were spaced one inch apart up the side wall and



Fig, 1. The cloud chamber used for measuring homogeneous nucleation rates.

across the top. Comparison of temperatures on both sides of the top glass immediately indicated that there was more than one-half degree gradient through the glass. Moreover, about one-half degree difference in temperature existed between the center and the edge of the vicwing window, see Fig. 2. The addition of another glass plate on top of the cloud chamber for added insulation did not remedy the problem. Fine heater wires were stretched across the viewing window for independent temperature control of the center of the top glass. One-half watt of heater power sufficed to eliminate the temperature variation across the top glass.

The vertical temperature gradient had to be reduced drastically. It was evident that the top half of the cylindrical wall was too cool. This allowed condensation to occur on the walls where the light beam from the flash lamps passed through the cylinder. This was very undesirable because the condensation scattered the light beam. Heater tapes were mounted just above the light beam. This tape nearly filled the space between the clearing field ring and the light beam. After a week of adjusting the voltages on the three side heaters and two top heaters, good temperature control of the interior chamber was achieved.

As the gradient was brought to less than one-tenth degree, uneven condensation occured at various parts of the chamber top and sides. This indicated that there were gradients across the chamber which were not previously recognized. Before this problem was solved: (a) the convection currents from the air conditioner had to be completely baffled from the chamber by draping one-quarter inch rubber sheet around the chamber frame and associated plumbing, (b) extra foam rubber was added around each of the four sides of the chamber and (c) four separate



Fig. 2. Arrangement of the cloud chamber heaters.

heaters were added in the top cavity, one on each of the four sides. Temperature gradients across the top cavity due to the forced air convection currents from the room air conditioner are eliminated by individually adjusting the power input to each of the four heaters. Elimination of the cross gradients is best accomplished by lowering the gradient and allowing slight condensation to take place on the top glass and sides. Observation of the condensation tells the location of the cooler areas.

During the regular cycling of the chamber, the temperature of the bottom of the chamber drifted out of control. This was due to the heat pumping action of the cycle. Elimination of this problem consisted of installing new heaters in the aluminum plate under the piston and controlling the temperature of the air coming into the manifold system.

The temperature control during the periods of data taking was not quite as good as the temperature control during the time when the thermocouples were installed in the chamber. Temperature control did not prove dependable enough to use a gradient under 0.05° C and a gradient of about 0.1° C was finally used for convenience. The temperature of the central volume of the cloud chamber in which measurements were being made was therefore known to within the same accuracy as the water temperature, $\pm 0.05^{\circ}$ C.

2-2. Pressure regulation. Leaks always occur in the plumbing under the cloud chamber. These leaks must be counteracted by letting excess air into the manifold system at a rate which exactly compensates for the leaks. Up to the time of this series of experiments, there had been no need for the initial pressure regulating system to be adjustable. The pressure regulator described by Allard consisted of a mercury manometer with platinum wire contacts which controlled a regulator valve. This system was naturally oscillatory; the best regulation possible after careful adjustment gave pressure oscillations of about 2 mmHg.

In the new control system a pressure transducer takes the place of the mercury manometer and a servo motor attached to a variable orifice valve, Fig. 3, does the regulating. A trickle of air is continously leaked into the manifold system as with the earlier system and the servo motor controls the valve opening so that the proper leak rate is established to maintain a constant pressure.

Some difficulty was encountered when trying to get the servo system to work. When the servo valve was allowed to pass by the completely "open position" and on to the completely "closed position" or vice versa, an oscillating condition occured. With this arrangement control could only be established when the system pressure was nearly regulated beforehand. Installation of a clutch and stops at the fully opened and fully closed positions eliminated this problem so that the servo system could bring the pressure to regulation from an expansion or compression configuration very quickly without disrupting the sense of the servo control.

It was also found that the location of the pressure transducer controlling the servo motor is critical. It must be located in the same pressure line as the servo valve and be very close to the orifice in order to avoid pressure oscillations. The same biasing technique is used with this transducer as with the pressure transducer in the sensitive volume. The starting pressure may now be held to within one mmHg with practically any volume of trickle air leaking into the manifold



regulator valve

Fig. 3. Block diagram of the pressure regulation system.

system. Recovery is so good that recovery times of less than ten seconds are required to establish any given pressure. The regulating ability of this system seems to be limited only by the capability of the pressure transducer.

<u>2-3.</u> The pressure measuring system. The precision measurement of the pressure in the sensitive volume throughout the expansion is the most critical part of a nucleation experiment. If the supersaturation is to be determined within one percent accuracy, the pressure must be measured to within one mmHg. This requires much better resolution than can be obtained with an ordinary recording system. The stability of all components in the pressure measuring system must be of the order of 0.01 percent. The pressure measuring system employed in this work was essentially the same as that described by Packwood⁵⁰. A block diagram of the pressure measuring system is shown in Fig. 4.

The pressure transducer is a one-quarter inch diameter, flush diaphragm, strain gauge type pressure transducer. Its diaphragm is mounted nearly flush with the inside cylindrical wall of the sensitive volume. It reads absolute pressure. The transducer is excited by a highly isolated transducer power supply which possesses 0.1 micromicrofarad capacitance between its output and power line sides. This specification is necessary to keep the common mode signal to the California Instruments wide band D.C. amplifier at a low level. Triply shielded twisted pair lead wires are used to interconnect all the units. Three separate mutually insulated shields are necessary to keep the A.C. noise level to a minimum. It was also necessary to mount the amplifier and the isolated power supply as close to the pressure transducer as possible.



Fig. 4. Block diagram of the pressure measuring system.

In order to obtain the desired accuracy in the recorded pressure signal, a type of scale expansion system was employed. Approximately 1.2 volts D.C. signal emerges from the California Instruments amplifier. Most of this signal is biased out by a constant D.C. voltage from the automatic bias module, having a 0.01 ohm impedence. The remaining 40 millivolts signal is recorded by a light beam oscillograph. Four different bias levels are provided by the automatic bias module. The automatic bias module's circuitry maintains a high level of isolation from both power line and chassis ground. The peak to peak noise level in the recorded signal corresponds to about 0.5 mmHg. Figs. 5-7 show typical data output from the oscillogram.

2-4. Pressure calibration. Pressure calibration was greatly facilitated by the addition of a Texas Instruments Model 145 Pressure gage with a precision servo nulling readout. This type of gage uses a quartz spiral bourdon tube which exhibits no measureable hysteresis and retains its calibration indefinitely. The servo readout follows pressure changes automatically, allowing instant comparison of recorder and prossure gage readings. Hysteresis in the cloud chamber pressure transducer and in the recorder galvanometers presented a problem. The calibration of the pressure transducer had to be accomplished by approaching the desired calibration point in the same manner that the cloud chamber would reach that same point in a normal data taking cycle. Approximately the same magnitude of pressure excursion was used in the calibration procedure as in the data taking cycle. It is probable that some of the difficulties encountered by Allard⁴⁹ and Schmitt⁵¹ resulted from improper calibration procedure.



Fig. 5. Sample of visicorder data. Galvanometer sensitivity 9.0mm/in, chart speed 10in/sec.



Fig. 6. Sample of visicorder data. Galvanometer sensitivity 9.0mm/in, chart speed 40in/sec.



Fig. 7. Sample of visicorder data. Galvanometer sensitivity 9.0mm/in, chart speed 40in/sec.
As the chamber began cycling, the zero offset drifted for about an hour. It is believed that this is the result of the establishment of a slightly different operating temperature brought about by the heat pumping characteristics of the cloud chamber. Calibrations were run every fifteen minutes until three consecutive calibrations, identical to within 0.3 mmHg., were obtained. Thereafter, the calibration was repeated every half hour. Note that by judicous choice of cycle parameters this heat pumping can be almost eliminated.⁵⁶

Since the pressure transducer used a semiconductor strain gage element, it is quite sensitive to changes in its ambient temperature. It was found that the equilibrium temperature of the strain gage element was dependent upon the thermal conductivity of the gas used in the cloud chamber. The calibrations for different gases differ slightly in their zero offset. However, the heat capacity of the pressure transducer was sufficiently large so that temperature changes during the expansion had a negligible effect on the transducer's calibration.

2-5. Photographic technique. In order to determine the nucleation rate, the number of droplets per cubic centimeter must be determined with considerable precision after the expansion. The necessity for imaging individual droplets places strict requirements on the illumination system and photographic technique. Conditions are accurately known only within the central region of the sensitive volume so only this portion is illuminated by means of a horizontal sheet of light whose vertical thickness is about one centimeter, see Fig. 8. Figs. 9 and 10 show the quality of the collimation of the light beam used in this work. The top and bottom edges of the beam exhibited a sharp cutoff in intensity. The droplets have diameters from ten to fifteen microns at the time



Fig. 8 Flash Tube Light Source

Lamp number 1: collimated July 18,1967 Louis Allen Don Hoffman



Lamp number 2; collimated July 19, 1967 by Louis Allen



they are photographed. It was found that one lamp does not give sufficient illumination. Two lamps were flashed simultaneously on opposite sides of the cloud chamber. The total energy input to the lamps is about 800 joules. The camera was set eighteen inches above the beam. Sufficient intensity and depth of field were obtained with an f-stop setting of 3.5. A grid of wires spaced at one centimeter intervals was photographed and used for calibration of the field of view of the projected film. see Plate 1.

There is an optimum time for photographing the droplets after they are nucleated. Droplet growth is dependent upon several factors which were varied during the course of the experiment, namely supersaturation and the transport characteristics of the inert gas. The optimum time for photographing the droplets was experimentally determined in each case. Growth times varied from 0.05 second in helium at high temperatures to nearly half a second in argon at low temperatures. Plates 2 and 3 show typical homogeneous nucleation for several different droplet densities.

An anomoly showed up in the photographs which was not expected. Droplets photographed at precisely the time they are coming into visible size form rather large diffraction rings on the photograph. Pictures taken slightly later have sharply focused images. This is probably a case of Fraunhofer diffraction through the lens. A calculation assuming Fraunhofer diffraction gives droplet size of the order of ten microns which is the size calculated from the droplet growth computer program. It appears that this technique could be used to determine the droplet growth rate for an accurate verification of the droplet growth law, see Plate 4.



Plate 1. Grid used for calibrating the camera magnification. A 1 cm. grid made from .4 mil tungsten wire is placed in the sensitive volume of the chamber and photographed.



Plate 2. Nucleation. Examples of typical data are shown with the same magnification as the grid in Plate 1. Upper 250 drops/cm³, lower 140 drops/cm³.



Plate 3. Nucleation. Examples of typical data are shown with the same magnification as the grid in Plate 1. Upper 42 drops/cm³, lower 2.5 drops/cm³.



Plate 4. Diffraction from small droplets. The upper picture shows droplets as they appear when small enough to give noticeable diffraction rings on the film. The lower picture shows the same droplets 0.1 sec later. Another strange effect showed up on the film developed on Feb. 8, 1968. Between some of the frames where the camera stopped, lines of static electricity or something similar sensitized the film. There is no shutter in the camera and the lines seem to have been catalyzed by light reflected onto the film. The static electricity itself was probably the result of strains induced into the emulsion by the motion of the film. During this particular data run the room and consequently the camera was maintained at 40°F. The atmosphere in the room was very dry. This particular combination of physical conditions is probably responsible for the effect. A developed print of this effect is shown compared with a normal print in Plate 5.

<u>2-5.1</u>. Film and development. Due to the very small area of the images, much denser blackening is required than in the case of ordinary photography. Moreover, grays are of no interest so that a fast, high contrast film can be employed. A degree of over development materially increases contrast and thereby the effective film speed. A wide range of results can be obtained with different film and developer combinations. Film and developer combinations which yield high speed and high contrast tend to yield a large grain size in the developed film. If the grain size becomes too large, the effective image diameter is increased.

Virtually all interesting film and developer combinations have been investigated in this laboratory, carefully noting the effective relative speeds and the maximum obtainable resolution. It was found that Eastman Kodak Linograph Shellburst film developed in Acufine film developer made by Baumann Photo-Chemical Corp. gives the greatest relative speed as well as a fine grain size. Eastman Kodak Tri-X is not quite as fast and gives considerably larger grain size.



Plate 5. Static. Stresses in the emulsion of the film under certain conditions when developed display a lightning like effect which seems to be catalyzed by light. These are frames exposed during the waiting interal between expansions. This effect is shown compared to a normal frame. A seven minute developing time is used in full strength developer at 20°C. An 18 minute developing time is used in half strength developer at 20°C when the Nikor developing machine is used.

2-6. The machanical Brake. During the normal operation of the cloud chamber, the piston is in free suspension between the upper and lower gas volumes. Such a system is oscillatory. The original design of the cloud chamber used a hole plate suspended in the water volume for damping the piston oscillations. Although the hole plate did reduce the waves in the surface of the liquid pool, it was inefficient as a damping mechanism. A solenoid operated brake was connected to the guide cylinder. It was originally designed so that the braking action could be electrically controlled. However, it was found that it could be adjusted for a constant slight drag which was sufficient to critically damp the piston. Small oscillations in the rubber diaphragm were unavoidable. The remaining oscillations were so small, however, as to be insignificant.

<u>2-7.</u> Program board. A programming patch board was added to the cloud chamber in order to facilitate the programming of different experiments, see Plate 6. It was hoped that this would reduce the down time required for reprogramming and add to programming versitility. The program board contains 1632 contacts. These are used to interconnect the timing units with the control apparatus of the cloud chamber. A sequencing unit was installed in conjunction with the program board. This allows a sequence of up to ten expansions, each of which can be different. Moreover, a numbering system for the photographs is provided. The entire installation consists of more than 10,000 connections and three miles of wire.

A patchboard is wired for each type of experiment. As long as the patchboard remains intact, the experiment may be repeated at any time.



Plate 6. Program board. A rear view of the program board is shown. The patchboard used for the author's data is in place. If two different experiments can use the same cloud chamber configuration, the appropriate program board may be installed and in a matter of minutes that experiment can be underway. This system makes more efficient use of the cloud chamber facility. It is also possible for several different cloud chambers to be run at alternate times using the same electronic control system and data processing equipment.

<u>2-8.</u> Purification of the water. For homogeneous nucleation rate measurements from supersaturated vapor, every effort must be made to obtain the purest possible vapor so that one may be reasonably assured that the nucleation is indeed homogeneous and not influenced by impurities. The purification of water is hampered by the fact that it is an almost "perfect" solvent. Under normal conditions water is saturated with silicates, various metal ions, and all atmospher ic gases including carbon dioxide. Water has such an affinity for impurities that freshly distilled water left open to the atmosphere for a few minutes will not pass conductivity tests for purity. This is due mainly to dissolved gases.

Since the nucleation rate studies were to be done with water vapor in an atmosphere of a pure rare gas, a special purification procedure had to be devised to provide pure water free from not only dissolved solids and liquids, but free from contaminating gases. These gases might affect either the vapor pressure or bonding characteristics of the vapor molecules and modify the nucleation rate.

Various methods of purification were considered, including ion exchange techniques in conjunction with distillation. The final conclusion reached was that distillation is the most effective means of obtaining very pure water, provided that several stages are used with each stage performing a different function.

General distillation procedure was worked out with the aid of Dr. James L. Kassner, Sr., an expert in the field.

The first operations were devoted to eliminating organic compounds since these impurities were deemed to be the least desirable, the most likely to contaminate the vapor and the hardest to remove. One should keep in mind that the amount of a substance present doesn't have to be large in order to have a great number of molecules present. For instance, a tolerably good vacuum of 10^{-6} torr still has ten billion molecules per cubic centimeter. Compared to normal operating pressure, this level of impurity represents about 1×10^{-7} per cent at a total pressure of one atmosphere and only one impurity atom for each 10^7 water molecules. Keeping impurity levels down to just a few molecules per critical embryo becomes impossible and one must settle for the greatest dilution of impurities pessible.

Ordinary distilled water contains some organic matter. Tap water was used as the starting water since it requires no less treatment for purification then ordinary distilled water. Potassium permanganate (ten grams per liter) with enough potassium hydroxide to assure an alkaline solution (PH of about 8 or 9) was added to the water and left standing in five gallon glass stoppered jugs for a few days. This solution was then cooked for twelve hours and refluxed for twelve hours (that is boiled and recondensed into the same flask) in such a way that any volatile gases had ample opportunity to escape. Cooking and refluxing are done so that all of the organic compounds are broken up either into volatile gases which escape or else into nonvolatile compounds which are removable by distillation. Carbon ends up as potassium carbonate provided there is sufficient potassium hydroxide in the solution. This liquid was then distilled through a two stage continuously running still at about one-fourth liter per hour, see Fig. 11 and Plate 7.

