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Status Report VI NASA Grant NAG 5-865

For the Period 1 March 1990 - 31 December 1990

MICROGRAVITY NUCLEATION AND PARTICLE COAGULATION EXPERIMENTS SUPPORT

Submitted to:

National Aeronautics and Space Administration Goddard Space Flight Center Greenbelt, Maryland 20771

Attention:

Dr. Joseph A. Nuth, III Code 691 Laboratory for Extraterrestrial Physics Space Sciences Directorate

Submitted by:

L. U. Lilleleht Associate Professor

F. T. Ferguson Graduate Student

J. R. Stephens Matrix, Inc.

SEAS Report No. UVA/528260/CHE92/106 January 1992

DEPARTMENT OF CHEMICAL ENGINEERING

SCHOOL OF

ENGINEERING & APPLIED SCIENCE

University of Virginia Thornton Hall Charlottesville, VA 22903

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UNIVERSITY OF VIRGINIA School of Engineering and Applied Science

The University of Virginia's School of Engineering and Applied Science has an undergraduate enrollment of approximately 1,500 students with a graduate enrollment of approximately 600. There are 160 faculty members, a majority of whom conduct research in addition to teaching.

Research is a vital part of the educational program and interests parallel academic specialties. These range from the classical engineering disciplines of Chemical, Civil, Electrical, and Mechanical and Aerospace to newer, more specialized fields of Applied Mechanics, Biomedical Engineering, Systems Engineering, Materials Science, Nuclear Engineering and Engineering Physics, Applied Mathematics and Computer Science. Within these disciplines there are well equipped laboratories for conducting highly specialized research. All departments offer the doctorate; Biomedical and Materials Science grant only graduate degrees. In addition, courses in the humanities are offered within the School.

The University of Virginia (which includes approximately 2,000 faculty and a total of full-time student enrollment of about 17,000), also offers professional degrees under the schools of Architecture, Law, Medicine, Nursing, Commerce, Business Administration, and Education. In addition, the College of Arts and Sciences houses departments of Mathematics, Physics, Chemistry and others relevant to the engineering research program. The School of Engineering and Applied Science is an integral part of this University community which provides opportunities for interdisciplinary work in pursuit of the basic goals of education, research, and public service.

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SUMMARY

Major effort during the current reporting period has been devoted to the evaluation of our very successful first series of microgravity test runs in February 1990. Although the apparatus performed well, it was decided to "repackage" the equipment for easier installation on the KC-135 and access to various components. It will now consists of three separate racks: one each for the nucleation chamber, the power subsystem and the electronics packages. The racks have been fabricated at the University of Virginia and the assembly of the repackaged units is proceeding well.

Preliminary analysis of the video data from first microgravity flight series has been performed and the results appear to display some trends expected from Hale's Scaled Nucleation Theory of 1986.

The data acquisition system is being refined by Dr. Stephens of Matrix, Inc. He has also been exploring and developing potential working relationships with the Microgravity Advanced Research Support (MARS) Center in Naples, Italy.

Several presentations have been made on our progress as discussed in the body of this report and abstracts included in the Appendices.

INTRODUCTION

The formation and interaction of fine-grained refractory particulates is of interest to diverse fields of science, including space sciences. Uniform, "quiescent" suspensions of mono-disperse particles in low-pressure gas are very difficult to produce in terrestrial laboratories by injection or resuspension of previously characterized particulates. Many of these difficulties, however, can be avoided in a microgravity environment thus making it possible to produce such suspensions for characterization.

This project is a part of a program initiated by researchers at NASA Goddard Space Flight Center to study the formation and growth of cosmic dust grain analogues under terrestrial as well as microgravity conditions. Its primary scientific objective is to study the homogeneous nucleation of refractory metal vapors and a variety of their oxides among others, while the engineering, and perhaps a more immediate objective is to develop a system capable of producing mono-disperse, homogeneous suspensions of well-characterized refractory particles for various particle interaction experiments aboard both the Space Shuttle and the Space Station. Both of these objectives are to be met by a judicious combination of laboratory experiments on the ground and aboard NASA's KC-135 experimental research aircraft.

The University of Virginia Department of Chemical Engineering has participated in this Microgravity Nucleation and Particle Coagulation Program since January 1, 1987 under Grant NAG-5-865. The contributions of the University of Virginia and its Matrix, Inc. subcontractor during the past project period are summarized here.

RESEARCH ACCOMPLISHMENTS

University of Virginia Effort

Major effort during the current reporting period has been devoted to modifications of the nucleation apparatus as a result of our first experiences with microgravity test runs in February 1990. The equipment has been completely "repackaged" for easier installation on the KC-135 and access to various components. It now consists of three separate racks:

CHAMBER RACK - which also contains the vacuum pump, inert gas cylinder and miscellaneous attachments to the nucleation chamber, such as the laser light source, video camera, pressure gauges, accelerometers, etc.

POWER RACK - including switch boxes, an inverter, a UPS system, variacs and transformers; and

ELECTRONICS RACK - with computer, monitor, the data acquisition system and various controls.

Other modifications inside the nucleation chamber include an improved mechanism for operating the vapor spout shutter, a series of thermocouples stretched diagonally across the interior of the chamber for temperature measurements, a timer, and LED's to indicate when data are actively being acquired.

Preliminary analysis of the video data from the February 1990 microgravity test series has been performed. Although there was considerable uncertainty about the actual temperatures at the visually observed nucleation front, the results appear to display some trends expected from Hale's Scaled Nucleation Theory of 1986.