It should be noted that the water was taken through the distillation apparatus beginning with twenty gallon batches. The first few liters as well as the last few liters, from each batch at each stage were discarded in the sense that the water was not kept as pure water but used for cleaning bottles and flasks, see Fig 12, and other such procedures necessary to the successful operation of the stills. This technique requires that four gallons of water start through the still to get one gallon of pure water out.

This permangnate solution was distilled and redistilled immediately after refluxing. Both stills were then dismantled and thoroughly cleaned so that the second stage of purification could begin. A very small amount of phosphoric acid (ten ml in 3000 ml) was added to the first still and the entire batch run through the stills in the same manner that the permanganate solutions was run through the still. Phosphoric acid was added to form insoluable phosphates of the heavier elements present and to make the solution acid.

It is not commonly known, but very pure water has a tendency to superheat. Boiling beads of many different materials were tried. Without a single exception, either the substance did not aid the boiling or interacted with the water and dissolved. Substances tried included ceramic beads, glass beads, carbon chips, various stainless steels covar metal alloy and other materials. Covar worked nicely but dissolved very quickly. Pure nitrogen works very well as a nucleating agent when slowly bubbled through the liquid in the boiling flask. This method was prohibited in this experiment, however, since the purification had to eliminate gaseous impurities as well as dissolved liquids and solids. In fact,





Plate 7. First stage still. The arrangement of the first stage still is shown. Water enters on the right and emerges on the left.



Fig. 12. Method of rinsing glassware. The flask is rinsed with pure water from condensing steam.

a system of steam generators had to be devised to keep the water in the stills agitated with superheated steam in order that purification beyond the first stage could be used with any degree of success, see Fig. 13.

The steam generator is simply a glass tube inserted in the boiling flask, drawn out to a point, bent into a double U shape and wrapped with a nichrome heater. A head of water is kept on the generator by condensing a small amount of water into the generator tube before it gets to the condensing column. This head is kept from flowing into the boiling flask by the nichrome heater which is adjusted so that about 200°C steam only enters the boiling flask.

No steam generator ever failed in service and they were kept in continuous operation for a period of two months which attests to the dependability of the steam generators. When distilling in the final stages of distillation, the need for the steam generators can be dramatically demonstrated by shutting off one and watching the temperature in the boiling flask rise degree by degree with no boiling. This is a dangerous procedure because the great amount of energy stored in the water is released with explosive force when boiling does begin again.

When a bubble of steam comes to the surface in the boiling flask, it bursts and sprays tiny water droplets in all directions, some of which are light enough to be carried into the condensing column. Smith⁵³ has shown that evaporating droplets do not evaporate completely. This is probably due to surface active materials which are concentrated in the surface. Thus, small re-evaporation nuclei which form as the result of the evaporation of sprays can effectively transport low vapor pressure organic materials through a still. This action effectively cancels part of any purification which might be effected by the distillation.



Fig. 13. The steam generator.

It was found that this problem could be effectively overcome by using small stainless steel chips closely packed in an 18 inch column above the boiling flask.

This method is especially effective when enough cooling is provided on the column containing the stainless steel chips so that sufficient water is condensed on the chips to continuously wash them clean. Resultant distillation rates are consequently lowered. The nuclei leaving the boiling pot are taken out by the chips of the column packing by both importion and diffusion. Impaction requires a finite flow velocity and sharp edged plates while diffusion requires time and a small diffusion distance. The distillation rate was about one-fourth liter per hour.

A commercial Corning still was modified for use as an intermediate distillation unit before the water was put into the final stage, see Plate 8. The final stage is designed to remove the last traces of atmospheric gases. This intermediate distillation was considered necessary because the water was of necessity kept in ordinary glass jugs after the first two distillation stages.

The final stage of the distillation is not a continuous operation but a batch operation. This stage has a five gallon flask, so that a reasonable batch may be processed, see Plate 9. A single batch of this size suffices for any cloud chamber experiment yet devised in this laboratory. Water is continuously boiled and recondensed in the final stage while maintaining the pressure at five to ten pounds above atmospheric pressure. Gases dissolved in the water are released as the temperature rises and are allowed to leak out through a small capillary leak. This action is continued for two or three days or until about onefourth of the water is lost to the atmosphere. For an operation of this



Plate 8. Corning still. A front view of the modified corning still is shown.



Fig. 14. Third stage still.



Plate 9. Third stage still. The lower part of the third stage still is shown. The packed column is on the left, the temperature sensor is in the center and the pressure sensor is out of the picture on the right. type, continuous automatic pressure and temperature sensing are required to maintain safe operation. The pressure stays fairly constant because the temperature is held to a preset value of approximately 108°C.

Distillation from the final stage was done directly into a five gallon jug containing a helium atmosphere. There is no reason, however, the distillation could not be done directly into a vacuum bottle so that all gases are eliminated and the purest possible water obtained. Since the final distillation occurs in the cloud chamber, the materials dissolved from the glass are not troublesome. If the glass jugs are used for the same purpose for some time they may eventually become very clean.

A system of more automatic operation of the first stages than had been used is currently being incorporated into the system. Improvements include automatic filling and temperature control of the first stage stills. Other improvements include continuous conductivity and periodic mass spectographic checks of purity. There is every reason to believe that this distillation system is very effective.

2-9. Preparation of the chamber. The process of readying the cloud chamber for a particular data run begins several days in advance of the data taking process itself. Assuming that all equipment is in operating condition, the chamber is thermostated at the desired temperature. Room temperature must be kept five to ten Fahrenheit degrees below the chamber temperature for proper thermal regulation. Air tanks must be maintained at a temperature near that of the cloud chamber. Otherwise too much heat is pumped into or out of the lower drive chamber for good temperature stability. Some adjustment of heater controls is usually required to maintain proper heat input. The constant temperature bath which is used as a thermocouple reference is usually maintained within one-half degree of the cloud chamber temperature. These thermocouples are used to thermostat the cloud chamber.

In order to assure gas purity, the cloud chamber is flushed several times immediately before each data run. This procedure eliminated gaseous impurities which may have diffused from the glass walls or the water. If a change in the gas type was made, flushing was done on two consecutive days prior to operation of the chamber. This allowed time for the former gas to diffuse out of the water pool in the chamber.

Even though it would have been desirable to use new water for each set of data, this was not possible because of the difficulty in changing the water in the chamber. No check was made of water purity after the water was in the cloud chamber, but because of the close agreement of the data taken at widely spaced intervals, it is believed that neither water purity or gas purity affected the nucleation rates measurably during the course of the entire experiment. As discussed in the section on water purification, there is little hope of having an atmosphere which is completely free of gaseous impurities. Since the author's data corresponded so well with that of the other researchers in this laboratory who used various means of water purification, it is felt that water purity is not a problem in these experiments.

<u>2-10.</u> The cloud chamber program. For the measurement of homogeneous nucleation rates, the cloud chamber is programmed as shown in Fig. 15. Expansion AB requires about 0.2 sec. The interval BC can be varied from 0.01 sec. to about 1. sec. The slight compression CD reduces the supersaturation by an amount sufficient to stop all subsequent nucleation.





After the droplets have had ample time to grow to photographable size, the xenon flash is triggered and the droplet density is photographed.

An electrostatic clearing field of 80 volts per cm. is used to sweep out ions which are produced in the cloud chamber between expansions. The clearing field is turned *off* just prior to the onset of condensation. Any tracks which are formed during the sensitive time of the chamber appear as easily recognizable ion tracks. Such tracks are carefully avoided when drop counts are made. Therefore, the data presented in the course of this work is not biased by the presence of ions.

Two cleaning expansions were used between data expansions to insure that re-evaporation nuclei had been eliminated. The second cleaning expansion was photographed in order to determine the background level. Except for additional sophistication in the experimental technique, the method employed is basically the same as that employed by Allard⁴⁹.

CHAPTER III

CLOUD CHAMBER THERMODYNAMICS

The most important piece of information to be extracted from the data for a cloud chamber expansion is the state of supersaturation of the atmosphere as a function of time during those parts of the cloud chamber cycle when nucleation is taking place. When the cloud chamber was used as a particle detector for high energy nuclear physics, the supersaturation needed to be controlled with only moderate accuracy so that drop formation occurred only on ions and an exact knowledge of the state of supersaturation was not necessary. In addition, it was useful to know for what period of time the cloud chamber was actually sensitive to the ions. Experiments measuring homogeneous nucleation rates and droplet growth are critically dependent upon an exact knowledge of the supersaturation as a function of time during the cloud chamber cycle.

There is no way to directly measure the water vapor content of the cloud chamber atmosphere during an expansion. The cloud chamber establishes a state of supersaturation by means of an adiabatic expansion as shown in Fig. 16. In the initial condition a noncondensible gas is saturated with water vapor at temperature T₁. The dashed line shows the course taken by an adiabatic expansion. At a representive time the water vapor pressure has been reduced from P₁ to P₂ by the expansion itself. The temperature has dropped from T₁ to T₂. The equilibrium vapor pressure corresponding to T₂ is P₂ so that the supersaturation ratio established by the expansion is P₂/P₂.



Fig. 16. Creation of supersaturation in helium and water vapor by means of an adiabatic expansion.

Ц

3-1. Measuring temperature directly. It had been hoped that an instrument could be developed which would measure temperature with sufficient speed and accuracy to permit its use in the cloud chamber. In recent years fine wire bolometers and fine wire thermocouples have been available which are seemingly fast enough to measure temperature accurately in an expanding gas. However, in a moist gas condensation occurs on the thermocouple, liberating the latent heat of condensation. The wet thermocouple tends to approach the equilibrium temperature T_2 shown in Fig. 16. The exact temperature of the thermocouple will depend upon the rate of change of conditions in the cloud chamber and upon the surface properties of the water film on the thermocouple. Moreover, surface conditions do not reproduce nicely and even this condensation does not take place reproducibly. The prospects for overcoming these effects are not favorable so that all temperature data must be obtained from other sources when the cloud chamber is in a supersaturated state.

3-2. Reliability of volume measurements. Packwood⁵⁰ has shown that calculations of temperature made from volume expansion ratios give erroneous results. This can be readily understood in terms of the thermodynamic processes taking place in the cloud chamber. During the expansion the walls of the apparatus remain at temperature T_1 , Fig. 16. Since the interior gas is at a much lower temperature, T_2 , and since the wet cloud chamber walls would have to be at temperature T'_2 in order to be in equilibrium with the existing vapor density, P_2 , the rapid diffusion of both heat and vapor takes place from the wet chamber surfaces. The net effect is that boundary layers (affected by diffusion processes) expand and thereby produce a compressive effect at the interior of the chamber. The sensitive volume of the cloud chamber as a whole is nonuniform and it makes no sense to talk about the adiabaticity of the whole volume. In fact, volume measurements are useless because the computational complexity of dealing with the real nonuniform gas situation is unduly great.

If the expansion process is slow enough so that shock waves are not created, the pressure throughout the system will everywhere be the same. Moreover, diffusion processes are inherently slow so that a finite time is required for the central regions of the cloud chamber to be sensibly affected, Figs. 17 and 18. Even though the compressive effect has been active, the center of the chamber remains truly isentropic until actual diffusion reaches these regions in perceptible magnitude.

Ordinarily, the final temperature is calculated from one of the ideal gas relationships for a constant entropy process.

$$\Gamma V^{\gamma - 1} = \text{constant}
 (3-1)$$

$$P V^{\gamma} = \text{constant}$$

$$T P^{(1-\gamma)/\gamma} = \text{constant}$$

The first two involve the volume and are not useful where great accuracy is required. Before using the last equation, however, an appropriate gamma for the equation must be known. Since the cloud chamber operates with a gas mixture, the difficulty in finding an appropriate gamma is magnified even more than for a single component gas system, Packwood⁵⁰ does a thorough analysis of error propagation and concludes that an error of 0.06 in the supersaturation (at a supersaturation of 5.0) produces an error of 100% in the nucleation rate. This can be caused by an error of only 0.005 in the composite gamma used in the ideal gas relationship.



Fig. 17. Temperature Diffusion Profiles in a Finite Cylindrical Cloud Chamber.



Fig. 18. Vapor Diffusion Profiles in a Finite Cylindrical Cloud Chamber.

3-3. Method of Richarz. Therefore, if the third of Eqs. (3-1) is to be employed, a gamma which is averaged over the range of thermodynamic coordinates must be employed. Methods for surmounting this difficulty are discussed in the following section. Richarz⁵⁵ derived a relationship for determining the composite gamma of a system of two noninteracting gases when the respective partial pressures and gammas are known. The following is a variation of the translation of his procedure as given by Laby⁷.

Assign a mass of 1 to the mixture so that each component is $1-\mu$ and μ respectively. Let the densities by ρ , ρ' , ρ'' which are understood to be measured under standard conditions. The specific heats at constant volume are C_v , C'_v , C''_v , and the ratio of the specific heats γ , γ' and γ'' .

Conservation of energy requires

$$C_{v} = C_{v}'(1-\mu) + \mu C_{v}'' = C_{v}' + \mu (C_{v}''-C_{v}')$$
(3-2)

The specific volume of the mixture is

$$\frac{1}{\rho} = \frac{1}{\rho} (1-\mu) + \mu \frac{1}{\rho''} = \frac{1}{\rho} + \mu (\frac{1}{\rho''} - \frac{1}{\rho'})$$
(3-3)

Let $C_{p}, C_{p}^{\prime}, C_{p}^{\prime\prime}$ be the specific heats at constant pressure

$$C_p - C_v = \frac{R}{JM} = \frac{1}{\rho} K$$
 (K= constant) (3-4)

since M is proportional to p, and R and J are constant

whence
$$\frac{1}{\gamma - 1} = C_V K$$
, $\frac{1}{\gamma - 1} = \rho' C_V' K'$, $\frac{1}{\gamma'' - 1} = \rho'' C_V'' K''$ (3-5)
Now eliminate μ in Eqs. (3-2) and (3-3)

$$C_{v}(\rho''-\rho') = (\rho-\rho')\rho''C_{v}'' + (\rho''-\rho)\rho'C_{v}'$$
(3-6)

and by Eq. (3-5)

$$\frac{1}{\gamma - 1} = \frac{\rho - \rho'}{\rho'' - \rho'}, \frac{1}{\gamma'' - 1} + \frac{\rho'' - \rho}{\rho'' - \rho'}, \frac{1}{\gamma' - 1}$$
(3-7)

This equation looks at first sight to be most adaptable to the conditions of cloud chamber work. The problem which arises is that gamma (even for the most ideal gas, helium) is a function of the thermodynamic co-ordinates to such an extent that it is not constant over the range of even a typical 30° expansion.

3-4. Temperature-entropy diagram method. Accurate temperature calculations have been made using a method devised by Schmitt⁵¹ and Dawbarn⁵². This method makes use of the temperature-pressure-entropy diagrams for the individual components of the gas. The expected temperature change during the expansion is determined by interpolating between isobars on a line of constant entropy. This temperature change allows one to calculate a gamma which is automatically averaged over the thermodynamic co-ordinates (sometimes referred to as an effective adiabatic index). The formula of Richarz is then used to calculate the composite adiabatic index. This procedure neglects the entropy of mixing and the exchange of energy between the component gases. As a result the method gives good results only when one of the gases is the dominant species.

Another method which should be even more accurate involves making a composite entropy diagram for the gas mixture and finding the temperature directly from the diagram. This latter method has the serious drawback in that these composite entropy diagrams are very cumbersome to make and a new diagram is needed every time the initial temperature is changed since the mole fraction of vapor changes with temperature. In addition, this method is not adaptable for use with a high speed digital computer so all work has to be done by hand.

3-5. Comparison of methods of temperature determination. All temperature calculations ultimately come from an equation of state for the gas. Equations of state use three variables to completely describe the gas. Cloud chamber expansions are adiabatic so one of the variables used must be entropy. It may be set equal to a constant during the calculation. Derivations of these equations are given in standard thermodynamics texts.^{57,58} Pressure, temperature and entropy are the variables chosen when pressure is measured during the cloud chamber expansion.

The most accurate method is that which integrates directly the equation

$$ds = C_p \frac{dT}{T} - \left(\frac{\partial V}{\partial T}\right)_p dP \qquad (3-8)$$

This equation refers to one mole of a single gas. For a gas mixture the mole fractions n_1 and n_2 are used where $n_1+n_2=1$ and the total entropy change is the sum of the individual entropy changes.

$$ds = n_1 ds_1 + n_2 ds_2$$
 (3-9)

During an expansion

$$ds = 0 = n_1 ds_1 + n_2 ds_2$$
 (3-10)

A numerical solution of this equation including the most accurate values of the heat capacities and compressibilities, $(\partial V/\partial T)_p$, has been used to

provide calculations of temperature. Calculations of temperature using Richarz's method for average gamma as described by Kassner and Schmitt³⁷ were also used and compared with the results of the preceding method. It is concluded that there is essentially no difference in the results obtained with the two methods.

When immediate calculations are needed and no computing machine is available, the graphical method of temperature determination is useful. This makes the graphical method very adaptable to uses involving Aitken nuclei counters because of the independence from office machines.

The course of a perfectly adiabatic expansion is a vertical line on the entropy diagram. Latent heat is easily accounted for through the definition of entropy ds=dQ/T. When using air, there is so little difference in the diagram with a small mole fraction change that small changes in the mole fraction due to droplet growth are negligible.

Use of this technique complete with droplet growth corrections is outlined in detail by Kassner⁵⁹ et al. Refering to the diagram, Fig. 19, the expansion begins at the top of the diagram proceded straight down to point A where a correction is made for vapor depletion (negligibly small at this time). At points B and C the corrections for latent heat begin to be sizable. An excessively large droplet concentration, 10,000 droplets per cm³ was assumed in the calculation to make the effects show up vividly. Comparison of this method with the exact integration technique shows essentially no difference in accuracy.

In conclusion, any of the three basic methods of temperature calculations may be used with confidence. Each has its advantages and disadvantages depending upon the situation. Richarz's method for average gamma is most useful where a desk calculator is used and vapor



Fig.19. Determination of the True Temperature from the Temperature-Pressure-Entropy Diagram.

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i.



Fig. 20. Determination of the True Temperature by the Entropy Method. The Diagram Corresponds to the Expansion Shown in Fig. 19.

depletion effects are negligible. The graphical technique requires a tedious plotting of a new entropy diagram for each new mole ratio, but is very adaptable where no calculating machines are available and where the same initial conditions are used. The exact numerical integration technique is not adaptable to hand calculations, but is the only method easily programmable for large computing machines when taking all corrections into account. It appears that all three techniques will continue to be used as each has its own range of usefulness making it uniquely adaptable to a given situation.

<u>3-6.</u> Experimental test of equations of state and measuring techniques. Use of temperature entropy diagrams requires knowledge of entropy values to quite high accuracy. These values have ordinarily been derived by differentiating the equation of state which is risky at best. There is not and has been no doubt that the accuracy of these equations of state is quite good. The following question arises. Since the derived functions such as heat capacity and entropy are obtained by taking first and second order derivatives, just what accuracy is retained in these derived functions? F. Din⁶⁰ most aptly states the problem in his work on argon:

The first differential coefficient was difficult enough to derive even with nominal accuracy whilst values derived for the second differential co-efficient were uncertain in the extreme. It was decided to perservere and values of

$$\left(\frac{\partial \mathbf{r}}{\partial \mathbf{t}}\right)_{\mathbf{p}}$$
, $\left(\frac{\partial^2 \mathbf{r}}{\partial T^2}\right)_{\mathbf{p}}$ and $T\left(\frac{\partial^2 \mathbf{r}}{\partial T^2}\right)_{\mathbf{p}}$

were derived, tabulated and smoothed, values of C_p , entropy and enthalpy were then calculated by integration and it was immediately apparent that they were untenable. They showed

considerable irregularities when plotted and were in no sense systematic.

Considering that the data for argon is perhaps the most accurate and consistent of that for any gas, also that argon is the most ideal of all gases after helium and neon, it is not difficult to imagine inconsistencies arising in the entropy tables.

It was decided that in order to obtain good results with nucleation work, dependable data on the various gases in the cloud chamber must be obtained. The aim was to take sufficient data to check the existing data. It was felt that the accuracy that could be obtained with static measurements was not better than had been obtained by other experimenters, for instance in the case of argon at Leiden by Crommelin and Onnes⁶¹ and coworkers, at Reichsanstalt by Hobborn,⁶² by Masson^{63,64} and coworkers, at the Van der Walls laboratory by Michels⁶⁵ and coworkers and by Bridgman^{66,67}. Therefore, a means was sought to exploit the advantages of the cloud chamber. Data would not be taken recording pressure, volume and temperature, but recording only pressure and temperature during a constant entropy process. Data of this type plotted on a temperature entropy diagram, should yield a vertical line if our results are consistent with the diagram.

The remaining question is, just what sort of accuracy must an experiment provide so that the data obtained is of equal or better quality than the published data. A true error analysis of published data is difficult to carry out. One can judge, however, from the number of significant figures published in a table, the confidence with which an author rates his calculations. This confidence is expressed by publishing one more significant figure than the accuracy of the computations would indicate. This is a necessary evil, however, in order that internal

consistency might be obtained. Using this criterion, the data taken should exceed the accuracy of published data if pressure measurements are maintained to an accuracy of 0.5 mmHg and temperature measurements to 0.05 C°.

With very little modification, the Wilson expansion chamber of this laboratory may be used for making thermodynamic measurements. Temperature and pressure are recorded during an expansion or compression of a gas under conditions of constant entropy. These measurements must be made with extreme accuracy and speed.

<u>3-6.1</u> Obtaining a pure atmosphere. A new cloud chamber was constructed for this work. It has the advantage of a larger available piston motion so that sizable volume ratios might be obtained without partially filling the sensitive volume with water.

Assembly of the new cloud chamber was done with the greatest care. Every bolt, nut and screw was cleaned with the same care that one would use in a hospital surgical room. The sensitive volume was exposed only to the rubber diaphram, O-ring seals, stainless steel and glass.

In order that no water vapor or other volatile material enter the chamber, a dry ice and acetone cold trap was installed in the inlet line to the sensitive volume. This cold trap was kept in operation at all times during the period from assembly of the chamber to the completion of the data taking. Each time a new gas or gas mixture was used, a flushing operation was performed which virtually eliminated all traces of the former gas. A vacuum pump was used to remove as much gas as possible. New gas was then run into the chamber so that the final pressure was about three atmospheres. This gives a flushing action whereby three-fourths of the gas is removed each time so that the remaining gas of the former kind after filling is about twenty-five percent of the total. Usually eight flushing operations of this type were performed for each gas exchange with the result that the purity of the gas as it came from the cylinder was the determining factor in its purity in the cloud chamber.

A pressure transducer such as is used in this laboratory has a natural frequency of 40,000 Hertz. The oscillograph galvanometers have the slowest response of any component in the pressure detection system with a flat response from zero to 240 cycles per second (Heiland type M400-120, 8.62 MV/in, undamped natural frequency 400 cycles per second). Amplifier frequency response is good enough that no difference in gain is noticed for a d.c. signal or a ten kilocycle signal. The net result of the pressure detection system is that pressures are measured to ± 0.02 percent accuracy with a time response which is short compared to anything happening in the cloud chamber.

The thermocouple employed in these measurements was designed to minimize thermal pertubations due to the thermocouple itself while maintaining the lowest possible resistance. The thermocouple is shown in Fig. 21 and its position in the cloud chamber in Fig. 22. It is necessary to maintain the lowest possible resistances for the thermocouple element since the noise level at the output of the amplifier is roughly proportional to the input impedence. In the case of chromel-alumel the output is of the order of forty microvolts per degree centigrade. In order to read a temperature to one-hundreth of a degree, it is therefore necessary to know the voltage coming from the thermocouple to better than one microvolt.





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Fig. 22. The cloud chamber used for measuring gas temperature with a thermocouple.

Considering that wide band d.c. amplifiers generally have a noise level referred to the input of the order of tens of microvolts it is easy to see the difficulty in trying to accurately read an output signal to one microvolt. The Model 3101 California Instruments amplifiers used in this laboratory have a rated noise level of seven microvolts referenced to input. Four amplifiers were available so a complete check of each was made to determine which had the lowest noise level in the circuit configuration used with the thermocouple. The amplifiers were run in the potentiometric mode since it showed consistently lower noise levels than the differential mode.

Noise level tests with shorted input were made and recorded for each amplifier. It turned out that two of the amplifiers just met specifications while two were significantly better. Of the other two, one had a noise level of five microvolts peak to peak. The thermocouple used has a resistance of approximately 390 ohms. This value of carbon resistor was then placed across the input of each amplifier and a check made on the noise level. As expected, the noise level was higher, but only by about two microvolts in the case of the lower noise amplifier. A triply shielded twisted pair cable 25 feet long was constructed which has three 95 percent coverage shields. This cable was attached to the amplifier input and shorted, at the other end. Shorted at the end, it gave practically no noise increase, but with the 390 ohm resistor in place, the noise level was intolerable. It was therefore decided that the amplifier must be placed as closely as possible to the thermoccuple so that stray capacitive pickup would be minimized.

After the thermocouple was installed and connected to the amplifier, several configurations of grounding the shields were tried. At best, the

noise level referred to the input was down to about eight microvolts peak to peak. This included about five microvolts due to the amplifier, one microvolt due to the bias module and two microvolts due to stray pickup by the thermocouple.

It should also be mentioned that the thermocouple hanging out in the center of the cloud chamber as it did, acted as a very good antenna, picking up signals from every valve and a.c. power line in the vicinity. There was no alternative but to encase all operating valves in a copperclad one-sixteenth inch iron case. Even the sola transformer operating the amplifier had to be moved from under the chamber. When all these changes were made, the noise level was again down to about eight microvolts peak to peak of mostly sixty cycle noise with some 400 cycle noise from the chopper in the amplifier.

Luckily, the noise peaks from the pressure signal and from the temperature signal were in phase. There also was a flat place in the signal between each peak which corresponded to a period of zero noise level. Because of this, even though noise was present in the signal, readings were taken every 1/120 second during the quiet period of the noise cycle, thereby achieving the same effect as if the noise were a factor of ten smaller. One disadvantage of this is that in order to get a large number of data points on each run, the expansion or compression time had to be increased almost to the limit. A block diagram of the temperature measuring system is given in Fig. 23.

<u>3-7.</u> Thermal characteristics of fine wire thermocouples. The output response of fine wire thermocouples to a changing environment created by an expanding gas is more complex than many investigators have recognized. The seemingly instantaneous response to a fast expansion



Fig. 23. Block diagram of the temperature measuring system.

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has been erroneously assumed to be indicative of the accuracy with which the thermocouple follows the temperature change. It was the purpose of this investigation to place the interpretation of fine wire thermocouple measurements on a sound basis. The commercial availability of 0.0005 in. dia. thermocouple wire and the relative ease with which thermocouples can be fabricated from this size material strongly recommend it for temperature measurements. However, data taken in an expansion cloud chamber with such fine wire thermocouples indicated that the thermal capacity of the thermocouple itself was not negligible.

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. Now under these circumstances the rate of diffusion of heat away from the thermocouple will adjust itself so that a steadystate 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). The establishment of the steady-state requires of the order of 10^{-4} sec. and so we see a very fast response to sudden environmental changes. But the temperature being indicated by the thermocouple 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.⁶⁸

$$T(\mathbf{r}, \mathbf{t}) = 1 - 2 \sum_{n=1}^{\infty} \frac{J_0(\alpha_n \mathbf{r}/\mathbf{r}_0)}{\alpha_n J_1(\alpha_n)} e^{-\left(\alpha_n^2 \frac{\kappa}{C_p} \frac{\mathbf{t}}{\mathbf{r}_0^2}\right)}$$
(3-11)

where κ is the thermal conductivity, C_p is the heat capacity, r_0 is 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 $\alpha_4 = 11.79$. The chromel-alumel thermocouples used were made from 0.005 in. dia. 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 microsec. After ten microsec, the center is within two percent and after 100 microsec it is within 10^{-8} percent of the outside temperature. Therefore, the relaxation time of the thermo-couple 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.^{69,70}

Since things are everywhere the same in the direction of the axis of the *stretched out* thermocouple wire, the problem reduces to a two dimensional heat flow problem.

$$C_{p}\frac{\partial T}{\partial t} = \kappa_{g} \frac{\partial^{2}T}{\partial x^{2}} + \frac{\partial^{2}T}{\partial y^{2}} + f(x,y,\dot{x},t)$$
(3-12)

where $C_{\boldsymbol{p}}$ is the heat capacity of the gas at constant pressure and $\kappa_{\boldsymbol{g}}$ is

the thermal conductivity of the gas. $(f(x,y,\dot{x},t)$ is a source function which allows for varying expansion speeds and also allows for the motion of the thermocouple through the gas. Only numerical solutions of this problem were attempted.

Figs. 24 and 25 show the results for an expansion in dry argon. The 0.0005 in. dia. (12 microns) thermocouple was located 3 in. 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. 25 indicates close agreement between the theoretically predicted thermocouple temperature and the measured thermocouple temperature. The difference is about 1.5 C° after a short time for a gas velocity of 2 cm/sec. 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 0.1 C° rise in temperature in a cylinder of gas with a radius of 0.3 cm. This is only misleading since the small gradients make the dispersal of the evolved heat very slow.

Israel and Nix⁷¹ investigated the thermodynamic processes in the Pollak counter by inserting a fine wire thermocouple into the dry chamber, Fig. 26. 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.

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. Moreover, the equations of state for the gases tested check very well against the data obtained in these experiments.



Fig. 24. Measured temperature and pressure in dry argon.



Fig. 25. Temperature difference for the expansion shown in Fig. 24.





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CHAPTER IV

NUCLEATION MEASUREMENTS

The first homogeneous nucleation measurements to be made in this laboratory were made by Allard and Kassner.²⁰ Later, additional measurements were made by Kassner and Schmitt³⁷ with some improvements in technique and data analysis. One of the principal problems was related to the inability to adequately account for the various aspects of droplet growth: bulk depletion of available vapor, effect of diffusion profiles around growing droplets on subsequent nucleation (dead space) and competition for the available vapor supply by closely spaced droplets. Droplet growth measurements are conspicuously lacking and much effort has been expended in studying these effects.

The author has attempted to elucidate the disparity which exists in the literature on the nucleation rate of water vapor by providing comprehensive measurements of the nucleation rate as a function of supersaturation, temperature and sensitive time. Fig. 27 shows data representing the average nucleation rate as a function of average supersaturation for narrow nucleating pulses of sensitive time 0.01 sec. The circles represent data in argon while the crosses represent data in helium, both for an initial starting temperature of 22.5°C. Note that the data in Argon are noticeably and reproducibly higher. Dawbarn⁵² first observed this effect in this laboratory. Classical theory predicts no effect on the nucleation rate due to the nature of the non-condensible gas. This effect might be the manifestation of the alteration in the free energy of the critical cluster due to the inclusion of an argon



Fig. 27. Comparison of the nucleation rate in argon and helium.

atom in a clathrate like structure of water molecules in the embryo. Pauling^{72,73} has hypothesized such a hydration scheme for xenon in the bulk water structure. Fig. 28 shows a possible arrangement of 20 molecules when an ion is included in the cluster. Fig. 29 shows a possible arrangement of 24 neutral molecules into a clathrate like structure. These two arrangements are dodecahedral and tetrakaidecahedral respectively and are the smallest possible configurations where each oxygen molecule has three hydrogen bonds and the bond angles are maintained near the normal bond angle. If such a configuration actually exists, these structures should be particularly stable and there should be a minimum in the free energy curve for these particular configurations.

Parunge and Lodge⁷⁴ have measured the effect of nonpolar gases upon the freezing point of supercooled water. Their water was of sufficient purity that small droplets normally froze at -16°C. Their data is reproduced in Fig. 30. It is seen that there is a definite ordering of the water molecules due to the presence of the inert gas. Claussen and Polglase⁷⁷ have suggested that the larger voids required in the liquid by krypton and xenon might well be dodecahedral or tetrakaidecahedral, thereby accounting for the discontinuity in the freezing points. If the critical cluster for nucleation of the liquid from the vapor is of the clathrate structure, krypton and xenon might well fit into the critical cluster. This would result in a sizeable lowering of the free energy of the cluster and the nucleation rate of water vapor in these gases would be correspondingly increased.

Fig. 31 shows the number of droplets nucleated per cubic centimeter as a function of peak supersaturation for the narrowest possible pulses. Both the 12.5°C and 22.5°C data exhibit prominent inflections while the 31°C data gives the indication that an inflection may exist



Fig. 28. The arrangement of hydrogen bonds for twenty water molecules in a dedecahedral configuration.



Fig. 29. The arrangement of hydrogen bonds for twentyfour water molecules in a tetrakaidecahedral configuration.



FIG. 1. Increase in the temperature of spontaneous freezing of 0.1-ml dreps of solutions of two series of non-polar gases, compared with "pure" water, plotted against the entropies of hydration of the solute gases.

Fig. 30. Data of Parungo and Lodge.



Fig. 31. Number of droplets nucleated as a function of peak supersaturation. The starting temperature is (Δ) 41.5°C, (+) 32.5°C, (x) 22.5°C and (∇) 12.5°C.

for higher droplet densities than could be measured in this experiment. If the number of droplets (corresponding to the plateau of the inflection for the 12.5° and 22.5° curves and the number estimated where the shoulder might lie for the other two helium curves) are compared, it is found that the number is nearly proportional to the initial vapor pressure of water in each case. Schuster⁷⁵ has measured the nucleation rate of water vapor in an argon atmosphere as a function of supersaturation with an initial temperature of 24°C. using light scattering techniques. His data agrees reasonably well with the author's both in slope and magnitude. His data teproduced in Fig. 32 shows slight evidence of an inflection at about the same point as the author's.

Fig. 33 is a correlation of the author's data with temperature dependence data found in the literature. Curve No. 1 is the author's data for an estimated droplet density of 100 drops/cm³ or a nucleation rate of 10,000 drops/cm³sec. Curve No. 2 is the corresponding data for 1 drop/cm³ or a nucleation rate of 100 drops/cm³sec. The dashed lines represent the path of the adiabatic expansion of the cloud chamber for the four sets of data. Curve No. 3 is the data of Volmer and Flood¹⁵ for a rate of 4 drops/cm³sec as estimated by Allard and Kassner.²⁰ The latter corresponds very closely to the author's data for a rate of 1 drop/cm³sec both in magnitude and in slope. The ϕ 's represent Powell's data for an estimated nucleation rate of 10⁵ drops/cm³sec. The author's data, extrapolated to this nucleation rate, agrees reasonably well with that of Powell both in magnitude and in slope. Curve No. 4 represents the homogeneous nucleation rate data of Sander and Damkohler⁷⁶ for their estimated rate of 1 drop/cm³sec. The θ 's are Powell's ion limit data.



FIGURE 5-5 Variation of Modified Nucleation Rate with Supersaturation and Temperature

Fig. 32. Data of Schuster.



Fig. 33. Temperature dependence of the nucleation rate. The dashed curves are the paths of the adiabatic expansion for starting temperatures of (A) 41.0°C, (B) 32.5°C, (C) 22.5°C and (D) 12.5°C. Line (1) represents the author's data for 10,000 drops/cm³ sec, line (2) is the author's data for 100 drops/cm³ sec, line (3) is the data of Volmer and Flood and line (4) is the homogeneous nucleation data of Sander and Damkohler.

It is impossible to reconcile Sander and Dankohler's consistently low results unless they actually observed nucleation on ions. Curve No. 4 also agrees well with our results for nucleation on ions.

Fig. 34 shows the decrease in the measured nucleation rate as a function of time for supersaturations of 4.4, 4.9, 5.2 and 5.6. The dotted lines are the expected decrease resulting from the effects of droplet growth as calculated using the method given in the following chapter. It is seen that the decrease in the nucleation rate cannot be due to the effects of droplet growth. Moreover, the greatest deviation occurs at the supersaturation corresponding to the inflection in the curve of Fig. 31. In the data where the initial temperature of the cloud chamber was 12.5°C, the cut-off in the nucleation process with increasing sensitive time is even more pronounced. In this case, everything that is going to nucleate does so in the first 0.01 sec. for supersaturations between 6.0 and 6.5. Because of the unexpected nature of this data, both the 12.5°C and 22.5°C data were repeated several weeks after the initial data was taken. There was complete agreement with the previous data for both temperatures so it was felt that the data were accurate.

It is the cut-off phenomenon which indicates the presence of a heterogeneous nucleating agent. This effect is clearly not due to nucleation on ions. A clearing field of 80V/cm is maintained until the beginning of the expansion. Old ions are swept out and ion tracks show clearly at a much lower supersaturation.

Nucleation rate measurements have been carried out in this laboratory since 1960 and the results have all been self-consistent. The purity of the helium-water vapor system used in the experiments has varied widely without any change in the results. Water purification methods



Fig. 34. Nucleation rate as a function of time. The starting temperature is 22.5°C and the supersaturations are left to right respectively 4.4, 4.9, 5.2, and 5.6.

have included deionization columns, ordinary glass distillation systems, distillation columns preceded by a charcoal absorption cell and the method described in Chapter II. None of these have produced any variance in the results. It is difficult to imagine any ordinary impurity which would be present with the observed consistency and in the small concentrations observed in these experiments (30 to 100 molecules/cm³). Because the concentration of the impurity varies in proportion to the initial vapor pressure of the water, it seems likely that this particular nucleating agent is a neutral product formed through the action of ionizing radiation on the water vapor.

The data in the literature seems to be separable into two basic groups. One, such as the data of Volmer and Flood,¹⁵ where measurements have been made yielding very small droplet densities and another, such as the data of Powell,¹⁶ where measurements have yielded large droplet densities, see Fig. 33. Both of these are in good general agreement with the author's results, so it seems likely that the other observations have also recorded the same effect. Moreover, this mode of heterogeneous nucleation retains many of the characterististics of the homogeneous nucleation process and possesses a critical supersaturation limit very close to that predicted by the Becker-Doring theory. The similarity with the homogeneous nucleation process gives a clue as to the nature of the process.

Since the normal rate of ionization is due to both cosmic rays and natural radioactivity, the rate of production of molecules of the nucleating agent would be expected to be fairly constant with the largest fluctuations being due to cosmic ray showers. Cloud chambers cycling at a regular rate would be expected to experience about the same buildup between expansions whereas devices operating with irregular cycle times could easily experience large deviations in results. The scatter

of data points experienced in this work near and below the plateau is reminiscent of that found by Allard⁴⁹ in his data for nucleation on random ionization. It is believed that this may explain the scatter of results displayed by Katz and Ostermier³⁹ for water since they probably cycled their diffusion chamber at irregular intervals.

4-1. Theory. In classical nucleation theory it is assumed that small clusters of water vapor molecules possess the properties of bulk water, i.e. they have a definite temperature and their surface may be characterized by the bulk surface tension for water. When a vapor molecule impinges upon a cluster, a quantity of Gibbs free energy, kTlnS, is released in the transformation from gas to liquid, assuming that the cluster may be considered as bulk water. However, the volume of the eluster must increase with the addition of each molecule so that some of the above energy goes into the creation of new surface, the total surface free energy of the cluster of radius r being $4\pi r^2\sigma$. Hence, the energy of formation of a homogeneous cluster is

$$\Delta G_{\rm h} = -NkT \ln S + 4\pi r^2 \sigma \qquad (4-1)$$

= -4/3 \pi r^2 n_1 \ln S + 4\pi r^2 \sigma

where N is the number of molecules in the cluster, k is Boltzmann's constant, T is the absolute temperature of the gas, S is the supersaturation, σ is the surface tension and n_L is the molecular density of liquid water.

Since the volume has an r^3 dependence and the surface has an r^2 dependence, there is always a maximum point in the plot of free energy against radius. The radius to which the maximum free energy belongs

is called the *critical radius* and is found by differentiating the Gibbs free energy. The critical radius

$$\mathbf{r}^* = 2\sigma/n_{\rm L}kT\ln S \tag{4-2}$$

so that the free energy ΔG^* of the critical cluster becomes

$$\Delta G^* = 4/3 \pi r^{*3} n_L k T \ln S + 4 \pi r^{*2} \sigma \qquad (4-3)$$

$$= \frac{16\pi\sigma^{3}}{3[n_{L}kTlnS]^{2}}$$
(4-4)

where $4/3 \pi r^{*3}n_L$ is the number of molecules in the critical cluster.

It is conjectured that a chemical reaction may take place between a molecule of the heterogeneous nucleating agent and a water molecule. It is also assumed that clustering proceeds upon this complex chemical entity in much the same way that it does upon the clusters in the homogeneous case. The energy of formation of a heterogeneous cluster, i.e. a cluster including the chemical bond of energy ε , is

$$\Delta G_{c} = -4/3 \pi r^{3} n_{t} k T \ln S + 4 \pi r^{2} \sigma - \epsilon \qquad (4-5)$$

It is seen that because the ε term is independent of radius, the critical radius of the heterogeneous cluster is identical with that of the homogeneous cluster (neglecting the size of the single chemically active molecule) so that

$$\mathbf{r}^{\star} = 2\sigma/\mathrm{n}_{\mathrm{L}}\mathrm{k}\mathrm{T}\mathrm{ln}\mathrm{S} \tag{4-6}$$

$$\Delta G_{\varepsilon}^{\star} = \frac{16\pi\sigma^3}{3(n_{\rm L}kT1nS)^2} - \varepsilon \qquad (4-7)$$

and

The free energy of a cluster as a function of radius is shown in Fig. 35 for both the case of the homogeneous and the heterogeneous clusters.


A small stable cluster of radius r_c exists at saturation which serves to give the heterogeneous clusters a slight head start in the fluctuation process. The author is fully aware of the shortcomings of the classical liquid drop theory and in particular the difficulties which arise when the theory is applied to very small clusters.

The gas is assumed to obey a Boltzmann type distribution law. It is therefore assumed that the probability of occurrence of a process is determined by the energy required to establish the process, i.e.,

$$P = \exp(-\Delta G/kT) \tag{4-8}$$

where P is the probability of occurrence of a cluster whose free energy of formation is ΔG , k is Boltzmann's constant and T is the absolute temperature. If N_0 is the density of monomer water molecules in the gas and N_{ε} is the density of the heterogeneous nucleating centers, the expected density of clusters of size g of both the homogeneous and the heterogeneous types, $N_{\rm gh}$ and $N_{\rm gc}$, is

$$N_{gh} = N_0 exp(-\Delta G_h/kT)$$
(4-9)

$$N_{g\epsilon} = N_{\epsilon} exp(-\Delta G_{\epsilon}/kT)$$
 (4-10)

Fig. 36 shows how the number, N_{gh}, of clusters of size g varies with g. The minimum of the curve is the critical cluster size. The distribution is assumed to cut off at a value of g slightly larger than g* (say about 2g*, the exact value is not critical) so that an infinite supply of vapor is not required to maintain the distribution. It is assumed that each cluster which becomes larger than g* by the aquisition of another vapor molecule becomes a free growing droplet and will continue to grow to macroscopic size. In the classical theory, which is a precatastrophic



Fig. 36. Distribution of clusters.

theory, these droplets are assumed to be broken up by a Maxwell demon and returned to the vapor as monomers so that a steady state is maintained. The nucleation rate is the number of droplets growing larger than the critical size per unit time. Therefore, the nucleation rate is the product of the number of critical size clusters and the probability of the critical cluster acquiring another molecule. Hence

$$J_{h} = \frac{p \ 4\pi r^{*2}}{(2\pi m kT)^{1}/2} N_{0} exp - \left(\frac{16\pi\sigma^{3}}{3kT(n_{L}kT1nS)^{2}}\right)$$
(4-11)

$$J_{\varepsilon} = \frac{p 4\pi r^{\star 2}}{(2\pi m kT)^{1/2}} N_{\varepsilon} \exp \left(\frac{16\pi\sigma^{3}}{3kT(n_{L}kT\ln S)^{2}} - \frac{\varepsilon}{\kappa\tau}\right) \qquad (4-12)$$

where p is the vapor pressure of the water and m is the mass of a water molecule.

It is assumed that the number density of monomer water molecules, N_o , is so large that its value does not change sensibly during the nucleating process. N_c is much smaller, however, and the supply of these heterogeneous nucleating agents is quickly depleted. Therefore, a situation analogous to that of radioactive decay in nuclear physics exists.

$$-\frac{dN_{\varepsilon}}{dt} = P_{\varepsilon}N_{\varepsilon} \qquad (4-13)$$

where $P_{\epsilon} = J_{\epsilon}/N_{\epsilon}$.

This equation is integrated from time t=0 to time t=t and from the initial density $N_{\epsilon 0}$ to instantaneous density N_{ϵ} .

$$\int_{N_{\varepsilon 0}}^{N_{\varepsilon}} \frac{dN_{\varepsilon}}{N_{\varepsilon}} = -\int_{0}^{t} P_{\varepsilon} dt \qquad (4-14)$$

$$N_{\varepsilon} = N_{\varepsilon 0} \exp((p_{\varepsilon} t))$$
(4-15)

The total nucleation rate is the sum of the homogeneous and hetergeneous nucleation rates

$$J = J_n + J_{\varepsilon}$$
(4-16)

$$= \frac{p4\pi r^{*2}}{(2\pi m kT)^{1/2}} \left(\exp \left(\frac{16\pi\sigma^{3}}{3kT [n_{L}kT \ln S]^{2}} \right) \left(N_{0} + N_{\varepsilon 0} \exp \left(-P_{\varepsilon} t + \frac{\varepsilon}{kT} \right) \right) \right)$$

The second term in the right hand bracket is the heterogeneous contribution to the nucleation rate. Fig. 37 shows how the nucleation rate varies with supersaturation. The dotted line is the path taken by the cloud chamber during an expansion. Depletion of the heterogeneous nucleating centers brings the rate down to that of the purely homogeneous level in a short time.

If the classical liquid drop model is assumed to be valid, one can surmise several things about the free energies and cluster sizes from the data. Putting the free energy into the form of Eq. (4-3) instead oftthat of Eq. (4-4) brings out the dependence of the critical cluster size and surface energy terms more clearly.

$$J = \frac{p 4\pi r^{*2}}{(2\pi m kT)^{1/2}} \left(S^{\left(\frac{4}{3}\pi r^{*3}n_{L}\right)} \exp - 4\pi r^{2}\sigma/kT \right) \left(N_{e} + N_{eo} \exp\left(-P_{e}t + \frac{\varepsilon}{kT}\right) \right)$$
(4-18)

It is seen that the slope of the curve on a plot of lnJ vs. S for constant temperature will give the number of molecules in the critical cluster, $4/3\pi r^{*3}n_L$. This would indicate that the critical size may only be 35 and not the 80 predicted by the Kelvin-Thompson equation. This is not surprising since it has been shown⁷⁸ that the surface tension of small drops should be lower than that of the bulk liquid. If this is the case experiments, such as those using nozzles for which it is predicted



Supersaturation

Fig. 37. Comparison of homogeneous and heterogeneous nucleation rates. The solid curve is the total nucleation rate observed in the cloud chamber.

critical cluster sizes are of less than ten molecules, must be reconsidered.

It is also seen that the slope of the plot of InS vs. 1/T gives the surface energy per molecule in the critical cluster which differs in the two cases. The slope of the curves for the homogeneous and heterogeneous nucleation must be different because the heterogeneous slope has included in it an extra $\epsilon/(4/3\pi r^{*3}n_L)$ term. Moreover, the theory of this chapter predicts a smaller slope for the case of heterogeneous nucleation which is in agreement with experimental data. There is also the additional effect due to the initial vapor pressure of the water vapor which is another temperature dependence which must be included when intercomparing data of different starting temperatures.

CHAPTER V

DROPLET GROWTH

In order to assess the effects of droplet growth upon the nucleating vapor, a detailed calculation is required which accounts for vapor depletion, release of latent heat and the effect of the nonuniform vapor and temperature profiles around the growing drop. An exact calculation is impossible without resorting to the theory of nonuniform gases. Computational complexity of such considerations are discouraging. It is apparent that a simpler version of the theory will be quite adequate for the purposes of this work provided that the depletion of vapor and the evolution of latent heat never become a dominating effect.

The problem of the diffusional growth of liquid droplets from the vapor has been considered by numerous investigators. Frisch and Collins⁷⁹ extended the range of validity down to droplets whose diameter was of the order of magnitude of the mean free path of the gas molecules. They relegated the entire thermal diffusion process to a simple accommodation coefficient which would appear to lose the complex interdependencies of the thermal diffusion process on other physical parameters.

Bagge, Becker and Bekow⁸⁰ formulated a solution for the mass flux which connects continuously to the surface of the drop via a connection with kinetic theory. However, they do not follow similar considerations in the case of thermal transport. Mason⁸¹ takes into account kinetic interactions with the surface by deriving a modified diffusion coefficient. Although it seems to be implicitly implied, he makes no mention of the concept of either a temperature or vapor jump at the surface of the drop. Beucher⁸² develops a purely kinetic treatment of droplet growth which is valid down to the size of the critical nucleus defined by nucleation theory. He finds that the heat capacity of the droplet accounts for about half as much power as the creation of new surface, both effects dying out simultaneously at about twice the critical radius.

Neiburger and Chien⁸³ calculated droplet growth assuming macroscopic diffusion processes. They accounted for curvature, hygroscopicity and heating caused by the release of latent heat. They, however, neglected the effective modification of the diffusion coefficients at the droplet surface and, hence, did not account for a temperature or vapor density "jump" at the droplet surface.

Schuster⁷⁵ calculated droplet growth taking into account the effects of dcuble diffusion of heat and vapor. He assumes, for simplicity, that the shape of the diffusion profile outside the droplet may be described by the function $1 - \exp(-R_0/r)$ where R_0 is a parameter of the order of the mean free path. This function is used to calculate the vapor density at a point just outside the surface of the droplet. He used kinetic theory to calculate the actual rate of growth of the droplet. His solution is iterated through time in much the same manner as the author's. This technique in conjunction with a series approximation for the above mentioned exponential allows the problem to be solved in closed form.

Carstens⁸⁴ intercompares the predictions of the steady state and the non-steady state versions of the diffusion droplet growth theory. It was found that a cellular approach to the quasi-steady state theory very closely approximates the nonsteady state theory. Reiss and La Mer⁸⁵ demonstrated the effectiveness of the cellular model but only considered

vapor diffusion. More recently the cellular model has been used by Zung⁸⁶ to describe the evaporation of clouds and sprays. Smith⁵³ described the evaporation of very small water droplets utilizing a kinetic connection at the droplet surface. However, he did not use a cellular model.

Carstens and Kassner⁸⁷ discuss in some detail various aspects of droplet growth theory as they apply to cloud chamber measurements. The droplet growth theory utilized in this work is largely based upon their conclusions.

5-1. Droplet growth equations. Most cloud chamber experiments are arranged such that droplet growth occurs in a medium composed of a vapor in dilute solution with an inert gas. Vapor diffusion and thermal diffusion are both controlled by the nature of the inert gas. In the region where droplet radii are much larger than the mean free path of the gas molecules, the quasi-steady state diffusion equations constitute an adequate description of droplet growth.

$$\nabla^2 \rho = K_o(t) \tag{5-1}$$

$$\nabla^2 \mathbf{T} = \mathbf{K}_{\mathrm{T}}(\mathbf{t}) \tag{5-2}$$

where ρ and T are the vapor density and temperature and $K_{\rho}(t)$ and $K_{T}(t)$ are homogeneous source functions which account for changes in the bulk vapor and temperature of the gaseous system brought about by external means.

Power balance is required at the droplet surface. The latent heat liberated by the condensing flux of vapor molecules must be carried away by the process of thermal diffusion which is accomplished principally by the noncondensible gas under our particular conditions. It is assumed

that the droplet maintains a uniform temperature throughout its interior and that the steady state temperature of the droplet is always maintained.

$$\left. DL \frac{d\rho}{dr} \right|_{r=a} = \kappa \frac{dT}{dr} \right|_{r=a}$$
(5-3)

where D and κ are the mass and thermal diffusion coefficients respectively, L is the latent heat of condensation and a is the radius of the drop. The contribution of surface free energy and the thermal capacity of the drop are negligible throughout the region of interest in this work.

The specification of the outer boundary condition requires some physical insight. The principal defect in the steady state theory lies in the fact that the diffusion profiles prematurely extend to infinity. Since we expect to be able to calculate the diffusion profiles outside the droplet for the purpose of calculating the dead space, it is desirable to employ the cellular model. The imposition of an impermeable sphere of radius R which serves as the reservoir for heat and vapor for a droplet, eliminates to a large extent the errors in the diffusion profiles introduced through the use of the quasi-steady state equations.

For a truly isolated drop, R should be chosen so that the integral of the vapor depletion throughout the vapor density profile gives the mass of the drop. Under such a constraint $\rho(\mathbf{R})$ would remain constant throughout droplet growth and R would be moved out indefinitely as growth proceeds. Similar arguments apply to the thermal diffusion process, in general R(T) and R(ρ) being different. However, where a system of droplets exists the period of such isolation is short lived and the R spheres for different droplets begin to overlap. At this point a type of averaged competition for the available vapor becomes active. Strictly speaking, a distribution of R's should be employed such that the $\frac{d\rho}{dT}\Big|_{\mathbf{n}}$ are the same for all droplets. However, Kassner and Carstens⁸⁸ have shown that the distribution of R's does not appreciably affect droplet growth rates until droplet densities exceed 10^6 drops/cm³. Therefore, it is feasible to use the same size impermeable sphere for all droplets, requiring only that the volume of these spheres fill all space. Fig. 38 illustrates the mechanics of choosing the radius of the impermeable sphere.

When the drop size is of the order of the mean free path, λ , its growth is dependent upon kinetic theory. A connection may be established between the macroscopic and microscopic regimes by equating the corresponding flux expressions:

$$\left. D \frac{do}{dr} \right|_{r=\alpha} = \left[I_{v} - I_{c} \right] \delta$$
(5-4)

$$\kappa \frac{dT}{dr} \bigg|_{r=a} = \left[I_g C_g \delta_g + I_\rho C_\rho \delta_\rho \right] \Delta T$$
 (5-5)

where ΔT = the temperature jump between drop and vapor at the liquidvapor interface, C_v = the specific heat per molecule of the vapor, C_g = the specific heat per molecule of the non-condensible gas, δ_g = the average proportion of energy transfer between the non-condensible gas and the liquid surface molecules, δ_v = the average proportion of energy transfer between the vapor and the liquid surface molecules, δ = the sticking probability of the vapor molecules on the liquid surface. I_g = the flux of non-condensible gas molecules impinging on the drop surface, I_c = the flux of vapor molecules condensing on the surface of the drop as if it were in an equilibrium atmosphere, I_ρ = the flux of vapor molecules impinging on the drop surface.



Fig. 38. Choosing the radius of the impermeable sphere. When droplets do not compete for vapor, the radius R of the impermeable sphere is chosen so that there is no vapor depletion at R. Fig. a. When droplets are closely spaced this is not possible, Fig. b. A practical solution chooses R, not so that $\frac{\partial P}{\partial t}|_{R} = 0$, but so that $\frac{\partial P}{\partial t}|_{R} = 0$ and bulk corrections are made for vapor depletion (the error in the correction is the shaded area in Fig. c).

The existence of a temperature jump at the surface of the drop implies that the droplet is unable to maintain the equilibrium vapor pressure immediately outside its surface. Both a temperature jump and a vapor density jump exist.

$$\Delta T = T^{eq}(a) - T(a)$$
 (5-5)

$$\Delta \rho = \rho^{eq}(a) - \rho(a) \tag{5-6}$$

It is assumed that $I_{\rho} C_{\rho} \Delta T$ may be neglected because of the dilutness of the vapor. Then Eq. (5-4) and (5-5) become:

$$\frac{d\rho}{dr}\Big|_{r=\alpha} = 1/4 \{-\overline{v}_{V}[T^{eq}(\alpha)]\rho^{eq}(\alpha) + \overline{v}_{V}[T(\alpha)]\rho(\alpha)\}\delta \quad (5-7)$$

$$\frac{dT}{dr}\Big|_{r=\alpha} = 1/4 \rho_{g}\overline{v}_{g}[T(\alpha)]\Delta TC_{g}\delta_{g} \quad (5-8)$$

where $\overline{v_g}(T)$ = the average molecular velocity of the non-condensible gas at the temperature T, and $\overline{v_V}(T)$ = the average molecular velocity of the vapor at temperature T.

The solutions of Eq. (5-1) and (5-2) in terms of the boundary values are for the steady state:

$$\rho(\mathbf{r}) = \frac{\mathbf{R} - \mathbf{r}}{\mathbf{R} - \alpha} \frac{\alpha}{\mathbf{r}} [\rho(\alpha) - \rho(\mathbf{R})] + \rho(\mathbf{R})$$
(5-9)

$$T(r) = \frac{R-r}{R-a} \frac{a}{r} [T(a) - T(R)] + T(R)$$
 (5-10)

These solutions substituted into the power balance equation give:

$$\frac{\rho(R) - \rho(\alpha)}{T(\alpha) - T(R)} = \frac{\kappa}{LD}$$
(5-11)

The steady state solutions also give for Eqs. (5-7) and (5-8):

$$\rho(a) = \frac{\frac{R}{R-a} \frac{4D}{a} \rho(R) + \overline{v}_{v} [T^{eq}(a)] \rho^{eq}(a) \delta}{\frac{R}{R-a} \frac{4D}{a} + \overline{v}_{v} [T(a)] [1 + \frac{\Delta T}{T^{eq}(a)}]^{1/2} \delta}$$
(5-12)

$$\Gamma(\alpha) = \frac{\frac{R}{R-\alpha} \frac{4D}{\alpha} T(R) + \overline{v}_{g} [T^{cq}(\alpha)] \delta_{g}}{\frac{R}{R-\alpha} \frac{4D}{\alpha} + \overline{v}_{g} [T(\alpha)] [1 + \frac{\Delta T}{T^{eq}(\alpha)}]^{1/2} \delta_{g}}$$
(5-13)

To obtain the asymtotic behavior of $\rho(a)$ and T(a)

let

$$K = \gamma' \pi_g \overline{v}_g \lambda_g$$

 $D = \gamma \overline{\gamma} \lambda$

where γ and γ' are constants of the order unity, λ_v and λ_g are the mean free paths of the vapor and non-condensible gas. Eqs. (5-12) and (5-13) reduce to

$$\rho(a) = \frac{\frac{R}{R-a} 4\gamma(\lambda_{v}/a)\rho(R) + \rho^{eq}(a)\delta}{\frac{R}{R-a} 4\gamma(\lambda_{v}/a) + \left[1 + \frac{\Delta T}{T^{eq}(a)}\right]^{1/2}\delta}$$
(5-14)

$$T(\alpha) = \frac{\frac{R}{R-\alpha} \left(\frac{8}{5}\gamma'\right) \frac{\lambda g}{\alpha} T(R) + T^{eq}(\alpha) \delta_{g}}{\frac{R}{R-\alpha} \frac{\lambda g}{\alpha} \left(\frac{8}{5}\gamma'\right) + \left[1 + \frac{\Delta T}{T^{eq}(\alpha)}\right]^{1/2} \delta_{g}}$$
(5-15)

These equations are simplified by the approximation $\left[1 + \frac{\Delta T}{Teq}\right]^{1/2} = 1$ and substitution of n and n' for the remaining variables and constants gives

$$\rho(a) = \frac{n \ \rho(R) + \rho^{eq}(a)}{1 + n}$$
(5-16)

$$T(a) = \frac{n'T(R) + T^{eq}(a)}{1 + n'}$$
 (5-17)

In the limit that the mean free path is much smaller than the drop-

let radius:

$$\lim_{\alpha/\lambda_g \to \infty} T(\alpha) = Teq(R)$$

$$\lim_{\alpha/\lambda_g \to \infty} \rho(\alpha) = \rho(R)$$

These imply the absence of local vapor depletion.

Another relationship is provided by the Kelvin-Thompson equation which takes into account the change in equilibrium vapor density with drop radius.

$$a = \frac{2\sigma}{\rho R T} / \ln \frac{\rho(a)}{\rho eq(R)}$$

 $\frac{\rho(a)}{\rho^{eq}(R)} = \exp\left(\frac{2\sigma}{a\rho RT}\right) = \exp\left(\frac{B}{aT}\right)$

 \mathbf{or}

where σ = the surface tension, ρ = the density of the condensed vapor, R = the gas constant.

This equation when combined with the Clausius-Claperyon equation (integrated for an ideal gas) gives:

$$\rho^{eq}(a) = \frac{\rho^{eq}(R)T(R)}{T^{eq}(a)} \exp\left(a\frac{T^{eq}(a)-T(R)}{T^{eq}(a)T(R)}\right) \exp\left(\frac{B}{aT^{eq}(a)}\right) (5-18)$$

where $a_0 = QM/R$, Q = the latent heat of condensation, M = the molecule weight of the vapor.

There is also the equation relating mass influx to droplet growth.

$$\frac{d}{dt} \left(\rho \frac{4}{3} \pi a^{3} \right) = 4 \pi a^{2} D \nabla \rho \Big|_{r=a}$$

$$a \frac{da}{dt} = \frac{RD}{R-a} \left[\rho(R) - \rho(a) \right]$$
(5-19)

or

The droplet growth process has now been defined in terms of the variables a, $\rho(a)$, $\rho^{eq}(a)$, T(a), and $\overline{T^{eq}}(a)$ by the Eqs. (5-11), (5-16), (5-17), (5-18), and (5-19).

5-2. Solution of the droplet growth equations. A solution of the droplet growth equations may be obtained as follows. The power balance Eq. (5-11) is combined with the first continuity Eq. (5-16) to eliminate $\rho(a)$.

$$\rho(\alpha) = \frac{n\rho(R) + \rho^{eq}(\alpha)}{n+1} = \rho(R) + \frac{k}{LD}[T(R) - T(\alpha)]$$
 (5-20)

Now combine Eq. (5-17) with the above to eliminate T(a).

$$T(\alpha) = \frac{LD}{k(n+1)} \rho(R) - \frac{LD}{k(n+1)} \rho^{eq}(\alpha) + T(R) = \frac{n'T(R) + T^{eq}(\alpha)}{n'+1}$$

$$\rho(\alpha)^{eq} = \frac{k}{LD} \frac{n+1}{n'+1} (T(R) - T^{eq}(\alpha)) + \rho(R) \qquad (5-21)$$

$$= \frac{\rho^{eq}(R)T(R)}{T^{eq}(\alpha)} \exp\left(\alpha_{0}\frac{T^{eq}(\alpha) - T(R)}{T^{eq}(\alpha)T(R)}\right) \exp\left(\frac{B}{\alpha T^{eq}(\alpha)}\right)$$

$$T^{eq}(\alpha) \left[\rho(R) + \frac{k}{LD} \left(\frac{n+1}{n'+1}\right)T(R)\right] - \frac{k}{LD} \left(\frac{n+1}{n'+1}\right)T^{eq}(\alpha)^{2} - \rho^{eq}(R)T(R)$$

$$\cdot \exp\left(\frac{a_{0}}{T(R)}\right) \exp\left(\frac{B}{\alpha T^{eq}(\alpha)}\right) = 0 \qquad (5-22)$$

Eq. (5-22) gives the relationship between vapor density and temperature at the droplet surface.

The integrated form of Eq. (5-19) is

$$t_1 - t_2 = \int_{a_1}^{a_2} \frac{\mathbf{R} - a}{\mathbf{R} \mathbf{D}} \frac{\mathbf{r} d\mathbf{r}}{\left[\rho(\mathbf{R}) - \rho(\mathbf{r})\right]}$$
(5-23)

Knowing the droplet size at time t_1 , the droplet size at time t_2 is determined as follows. A trial value of radius a_2 is picked and a gaussian-quadrature method of integration is used to evaluate t_2 . If the calculated value of t_2 is not sufficiently close to the real value of t_2 , a new trial value of a_2 , picked by a bisection scheme, is used as the new upper limit of the integration. In the integration, values of $\rho(r)$ are determined from the steady state solutions in conjunction with the connection equations and Eq. (5-22). This process is easily carried out with an electronic computer.

Analysis of an actual data expansion from a cloud chamber is complicated by the fact that the narrowest possible pulse of supersaturation is so long that many different sizes of droplets are growing simultaneously. The procedure used to simplify this situation is to divide the supersaturation pulse into many small time increments. Each time increment is then assumed to have a constant supersaturation which is the actual value of supersaturation in the middle of the time step. Droplets are nucleated all during the time step but are assumed to all be born at the center of the time step so that they all have the same age and size.

5-3. Dead space calculation. A new radius of the impermeable sphere must be calculated after each time increment. This is because the population of droplets increases during each time increment so that a smaller volume is available for the impermeable sphere after each new family of droplets is born.

An approximate family population, AN_i , for the <u>ith</u> time interval is computed for the new family each time by using the bulk values of supersaturation and temperature. The exact population of the new family is computed by integrating the nucleation rate law over the volume of the impermeable spheres for all the preexisting families. Hence, the number in the ith family, N_i , is

$$N_{i} = \sum_{j=1}^{i-1} N_{j} \int_{\alpha_{j}}^{R} Rate \ 4\pi r^{2} dr \Delta t$$
 (5-24)

where Δt = the duration of the time step. Knowing N_i, the total droplet population is calculated

$$N_{T} = \sum_{j=1}^{i} N_{j}$$
 (5-25)

The radius of the impermeable spheres is then calculated for the (i+1)th time step.

$$N_{\rm T} \frac{4}{3} \pi R^3 = 1 \tag{5-26}$$

A dead space, V_D , is defined to be the volume around the droplets which would have no nucleation if the bulk values are used to calculate the nucleation rate and the total drops nucleated in a step is to be the value calculated using the exact profile, see Eq. (5-24).

$$V_{\rm D} = 1 - \frac{N_{\rm i}}{\Lambda N_{\rm i}}$$
(5-27)

The average radius of the dead space, r_D , is then defined by the relation

4/3
$$\pi r_D^3 = V_D 4/3 \pi R^3$$

 $r_D = V_D^{1/3} R$ (5-28)

The concept of dead space is useful as it is an indication of the extent to which the nonuniformity in the vapor and temperature distributions outside the droplets affects the whole volume. As long as the dead space is small, there is little doubt as to the validity of the droplet growth calculations. When dead space rises to a significant percentage of the total volume, the assumptions made concerning the impermeable spheres must be questioned. Moreover, the dead space is used to correct nucleation rates since no new nucleation occurs within the dead space.

5-4. The computer solution. A computer with moderate storage capacity is required for data analysis using the technique outlined in the preceding sections. A new family of droplets is born each time step and information such as radius, surface temperature, gas temperature at the surface, vapor density at the surface and dead volume must be kept for each family. The actual computer program used in this work follows the procedure outlined above. Numerical integrations and solutions are carried out with sufficient accuracy that errors from this are negligible. Because of the increasing number of families which must be accounted for with each additional time step, computer time rises as the square of the number of time steps in the calculation. Figs. 39-45 are sample plots of values calculated by the computer for one data expansion. A sample computer printout is given in Appendix III.



Fig. 39. Pressure vs. time.

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Fig. 40. Temperature vs. time for the expansion of Fig. 39.



Fig. 41. Supersaturation vs. time for the expansion of Fig. 39.



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Fig. 42. Radius vs. time for the expansion of Fig. 39.



Fig. 43. Number vs. radius for the expansion of Fig. 39.



Fig. 44. Vapor pressure vs. time for the expansion of Fig. 39.



TIME (sec)

Fig. 45. Dead space for the expansion of Fig. 39.

CHAPTER VI

SUMMARY

The expansion cloud chamber has been developed into a precision instrument, capable of yielding definitive information concerning nucleation and growth of water droplets.

The gas temperature was measured in the dry chamber during the course of an isentropic expansion. It was found that the finite heat capacity is not negligible and must be accounted for. A computer program was developed which solves the problem of heat flow from the thermocouple. When the measured gas temperature is compared with that calculated from the pressure measurement using Eq. (3-1) it is found that there is almost perfect agreement. With some refinement of technique, the expansion chamber, using the same method of measuring temperature and pressure as in this work, could be used to accurately calculate the heat capacity of the gas through the relationship:

 $C_{p} = T\left(\frac{\partial V}{\partial T}\right)_{p}\left(\frac{\partial P}{\partial T}\right)_{s}$

This method of calculation of the heat capacity is potentially more accurate than any now available.

The nucleation rate was measured as a function of temperature, supersaturation and sensitive time for water vapor in a helium atmosphere. It was found that there exists a form of heterogeneous nucleation occurring above the ion limit at about the supersaturation predicted by the Becker-Doring theory for homogeneous nucleation. This form of heterogeneous nucleation appears to occur upon chemically bonded centers whose concentration is very low and depends upon the vapor pressure prior to the expansion. The consistency of the number of these nucleating centers indicates that they may be the neutral product of natural radioactivity and cosmic rays. Experiments are planned to test this hypothesis by dosing the cloud chamber with x-rays and checking to see if the number of nucleating centers is increased.

A semiphenomenological theory was developed along the lines of the classical theory but which includes the chemical bond energy of the heterogeneous nucleating center. The theory predicts a different temperature dependence for the heterogeneous and homogeneous nucleation rates and at least qualitatively explains the essential features of the data. Modifications in the photographic technique are under way which will allow an extension of the data to higher and lower droplet densities. If the liquid drop theory satisfactorily describes the nucleation of water vapor, then: (a) the measured temperature dependence of the nucleation rate will yeild the free energy per molecule in the critical embryo and (b) the supersaturation dependence will yeild the number of molecules in the critical cluster.

A considerable disparity has existed in the temperature dependence of the critical supersaturation as reported by various experimenters. This disparity has been in large part resolved by properly interpreting the data in terms of the theory derived for the heterogeneous nucleation.

It was definitely established that the nucleation rate of water vapor is higher in an argon atmosphere than in a helium atmosphere. This may be related to a disruptive factor due to the higher velocity

of the helium atoms. It is however more likely due to the hydration of the argon atoms into the critical cluster. Such a hydration has been hypothized for krypton and xenon in liquid water. These two gases have been obtained so that the same nucleation experiment may be performed in an atmosphere of each of these. If such a hydration is involved the nucleation rate will be considerably higher in these gases.

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APPENDIX I

VARIATION OF PHYSICAL PARAMETERS

Because of the wide variation in the thermodynamic coordinates during the course of these experiments several of the physical parameters which are normally considered to be constant must be included as variables in the calculations in order that sufficient accuracy may be obtained. The variation of some of these parameters are discussed in the following paragraphs.

I-1. Equilibrium vapor pressure of water. A least squares fit was used for the vapor pressure of water⁸⁹, see Fig. 46.

Below 25°C

 $p = 4.58192 + .333075T + .010758T^{2} + .196622 \times 10^{-3}T^{3}$ $+ .216663 \times 10^{-5}T^{4} + .211191 \times 10^{-7}T^{5}$

Between 20°C and 60°C $p = 5.92556 + .139239T + .0215602T^2 - .94144 \times 10^{-4}T^3$ $+ .59993 \times 10^{-5}T^4$

where p is the vapor pressure in mmllg and T is the temperature in degrees Centigrade.

<u>I-2.</u> Surface tension of water. Data are available for only the temperature dependence of the surface tension of water.⁸⁹ A linear approximation is satisfactory for the range of interest in this work, see Fig. 47.

 $\sigma = 116.459 - .149228T$

where σ is the surface tension in dynes per centimeter and T is the absolute temperature in degrees Kelvin.



Fig. 46. Equilibrium vapor pressure of water.


Fig. 47. Surface tension of water.

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I-3. Latent heat of vaporization of water. A linear least squares fit was used for the latent heat of vaporization of water⁸⁹, see Fig. 48.

$$Q = 746.1 - .55T$$

where Q is the latent heat in calories per gram and T is the absolute temperature in degrees Kelvin.

<u>I-4</u>. <u>Heat capacity and compressibility of water vapor</u>. The best available compilation of water vapor entropy data is that of Vukalovitch.⁹⁰ The interpolated values of Cp, where Cp is the constant pressure heat capacity divided by the ideal gas constant, are given in Table II. Figs. 49 and 50 show plots of this data. There is a definite discontinuity around 340°K and 23.8 mmHg. Because of this the best curve fit is divided into three expressions.

For any pressure above 340°K

Cp = 3.1611

Below 23.8 mmHg and below 340°K

 $C_p = 2.9611 + .01201(T-240.)$

Above 23.8 mmHg and below 340°K

$$Cp = 2.9611 + .01234(T-240.) + \left(\frac{28.2 - p}{14.71}\right) [.1958 + .00082(T-240.)]$$



Fig. 48. Latent heat of vaporization of water.

TABLE	II.	The	heat	capacity	of	water	vapor.

-

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p mmHg T°K	7.3559	14.71118	22.0667	29.42236	36.778
240	3.1790	3.0919	2.9832	2.9611	2.9611
250	3.3115	3.2207	3.1075	3.0845	3.0845
260	3.4440	3.3496	3.2318	3.2079	3.20 79
270	3.5764	3.4784	3.3561	3.3313	3.3313
280	3.7089	3.6072	3.4804	3.4546	3.4546
290	3.8414	3.7361	3.6047	3.5780	3.5780
300	3.9738	3.8649	3.7290	3.7014	3.7014
310	4.1063	3,9937	3.8533	3.8248	3.8248
320	4.2388	4.0646	3.9776	3.9482	3.9482
330	4.3712	4.1019	4.1019	4.0418	4.0418
340	4.5037	4.1337	4.1953	4.1395	4.1395
350	4.1281	4.1279	4.1598	4.1599	4.1279
360	4.3767	4.1479	4.1807	4.1807	4.147 9
370	4.0936	4.1288	4.0952	4.0955	4.1288





and the second sec

Fig. 50. Heat capacity of water vapor.

t'S't

The compressibility, $z = (\partial V/\partial T)_p$, is plotted in Fig. 51. There is a negligible pressure dependence.

Below 235°K

$$z = 1.005 + .00054(T-325.)$$

Above 235°K

z = 1.005

<u>I-5</u>. <u>Thermal conductivity of Helium</u>. A linear relationship was used for the thermal conductivity of Helium as a function of temperature.^{91,92} The pressure variation is insignificant.

k = 4300 + 35T

where k is the thermal conductivity of helium in ergs per degree second centimeter and T is the absolute temperature in degrees Kelvin.

<u>I-6.</u> Heat capacity and compressibility of Helium, Argon, Nitrogen and <u>Air</u>. In making accurate calculations of temperature as discussed in Chapter 3 it is essential that the heat capacity and compressibility of the atmospheric gases be included as variables in the calculation. For that reason values of heat capacity, compressibility and the constant $(\gamma-1)/\gamma$ which is used in the isintropic ideal gas law, Eq. (3-1), have been tabulated for Argon, Nitrogen and air from data taken from NBS Circular #564.⁹³ Helium is so nearly ideal at the temperature and pressure used in this work that it may be considered ideal for all practical purposes.

The heat capacity is plotted in unitless numbers as Cp/R where R is the ideal gas constant. The compressibility is PV/RT + $T\left(\frac{\partial (PV/RT)}{\partial T}\right)_{p}$







Fig. 52. Thermal conductivity of helium.

where P is pressure, V is volume and T is the absolute temperature. The adiabatic constant is the compressibility divided by the heat capacity.

TABLE III. Compressibility, heat capacity and adiabatic constant of Argon.

n atm				
T°K	.7	1.0	1.5	2.0
245	1.00264	1.00391	1.005912	1.00804
295	1.00173	1.00256	1.00390	1.00526
345	1.00119	1.00180	1.00270	1.00360

T [°] K	.7	1.0	1.5	2.0
245	2.5065	2.5092	2.5139	2.5187
295	2.5042	2.5059	2.5089	2.5119
345	2.5029	2,5042	2.5063	2.5083

Adiabatic Constant

раст Т°К		1.0	1.5	2.0
245	.4000 2	.40009	.40014	.40025
295	.40002	.40008	.40013	.40020
345	.40001	.40005	.40007	.40011

.

TABLE IV. Compressibility, heat capacity and adiabatic constant of Nitrogen.

р Т°К	atm	.7	1.0	1.5	2.0
245		1.00257	1.00377	1.00578	1.00755
295		1.00161	1.00242	1.00349	1.00486
345		1.00109	1.00181	1.00255	1.00365
			Heat Capacity		
р Т°К	atm	.7	1.0	1.5	2.0
245		3.5072	3,5098	3.5141	3.5184
295		3.5066	3.5082	3.5110	3.5138
345		3.5098	3.5110	3.5129	3.5149
			Adiabatic Constan	t	
р Т°К	atm	.7	1.0	1.5	2.0
245		.28586	.28599	.28621	.28636
295		.28563	.28575	.28581	.28588

.28533

.28522

· .

345

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.28539

n atm	Co	ompressibility		
T [°] K	.7	1.0	1.5	_2.0
245	1.00246	1.00386	1.00579	1.00748
295	1.00182	1.00260	1.00390	1.00520
345	1.00103	1.00172	1.00241	1.00344
		Heat Capaci	ty	
T°K	.7	1.0	1.5	2.0
245	3,50015	3.50275	3.50712	3.51148
295	3.50365	3.5053	3.5081	3.5109
345	3.51345	3.5147	3.5163	3.5186
	4	Adiabatic Const	tant	
p atm T°K	.7	1.0	1.5	2.0
245	.2864	.2866	.2868	.286 9
295	.2859	.2860	.2862	.2863
345	.2849	.2850	.2851	, 285 2

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· .

TABLE	V.	Compressit	oility,	heat	capacity	and
		adiabatic	constar	nt of	Air.	

I-7. Diffusion coefficient for water vapor in helium. No experimental data were available for water vapor diffusing through helium. Therefore, a theoretical expression due to Jeans⁹⁴ was used. The resultant equation is approximately a linear function in the region of interest.

$$D = .000946T + .291$$

where D is the diffusion coefficient in cm^2/sec and T is the temperature in degrees Kelvin.

APPENDIX II

COMPUTER SOLUTION OF THE HEAT FLOW EQUATION FOR A CYLINDRICAL WIRE

The heat capacity of a thermocouple is very large in comparison to the heat capacity of a gas. Therefore, when the gas temperature is changing the thermocouple may not be at the same temperature as the gas. For this reason, an exact calculation of the heat flow from the thermocouple must be performed.

Due to the difficulty involved in incorporating a time dependent source term into an analytic solution of the heat flow equation, it was decided to seek only a numerical solution. Moreover, a different analytic solution is required for each set of boundary conditions, whereas the numerical solution, once it is properly programmed, will work for any set of boundary conditions.

In an iterative numerical solution, one assumes that the partial derivative may be approximated by finite differences.^{95,96} Intervals in space and time are picked small enough so that the temperature function may be assumed linear between them. Thus, the x-axis is divided into equal small increments with the points designated as T(1), $T(2)\cdots T(N)$. Stepping forward in time gives a function varying in time. Thus, T(n,1), $T(n,2)\cdots T(n,M)$, where one is looking at the time variation of the function T at the nth point along the x-axis.

The space derivative at the k-th point along the x-axis, T(k,r), may be taken in a forward or backward direction.

<

$$\frac{\partial T(k,t)}{\partial x} = \frac{T(k+1,t) - T(k,t)}{\Delta x}$$
(II-1)

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is the forward derivative where Δx is the x-distance between lattice points. Similarly,

$$\frac{\partial T(k,t)}{\partial x} = \frac{T(k,t) - T(k-1,t)}{\Delta x}$$
(II-2)

is the backward derivative.

•

Second derivatives are taken in a similar manner.

$$\frac{\partial^2 T}{\partial x} = \frac{\partial}{\partial x} \left(\frac{\partial T}{\partial x} \right) = \frac{\frac{T(k+1,t) - T(k,t)}{\Delta x} - \frac{T(k,t) - T(k-1),t}{\Delta x}}{\Delta x}$$
(II-3)

$$= \frac{T(k+1,t) - 2T(k,t) + T(k-1,t)}{(\Delta x)^2}$$

Before going to the full two dimensional equation, let us simplify by letting $\Delta x = h$, $\Delta y = h$, $\Delta t = k$ r = x coordinate, s = y coordinate, t = time coordinate. Eq. (3-12) becomes

$$\frac{T(r,s,t+1) - T(r,s,t)}{k} = \frac{K}{C_{p}h^{2}} [T(r+1,s,t)+T(r-1,s,t)+T(r,s-1,t) + T(r,s+1,t) - T(r,s,t)] + f(x,y,x,t) + T(r,s+1,t) - T(r,s,t)] + f(x,y,x,t)$$
(II-4)

With the equation cast into this form, it is seen that, if one knows the value of the function T at a point and the points immediately surrounding it at a time t, then the value of the function T may be calculated for a short time Δt in the future. This technique is used at all the points in the region of interest and repeated until the time of interest is reached. Extreme caution must be exercised in picking the h and k to be used in the approximations. Due to the iterative nature of the solution, when a time interval k (long enough for an equilibrium to be reached in the space interval h) is used, the approximation for short time in the differential equation is invalidated. If a large enough value of k is picked so that there is even a slight error in one time interval, the error tends to compile in successive time intervals. This is known as an exponential instability and the solution diverges exponentially from the true solution.

Fox⁹ has a good treatment of this problem. He shows that for $K/C_p = 1$

$$\frac{\Delta t}{(\Delta x)^2} \le \frac{5}{2}$$

this is about three times the relaxation time of a square in the lattice. Argon at room temperature has these possible values of h and k.

TABLE VI. Possible values of h and k for Argon

sec	.625×10 ⁻⁶	.25×10 ⁻⁵	1×10 ⁻⁵	4×10 ⁻⁵	16×10 ⁻⁵	6.4×10 ⁻⁴	2.56×10 ⁻³
cm	.000625	.00125	.0025	.005	.01	, 02	.04

Referring to the chart, a square the size of the thermocouple cross section requires time increments of the order of one microsecond. A problem arises when one considers that the mesh must extend outward to 0.3 cm from the thermocouple and the integration must take the time through a period of one second. In order that the problem could be solved with a finite size, finite speed computer, a grid mesh was set up with increasing mesh size as the points recede from the thermocouple, see Fig. 53.

$$A(5,1) A(5,2) A(5,3)$$

$$A(5,3) A(5,3) A(5,3) A(5,4)$$

$$A(3,1) A(3,2) A(3,3) A(4,4)$$

$$A(2,1) A(2,3) A(3,4) A(4,4)$$

$$A(1,1) A(2,4) A(3,5) A(4,5) A(5,5)$$

WIRE Aloo ()
$$A(2,5) A(3,5) A(4,5)$$

A(1,9) $A(2,6)$
A(2,9) $A(2,7) A(3,6)$
A(3,9) A(3,8) A(3,7) A(4,6)

A(4,9) . Λ(4,8) . Λ(4,7) . Λ(5,6) 5 El

· A(5,9) . A(5,8) · A(5,7)

Fig. 53. Mesh used for thermocouple heat flow calculation. The mesh extends outward to the 10th row. The spacing is: row 1-.000675cm, row 2-.000675cm, row 3-.00125cm, row 4-.0025cm.

This technique allows small space intervals near the thermocouple where it is necessary and large space-time intervals in the outlying area. The net result of this technique is that a medium speed, medium capacity computer is sufficient to solve a problem normally suited to a much larger machine. Accuracy is not sacrificed with this technique. Table VII gives a sample of the computer output. Fig. 54 shows the temperature profile around the thermocouple for the expansion shown in Figs. 24 and 25.



Fig. 54. Temperature profile around the thermocouple.

 $\vec{Z}_{ij}\vec{L}$

<u> </u>	PROGRAM EDR CALCULATING THERMAL LAG DE THERMOCOUPLE
С	READ IN INITIAL CONDITIONS
	<u>PEAD(1,100)P,T</u>
- 100	王(J)和昭A王(-2E18-8) N-0
	A = O A 1 CO = T
	<u>nn 90 [=1.10</u>
6.5	DA(1, J) = 1 DA(1, 1) = 0
	<u>30 8 [3=1,4</u>
	00.7.17=1.4
	01 5 16=1,4 DO 5 15=1.4
<u> </u>	K, CP (ARGONI DT=1E-5, DX=, 0025CM, CP1= HEAT CAPACITY OF RIRE
	CP = (.04033 + .000398*2) *P/T
	- YK=3。905日~05キ(キー273・15)※1・713日~07 - CR1~ 18708
	XKF=1.6
	C = (XK/CP) * XKH
	<u>CI</u> =(XK/(CP+CPI))≭XKH DO-(_I(CP+CPI))≭XKH
	7234 14-194 N=N+1
	$VEL = (-(N-1)*(54 \cdot E - 5)*(-215)+2 \cdot 03)*1 \cdot E - 5$
	VC(1)=1.0-(VEL/0.0025)
15	V(1 X) = 1 + 2 + 3(1 + V(1 X+1))
	$P_{2} = 3 + 3 = 1, 4$
	00 - 2 - 12 = 1, 4
C	HEAT FLOW CORRECTION AREA NUMBER ONE
	DA(1,1) = C * (A(2,1) + 2 * A(1,3) + A 100 - 4 * A(1,1))
	0A(1,3) = 0*(A(1,1) + A(2,2) + A(2,4) + A(1,5) - 4*A(1,5))
	$\frac{1}{2} \frac{1}{2} \frac{1}$
	DA(1,9) = C*(A100+2*A(1,7)+A(2,0)-4*A(1,9))
	A1CO=C1*(A(1,1)+2*A(1,5)+A(1,9)-4*A100)+A100
	$N_{1} = 1 + 9 + 2$ A(1 - 1) = DA(1 - 1) + A(1 - 1)
Ç	TEMPERATUPE CHANGE IN ENTIRE MATRIX DUE TO VOLUME CHANGE
	DT=097486*T*VEL
	1 = 1 + 01 0 = 0 + 0 + 2 (0.0.2.1 + 0.1 / T)
· · · · · · · · · · · · · · · · · · ·	A100=A100+CP*DTZCP1
	00.98 IL = 1,9
0.0	DI 98 JE = 1, 9 ACT = 0.1 - A(11, 11) + DT
	VC1 = VC(1)
	VC2 = 1 - VC1
·	$\Lambda(1,1) = VC1 * A(1,1) + VC2 * A(1) C$
	$A(1,3) = VC_1 + A(1,3) + VC_2 + A(1,3)$
	$\Lambda(1, 7) = \Lambda(1, 7) = \Lambda(1 + \sqrt{2} + \sqrt{2})$
	$A(1,9) = VC 1 \neq A(1,9) + VC 2 \neq A(2,9)$
1	CONTINUE CANTINUE (DA A-C-2-VC)
2	しんした。日本時にしていたすがすいすべすです。
	CALL DADEL (DA, A, C, 3, VC)
3	CONTINUE
,	
·+	$P_1 = P * 760$.

<u>II-1.</u> Computer program for calculation of the thermal lag of the thermocouple.

$\Delta[1] \left\{ = \lambda \left\{ 0 \right\} - \Delta \left\{ G_{+} \right\} \right\}$	
88 1 1 M - 1 20 1 20 1 20 1 20 1 20 1 20 1 20 1	
$\frac{16(3,1) \cdot A(2,1) \cdot A(1,1) \cdot A(0,A(1,9) \cdot A(2,9) \cdot A(3,9) \cdot A(6,9)}{16(3,1) \cdot A(2,1) \cdot A(1,1) \cdot A(1$	A(5.9).
$2\Lambda(6, 9), \Lambda(7, 9), \Lambda(8, 9)$	
CALL DADEL (DA,A,C,5,VC)	
UALE DADEL(DA)A, C, C, C, VOJ A CONTINIE	
$CALL DADEL(DA \cdot A \cdot C \cdot 7 \cdot VC)$	
7 CONTINUE	
CALL DADEL(DA,A,C,2,VC)	
$\frac{8}{101} \frac{100}{100}$	
101、2010のAIT/アビックサビノ・エサビビ・スサビビ・ゲサビビ・エオ 102、名句のMAT(17757、2)。	
CALLEXIT	
FND	
SUBPOLITINE DADEL (DA.A.C.I.VC)	
C TEMPERATURE COPRECTION DUE TO HEAT FLOW BEYOND CENTRAL	AREA
DOUBLE PRECISION A(10,9), DA(10,9), D1, T, P, 4190	•
DOUBLE PSECISION VC(0),VC1,VC2,VC3,VC4,VC0,VC0	
$\frac{1}{2} \sum_{i=1}^{n} \frac{1}{2} \sum_{i=1}^{n} \frac{1}$	2))
$1-3.5 \times \{\{1,2\}\}$	
$D_{A}(1,2) = C_{*}(A(1,2) + A(1,4) + .5*(A(1+1,4) + A(1+1,2)) - 3*A(1,4)$	
DA(1,4) = C*(A(1,3) + A(1,5) + A(1-1,3) + 25*(A(1+1,4) + A(1+1,4))	71
$\frac{1-3.5*(11,4)}{(1-3)^{5}}$.5))
$\frac{1}{1} + \frac{1}{2} + \frac{1}$	(6))
$= \frac{1}{0} \Lambda(1,7) = \frac{1}{2} \kappa(\Lambda(1,8) + \Lambda(1,6) + 0.5\pi(\Lambda(1+1,6) + \Lambda(1+1,7)) + 1}{1}$	
[0A(1,9)=0*(A(1,9)+A(1-1,7)+A(1,7)+0.25*(A(1+1,7)+0.15))]	y 2 i 1
1-3.5*A(1,8)	
$00 \ 10 \ 1A = 1.9$	
$10 \Lambda(I,JA) = A(I,JA) + OA(I,JA)$	
VC3=VC(I)	
VC4=1VC3	
$\frac{V(F=5) \times V(4)}{V(F=1) - V(C)}$	
$\Lambda(I, 1) = VC3 * \Lambda(I, 1) + VC4 * \Lambda(I-1, 1)$	
A(1,2) = VC3 * A(1,2) + VC4 * A(1-1,3)	
$\frac{\Lambda(1,2) = V(3*\Lambda(1,3) + V(4*\Lambda(1,4))}{V(4*\Lambda(1,4))}$	
ム()・4)=Vしつぞう()・4)=Vしサでパレノナフィ メノナードン=VCコッカノナ・5)+VC4×3(キ・6)	
(1, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (0, 5) = (
A(1,7) = VC5 * A(1,7) + VC6 * A(1+1,8)	
A(1,8) = VC5 * A(1,8) + VC6 * .5 * (A(1+1,8) + A(1+1,9))	
A(1,9)=VC5=A(1,9)+VC6=A(1+L,9)	
PETURN	

TABLE VII

. .

OUTPUT OF THERMOCOUPLE HEAT FLOW COMPUTER PROGRAM

Print out is in this order: Gas temperature, Pressure, A(9,1), Time, Temperature difference A(1,9) A(2,9) A(3,9) A(4,9) A(5,9) A(6,9) A(7,9) A(8,9)A(8,1) A(7,1) A(6,1) A(5,1) A(4,1) A(3,1) A(2,1) A(1,1)A1.00 289.74 1027.1 289.74 0.1587 1.41 289.74 289.99 290.15 290.28 290.41 290.55 290.69 290.82 291.14 290.78 290.61 290.42 290.21 290.01 289.85 289.76 289.74 289.61 1025.9 289.61 0.1613 1.41 289.51 289.86 290.02 290.15 290.29 290.42 290.56 290.69 291.02 290.65 290.48 290.29 290.09 289.88 289.72 289.63 289.61 289.48 1024.8 289.48 0.1638 1.41 289.48 289.73 239.89 290.03 290.16 290.30 290.43 290.57 290.89 290.52 290.35 290.16 289.96 289.75 289.59 289.50 289.48 289.35 1023.6 289.35 0.1664 1.41 289.57 289.56 289.77 289.90 290.03 290.17 290.31 290.44 290.75 290.40 290.22 290.04 289.83 289.63 289.45 289.37 289.35 289.22 1022.5 239.22 0.1690 1.42 289.54 289.53 289.54 289.54 289.77 289.91 290.04 200.18 290.31 290.64 290.27 290.10 289.91 289.70 289.50 289.33 289.25 289.22 289.09 1021.4 289.09 0.1715 1.42 289.41 289.41 289.51 289.65 289.78 289.92 290.05 290.19 290.51 290.14 289.97 289.78 289.58 289.37 289.20 289.12 289.09 288.96 1020.2 288.96 0.1741 1.42 289.28 289.28 289.38 289.52 289.52 289.56 289.79 239.93 290.05 290.38 290.02 289.84 289.66 289.45 289.24 289.07 288.99 288.96 289.83 1019.1 288.83 0.1766 1.42 289.15 289.15 239.27 289.39 289.53 289.66 289.80 289.93 290.25 289.89 289.72 289.53 289.32 289.12 288.95 288.86 288.84 288.70 1017.9 288.70 0.1792 1.43 287.02 280.02 289.14 290.27 289.40 289.54 289.67 289.81 290.13 289.76 289.59 289.40 289.20 288.99 288.82 288.73 288.71 283.52 1016.8 238.58 0.1818 1.43 288.90 288.89 289.01 289.14 289.23 239.41 239.55 289.63 290.07 239.64 289.46 289.28 289.07 288.86 288.69 288.60 288.58 288.45 1015.7 238.45 0.1843 1.43 288.77 288.76 288.88 289.02 289.15 239.28 289.42 289.56 289.88 289.51 289.34 289.15 298.94 288.73 298.56 288.47 238.45 288.32 1014.5 288.32 0.1969 1.43 288.64 228.63 238.77 288.89 289.02 239.16 289.29 289.43 239.75 239.38 289.21 289.02 288.82 288.61 288.44 288.34 288.32 283.19 1013.4 283.19 0.1894 1.44 -288.51 288.50 289.44 268.76 288.90 299.03 289.17 287.30 289.63 289.26 289.08 283.90 288.69 288.48 288.31 283.22 288.19 288.06 1012.3 288.06 0.1920 1.44 289.38 288.30 288.51 288.64 288.77 288.91 289.04 289.18 289.50 289.13 288.96 288.77 288.56 288.35 288.18 288.09 288.06 287.93 1011.2 287.93 0.1946 1.44 289.25 288.25 288.38 288.51 288.64 238.78 238.91 289.05 289.37 299.00 288.83 288.64 288.44 288.23 288.05 287.96 287.94 <u>297.81 1010.0 287.81 0.1971 1.44</u> 288.12 298.12 288.26 293.38 288.52 288.65 288.79 288.92 289.25 239.88 288.71 288.52 288.31 288.10 287.93 287.83 287.81 287.68 1002.9 287.68 0.1997 1.44 -283.00 287.00 238.13 268.26 289.30 288.53 298.66 283.80 299.12 298.75 288.58 288.39 288.15 287.97 287.80 287.70 287.68 297.55 1007.8 287.55 0.2022 1.45 287.87 287.86 238.01 238.13 238.27 288.40 288.54 288.67 288.99 293.62 288.45 288.26 288.06 287.85 287.67 287.57 287.55 287.42 1006.7 237.42 0.2048 1.45 287.74 287.74 287.85 288.01 288.14 288.77 288.41 288.54 288.87 238.50 288.33 288.14 287.93 287.72 287.55 287.45 287.42 <u>287.29 1005.6 297.29 0.2074 1.45</u> 287.61 287.67 237.76 237.38 283.01 233.15 238.28 238.42 288.74 238.37 288.20 288.01 287.80 287.59 287.42 287.33 287.30 287.17 1004.4 287.17 0.2099 1.45 -287.49 287.54 287.53 287.76 287.99 298.02 288.16 288.29 288.62 288.25 288.07 287.88 287.68 287.47 287.29 287.20 287.17

APPENDIX III

COMPUTER SOLUTION OF THE DROPLET GROWTH EQUATIONS

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ICCTK7
                                                                                                                                                                                              TEAM OSTAGE BASTE FORTRAN IV (F) CHAPTLATION
                                             TOUTS R. ALTEN IOPA NORWOOD HALL
SCHEDALTZOD MUCLEATION PATE PATE PROGRAM
                     \frac{1}{2}
                     \hat{\phantom{a}}
                                 ccáv ž
                                              PATE LAN = PAPLEM PATE LAW
PEAL K
                     2
                                                          DIMENSION AT(5(), A1(50), A2(50), CMT(50)
DIMENSION (T(17), AT(13)
                         NINENCION (T(1), ALCOLT, ALCOLT, ALCOLT
NENCION (T(1), AT(1))
NETF(2,1021)
NETF(2,1021)
NETF(2,1021)
NETC(511())
NETC(1,1020) (T(11), H=1, H(0), (%T(1)), J=1, H(0)
NETC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, 20, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, PI, 20AS, 20, 29, C(,CA'0), 200%, K
NEAC(1,1021) #TECS, ETEV2, 200%, PI, 200%, PI, 200%, PI, 200%, F
NEAC(1,1021) #TECS, ETEV2, 200%, F
NEAC(1,1021) #TECS, F
NEAC(1,1021) #TE
                                                           AVPC=VPO
                                                         CTIETIMEI
CMTCTEO.
ACMTCTEC.
                                      NUMPERADED BY A ARE INCORECTED VALUES TER VALUES
                                                        ")=>
ICl=IP-?
UCl=ICALC(PC,P1,T0,T1,VPC,VP1)
CALL TCALC(PC,P1,T0,T1,VPC,VP1)
CALL VDREC(T1,VPE0)
SUPP=VP1/VPE0
CALL VDREC(T1,VPE0)
ASUCR= AVP1/VPE0
PEAD (1,1(C1) P2,T1M52
PT2=T1ME2-T1M51
PT=(D1+D12)*.5
                          1.13
                         1 VAPON PRESSNEES DE 1,2010.47

C2 TO2=101-1

IF(IO2)205,205,07

G7 D1 C2 11=1,T02

GEAD (1,1001)P2,TIME2

WRITE(3,1020)

1023 FOPMAT(77,* THE DEMAINING DATA CARDS AFE THE FOLLOWING:1,7)

G3 WPITE(3,1020)P2,TIME2

1027 FOPMAT(F2C.2,F15.6)

WRITE(3,1030)
```

151

```
10 C D C C D C T ( / / / / )
200 T C D C D
                        ET CALL MAATE (ATT, AVEL, ASUDE, APATE)
                                               CHEAT
SOUTE(2,1000) T
1000 FOS AT( TIME INTERVAL MO.', [4,//)
FOTTE(7,1002) F1.5T
1000 F02 MAT(AY, FORESSUBE =: F1).3, MM HG!, 10Y, FOURATION OF THIS STEP
1 == .e10.6, SECGNOS',///)
15(FOTTOT-1, 100, 00.97
00 FOTT(1)=(1,-FONTOT)%S1(E*OT
SOUT(1)=(1,-FONTOT)%S1(E*OT
SOUT(1)=(1,-
        1015 FOT (1)
                                                                                                                                                     DRODS VICLEATED IN THE EREE VOLUME = = + F15.6,//)
                                                  1 <sup>v</sup> = 1
            111 00 00 J=1,14

• J207 00 00 J=1,14

• J207 00 00 J V KATIOM 05 A2(J)

I 0 (J-1) 20,21,20

21 A1(J)=1,-5-05

0 J= A1(J)

A2(J)=100A1(J)

20 J0 22
                                                  21 TO 22
                        20 FAJ=(A1(J)-A7(J))*A1(J)

20 FAJ=(A1(J)-A7(J))*A1(J)

20 FAJ=(A1(J)-A7(J))*A1(J)
                                                0 320
                                                ^{11} = -1
                                                े र जिल्ला में सुर
जिल्ला में सुर
                     A SUMEA
Happervittel
AD 1A TIF1, IIA
AD 1, AD 1
                                                  3 Y 2 6
                        17 OT=(07140T2)*.5
                        /* [ 1841 MIT
CMECK EDWOD, ADDLY BISECTION METHOD OF ITEPATION
IF(ADS(SUM-DT)-DT/SD.)10,10,1
1 TE(SUM-DT)2,10,4
2 TE(CM)3,2,7
£.
                                             3
                                                TE(CK)7,5,5
                                 4
                                 5 CM=-1.
                                3 U=U+1
                        3 U=U+1
7, A2(J)=A2(J)+2.*CK*PAJ/(2.**(N+1))
00 TO 6
10 CALL TORAO(K.PI,PGAS,RG,RN,NT,CAMU,PMM,P1,T1,VP1,A2(J),NTMVP,
1%TMGS,TE0,TFA,VPA)
IC(N1)114,113,114
                114 CN=0.
CNT(I)=C.
                                                °ATE=∩.
                                              ARATE=0.
CO TO 84
                       CALCULATION OF DEAD VOLUME
Ç,
                 113 IF(J-1)81,83,83
```

0.0 17(1-1) 05,04,07 5 3 6-12-1 - CM=C - 4 - CM - TD - 04 - CMT (IN=CMTA TO 94 $r \rightarrow$ VP=(PATIO*(2-Y)*(TPA*YPA-T1*VP1)/X+T1*VP1)/TP SP=VP/VPTO PATE VP/VPTO PATE VP/ CONTINUE IF(M1)107,115,107 115 CMTATECMT(I)+CMTAT (F(CNT(I)/CMFAT -1.5-10)94,95,95 C4 WRIFF(0,1327) CMFAT,CNT(I) C4 \$\RITE(1,1327) CMT21,GNT(1)
N1=]
1027 EDSMAT(77,5X,100000AA CUT-9FF SINCE TOTAL DROP COUNT.',515.4,',
15A2 EXCENDS D2DD COUNT TU FULS SIE2,',E16.5,'.')
C5 TC 107
C5 V0=].-CNT(1)/CCTA
S5T 2 ECS NEXT TIME INTERVAL
NPTT(2,1010) V0
IF(CMTDT-).)105,105,105,105
105 S =0.62
C00 TC 107
106 S =(3.7(4.*DI*CNT0T))**(.33233)
107 CONTINUE
D2U=C 0 C/ C/C(1) 000 D2H=C.0 D3 PF J=1,IM 03 PF J=1,IM C3 D2H=(A./2.) **DI*C(T(J)*(A2(J)**3-A1(J)**3) + DPH C4LL CHLAI(T1, HEAI) C=D2H*HEAI C=D2H*HEAI DTFMP=2.*0*0GAS*F1/(PI*0M**5.*1.9872) DTFMP=2.*0*0GAS*F1/(PI*0***TMVP) T1=T1:DTEM2 NUM_FILE(), NUM_FILE() = NT(Y) T1 = T1 + D1 = M2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D1 + D1 + D2 Y2 = T1 + D TC=T1 VPC=VP1 PO=P1 21=P2 $\Delta TC = \Delta T1$ $\Delta VPC = \Delta VP1$ TIME1=TIME2 1

```
DE1=012
                                                                                \begin{array}{c} 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 2 & (1, 1) = (1, 1) \\ \hline 2 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ \hline 1 & (1, 1) = (1, 1) \\ 1
                                                                                5 ° 13
                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     .
                                                                     205 SPITE(2.1021)
2051 SPITE(2.1021)
                                                                                                                                           STED
                                                                                                                                              ENIP.
                                                        SUBROUTINE TOPAD(K.PI, PGAS, PG, PW, NT, CAMU, P.M. P, T. VPR, AR, WT 1VP,
191365, TED, TEA, VPA)
DEAL K
                                                                      T T AL
                        OFAL K
Y=T+1F,
CALL VPDFD(T, VDFO)
OIFFDSID* COFFFICIENT(CN**2/SEC) AS A F OF T AND MF PATHS AND MOL WFIGHT
AD=.CCD04.6*T + .001
CALL THFOND(T, THFDM. THFDME)
FDA=4.*AD*SOFT(VDNO*21/(3.*FGAS*T))/AF
FTAF=4.*ED*D*C*SODT(VDNOS*PI*E/(0.*PCAS))/((P-VDP)*PM4*A8)
CALL CHEAT(T,0)
A=(FTA+1.)*THEE4/(O*AD*[FTAC+1.))
                                                                      \frac{1}{1} \sum_{i=1}^{n} \frac{1}{2} \sum_{i=1}^{n} \frac{1}
                                                               \begin{array}{l} \nabla = \{ \{ 1, 1, 2\} \} &= -1, 2 \\ \nabla = \{ 1 \} \\ \nabla = \{ 1 \} \\ \nabla = \{ 2 \} \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\ + 2 \\
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                                                                -X=X+DY
                                          1
                                                                             S=11
                                                                      ch th s
                                                              INDEX
FUDWAI(: DID VCI CUANESCE:)
ESTIF(S'IDUS)
1000
                                                                       TRAE(FTAP#TFTEC)/(FTAP+].)
NOA=TFA*(VER/T+RCAS*THERM*(T-TEA)/(Q*AD*PMM*WTWVP))
                                                                      N T = N T + N
                                                                       PETHEN
                                                                       1 MD
                                     SUPERMITTY ( VAREA)
CALCULATION OF ECUILIBRINK VAROR PRESSURE OF WITER
                                                   TC=T-273.15
V0F3=A.F8102+TC+.232075+TC**2*.010758+TC**3*1.96622E-4
1+TC**3*.216463E-5+TC**5*.211191E-7
                                                                  A FTHOM
                                                                   I'MD
                                                   SUPPOUTINE CALAT (T.HEAT)
HEAT=746.1-.55*T
VETUPH
                                                     611D
                                                    SUBBOHTIME SIGMA(T, SEG)
                                                SIG=116.459-.149228*T
E=TURM
                                                                                                                                                                                                                                                                                                                                                                                                                             . . . .
                                                   FAD
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SUPRONTINE THERMO (T, THERME) THERMAL CONDUCTIVITY (CAL/DEG SEC CM) AS A FUNCTION OF T(DEG K) HELTUM THERNE=.435+04+.0035F+C4*T THERMETHERME/4.186C+07 PETURN END

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SUPPORTING TOALO (21,22.T1.T2,VP1.V2) DEAL SEM, ME2. ADUS DEAL SEM, ME2. ADUS DEVENDIVEI 사인 아프 1 프라지션 "PAR="FUT"FR 00707-00-01 N=63S(10.*09TOT) IF(().*09TOT) IF(().*09TOT) 2.DP=DPTOT N=1 00 TO 4 a possorat/M 4 (NE=NEN*00 DEC=ME0*00 00=1.0 020=2.5 T = T 1 $\hat{p} = p\hat{1}$ PET I INET, N DEPERD MO-DAGEN -. TREPART O (A1()@ AD (T2,V0,CD3,00)) [T=T+T*(>C*DPC/DC+(@ 90%00*00*00)/(CDC+0090*C20)) T2=T DETUPN CHIQAN FUD SUFACUTINE NCAP (T, P2, CP2, R2) CM CULATION OF 2 AND CP FOP NATEP VAPOP. TE(T=242) 3.4.4 4 C92=2.1411 GH TC 7 J IF(02=2.8) 5.5.6 6 C92=2.461+(T=240.)*.01201 c0 TC 7 5 C92=2.9611+(T=240.)*.01234+ 1((20.2+P2)/14.71)*(.1953+(T=240.)*.00202) 7 CONTINUE JE(T=235) E.2-9 / (UNTINUE JE(T-235) E, 0.9 0.02=].005+.0005/*(1-325.) 0.010 10 0.02=].005 0.005].00NTINUE 0.00TINUE I THEN TND

TIME INTERNAT	· 2(1)。 7				
P 2 E S S U P E	= \$27.400 MM	HO DURATION C	9F TH IS STEP = 0.00	500C SECONDS	
FAMILY	INITIAL SADIHS	FINAL RADIUS	VE OF OF OP SUR	TEMP OF DROP SUR	GAS TEMP AT DROP
1 2 3 4 5 7	0.302141528-03 0.360015425-03 0.367766255-03 0.253940705-03 0.10840994-03 0.374900715-04 0.09989075-05	0.420937165-03 1.393559445-03 1.354467225-03 1.367591875-03 0.2562946935-03 0.191552375-03 0.974939715-04	2.7320 2.7437 2.7583 2.7792 2.8121 2.8724 3.0598	266.2312 266.2364 266.3562 266.4546 266.6084 265.8857 267.7153	264.8770 264.8184 264.7458 264.6423 264.4807 264.1868 263.2954
0610 VOLUM V&000 OF 01 UATENT MEAT V&000 OVER	SASE COSECTION I DEFITURE LIUN	= 0.3374935-02 = 0.1662775-09 = 0.1003505-06 = 0.149995-03	· · ·	AD 1 49 1 T I C	TOTAL DROPS
15-126 - 21163	TEADERVINGE	■ AVBUD BUESSADE	SUPERSATURATION	CORPECTED NUCLEATION RATE	DROPS THIS STEP
ADIABATIC	258 . 8316 258 . 8302	8.4957 8.4954	5.585413 5.585338	0.72672095E 03 0.72656421E 03	0.36336031E 01 0.36205597E 01

END OF THIS TIME INTERVAL

.

EVALTA	INITIAL PADINS	FINAL RADIUS	A5 or Dbub 208	TENP OF DROP SUP	GAS TEMP AT DROP
١	0.34902502E-03	0.392141530+03		266.5363	265.2322
2		0.350215435-03	2.2449	256.7595	265.1548
	0.199400046-03	C.25394979E+03	2.8992	267.0073	264.8906
5	<u>040000816-04</u>	0.189402945-03	2.0510	267-2878	264.5898
<i>t</i> ,	0.0000007F=05	0.374992715-04	3.1465	268.0842	20301833
DEVO VOLU	·• [⁻	0.2345045+02			TOTAL DROPS
VAPOR DEP EATEMT HE		3.760119E-10			
Add July 1	SSURE CORRECTION =	D.6825458-04	• · · · ·	ADIAPATIC	0.40492125E 01
τεμοριλτυ	FF CORPECTION =	0.1601805-03		$(1,3) \in \mathbb{C} \to (1,2) \to (2,2)$	0.40402241E 01
					DDDDS THIS STEP
		A460% 5862803%	1993 20 S 14 (18 24 (1994)	BOULTAILUN AALL	
ADIABATIC	250.3093	3.5241	5.395782	0.41317090F 03	0.20908537E 01
むけ ビアレト 長行 シー・	259 · 2023	8.5259	2.69757.24	ジャキチウエコンド住宅 ソラ	いってい ロフエモフフト ビキ

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TIME INTERVAL MG. 6

Units are: Length-cm, Volume-cm³, Temperature-°K, Pressure-mmhs, Beat-cal, Time-sec.

PRESSURE = 531.700 MM HG DUPATION OF THIS STEP = 0.005000 SECONDS

TABLE VIII. OUTPUT OF THE DROPLET GROWTH COMPUTER PROGRAM

동 국 학 .	140 . 7 40	0.6	no syer
E 0400000000000000000000000000000000000	$\begin{array}{c} 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 &$	$\begin{array}{c} r_{1}r_{1}r_{2}r_{2}r_{3}r_{4}r_{4}r_{1}r_{1}r_{2}r_{2}r_{2}r_{3}r_{1}r_{1}r_{2}r_{2}r_{2}r_{3}r_{1}r_{1}r_{2}r_{2}r_{2}r_{3}r_{1}r_{1}r_{2}r_{2}r_{3}r_{1}r_{1}r_{2}r_{2}r_{3}r_{1}r_{1}r_{2}r_{2}r_{3}r_{1}r_{1}r_{2}r_{2}r_{3}r_{1}r_{1}r_{2}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{1}r_{2}r_{2}r_{1}r_{2}r_{1}r_{2}r_{2}r_{1}r_{2}r_{2}r_{2}r_{1}r_{2}r_{2}r_{2}r_{2}r_{1}r_{2}r_{2}r_{2}r_{2}r_{2}r_{1}r_{2}r_{2}r_{2}r_{2}r_{2}r_{2}r_{2}r_{2$	51770074444444444444444 517007570575515920775 1

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- F 505056836465555555 - F 505050000000000000000000000000000000	$\begin{array}{c} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 &$	TE 10 7272 • 63 2771 • 10 2771 • 10 2771 • 10 2771 • 10 2771 • 10 2760 • • 77 2760 • • 63 2760 • • 63 2760 • • 63 2760 • • 63 2760 • • 63 2660 • • 63 260 • • 63 271 • 10	- 5 5 13 3 3 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4
CYP.	MA. 754 ppcss 1125.0	1,5 907 7500 203.65 272.25	225700 54300 1.400 3.402

cyp.	MA. 754	1,5 00	1257CC
TINE	ppc55	TEHD DDS CS	3900 1 - 50
0.005		272 26	3 77
C.215	040.0 054 7	271.06	3.94
	CCR P	270 73	4 27
0.035	941 () 941 A	260 71	4.55
0.745	040 2	255.60	4.57

<pre></pre>		277.47 271.70 271.56 277.6.34 277.6.34 277.6.46 277.6.46 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.30 277.6.300 277.6.300 277.6.300 277.6.300 277.6.300 277.6.300 277.6.3000 277.6.3000 277.6.3000 277.7.3000000000000000000000000000000000	34444444444 * · · · · · · · · · · · · · · · · · · ·	
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	$p_1 \rightarrow p_2 = p_2 $	$T_{0,2,2,3}$	slaad 44444444444444444444444444444444444	E T 0015000000000000000000000000000000000	$\frac{100}{100000000000000000000000000000000$	0.7 FERR 275 277 277 277 277 277 277 277	$\begin{array}{c} 10 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 $
EYD.	NM. 750	0.6 08	195700				
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TABLE X

ARGON DATA

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EXP. NO. 667 TIME DEESS C.COM DECS C.COM DEC S C.COM D	0.3 DROPS/CC TEMP SUPR 295.65 1.00 274.79 3.29 274.76 3.41 272.63 3.55 272.64 3.68 272.48 3.91 271.96 3.94	0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	$274 \cdot 0.7$ $372 \cdot 0.3$ $372 \cdot 0.3$ 349 $274 \cdot 0.5$ 345 $274 \cdot 0.6$ 345 $274 \cdot 1.6$ 345 $274 \cdot 1.6$ 346 $274 \cdot 1.6$ 344 $274 \cdot 1.3$ 344
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TIME PRESS 1196.2 0.005 985.5	TEMP SUPR 295.65 1.00 274.68 3.32	0.070 944.1 0.075 957.8 0.090 970.5 0.085 978.6	271.65 273.07 273.98 3.67 273.98 3.47

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EYP, TIME 0.005 0.010 0.015 0.025 0.025 0.030	NO. BC7 PPESS 1196.1 025.5 078.4 071.6 066.4 062.5 957.6	15,400005/00 TEMP SHPQ 295.65 1.00 274.69 2.32 273.90 3.48 273.15 3.66 272.56 3.79 272.13 3.90 271.57 4.64	00000000000000000000000000000000000000	042.2 041.5 043.1 052.1 052.7 070.2	264 84 260 76 260 64 270 96 272 16 273 00	14 44 44 47 44 47 47 47 47 47 47 47 47 47

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0.010	070.8	274.25	3.45
6.015	074.9	272.40	3,53
0.020	949 3	272.93	3.70
0.025	064 5	272 24	3 95
0.030	07.011	271 65	
0.035	045.0	271.27	4.09
0.040	050 0	271 50	4.25
0.045	046.4	270.30	4.39
0.050	042 6	240 37	4.51
0.053	04 1 .0	260,70	4 54
6.055	042 1	260 02	4.50
0.046	954 6	271.16	4.15
- 54 F	944	272 29	3 25
c.o.z.c.	970 0	272.96	3.70
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The author was born on October 5, 1940 in Forbes, Missouri, the eldest son of Mr. and Mrs. Louis B. Allen, Sr. He graduated from the Public High School in Savannah, Missouri in 1958 and entered the Missouri School of Mines where he held a Harry Kessler Scholarship for two years. He worked for the Missouri State Highway Department for a year, then reentered the Missouri School of Mines. He graduated with a B.S. in Physics in June 1963, a M.S. in Physics in June 1964 and has continued work toward the doctorate. He has held a teaching assistantship, a three year National Defense Education Act fellowship and a National Science Foundation Research Fellowship.

The author is married to the former Miss Barbara Leonard of St. Joseph, Missouri. They have three daughters, Trina, Wendy and Laura.

VITA