Further development of the mathematical models describing the evolvement of the temperature and vapor concentration profiles has been temporarily suspended awaiting experimental nucleation data from the improved apparatus.

The following is a list of presentations made by the University of Virginia members of the microgravity nucleation project:

"Apparatus for Microgravity Nucleation Studies of Magnesium Vapors", a poster paper presented by L.U. Lilleleht (co-authors F.T. Ferguson, J.A. Nuth, III) at the 22nd Annual Meeting of the American Astronomical Society Division of Planetary Science, Charlottesville, VA, October 22-26, 1990, (See APPENDIX A for an Abstract of this presentation.); "Microgravity Nucleation of Refractory Materials: Modeling of Transport Processes in the Nucleation Chamber", a paper presented by F.T. Ferguson (co-authors L.U. Lilleleht, J.A. Nuth,III, and J.R. Stephens) at the AIChE Annual Meeting in Chicago, IL, November 11-16, 1990, (See APPENDIX C for a copy of the manuscript.).

In addition to these, a presentation entitled "Microgravity Nucleation of Refractory Vapors" was made at the University of Virginia Chemical Engineering Colloquium Series on September 13, 1990 by L.U. Lilleleht. He and F.T. Ferguson participated also at the UVA School of Engineering and Applied Science Open House during the Engineers' Week on February 23, 1991.

Matrix, Inc. Effort

During this contract period the primary work was on evaluating the flight data obtained during February, 1990 and planning for the next flight series. The work centered on three areas: (i) evaluating the present equipment and data, (ii) helping our Italian colleagues to plan their microgravity studies which will be coordinated with ours, and (iii) preparing for the next flight series with improved apparatus and diagnostic capabilities. In addition to the various "packaging" changes of the apparatus, improvements that need to be made for future flights include better synchronization of the thermocouple, accelerometer and video data, improving temperature measurements, and increasing the brightness of the video images.

To address the need for quantitative video data, John Stephens has done some research on video cameras, recorders, and possibilities for obtaining multi-color video data. His recommendation is to use a sensitive monochrome video camera, such as the Cohu CCD camera, for example. Cameras of this type are at least five times more sensitive than the camera currently Further, the exposure time of the camera can be in use. increased to provide even more sensitivity, if required. To record high resolution images from the camera, a monochrome VCR recorder, such as a Sony VO-5800, will be required. This recorder also records time on the tape which the old recorder could not do. Finally, a color filter wheel could be added to the monochrome camera to obtain video images in various colors.

John Stephens also assisted our Italian colleagues in defining their program in microgravity condensation experiments which includes adding equipment to our flight experiment package. The Italians are interested in developing a video analysis system similar to that which we are planning. In addition, they have computational hydrodynamic modeling capabilities that will complement ours. Matrix assisted the Italian group also with their experimental design, in coordination with our experiments, and obtaining flight qualification physicals. We anticipate that some of the Italian group will fly with us on the next series.

To improve the synchronization of the thermocouple and video data, the data acquisition computer program has been modified to open the chamber shutter automatically and start the thermocouple measurements when the accelerometer indicates less than 0.1 G and terminate data collection when the G-level rises above 0.8 G. In addition, the program has been modified to turn on and off a light emitting diode during the microgravity phase of the flight which will be recorded by the video camera. These computer control modifications, together with the "repackaging" of the apparatus, should more accurate temperature, acceleration, and time data to be collected on the next series of flights.

A poster paper entitled "A Research Program to Study Microgravity Condensation of Cosmic Dust Analogue Materials", was presented by J.R. Stephens (with co-authors J.A. Nuth, L.U. Lilleleht, F.T. Ferguson, L. Colangeli, E. Bussoletti, C. Mirra, D. Mancini, A. Vittone, M. Cavacece, and L. Napolitano) at the COSPAR Meeting in The Hague, Netherlands, June 1990. An abstract of this is presentation is attached as APPENDIX B.

APPENDICES

A:	AAS-DPS	Poster	Abstract	by	L.U.	Lilleleht,	et	al	•
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B: COSPAR Poster Abstract by J.R. Stephens, et al.

C: AIChE Paper Presented by F.T. Ferguson, et al.

APPENDIX A

Abstract of a Poster Presentation by L. U. Lilleleht at the 22nd Annual Meeting of the American Astronomical Society Charlottesville, VA October 22-26,1990

NUCLEATION AND CONDENSATION OF REFRACTORY VAPORS

by

L.U. Lilleleht, F.T. Ferguson, J.A. Nuth III, J.R. Stephens

Laboratory data appear to indicate that refractory vapors do not follow the classical theories of homogeneous nucleation. We have therefore undertaken an experimental program to investigate this interesting process which may well have implications ranging from how particulate pollutants are formed and emitted from power plants to the origins of cosmic dust.

A nucleation chamber of a novel design will be discussed along with our experiences with magnesium vapor in both terrestrial (1-g) as well as in microgravity (0-g) environments aboard NASA's KC-135 research aircraft.

APPENDIX B

Abstract of a Poster Presentation by J. R. Stephens at the COSPAR Meeting in The Hague, Netherlands June, 1990

A RESEARCH PROGRAM TO STUDY MICROGRAVITY CONDENSATION OF COSMIC DUST ANALOGUE MATERIALS

by

J.R. Stephens, J.A. Nuth, L.U. Lilleleht, F.T. Ferguson, L. Colangeli, E. Bussoletti, C. Mirra, D. Mancini, A. Vittone, M. Cavacece, and L.G. Napolitano

Cosmic dust is mainly formed by nucleation processes in circumstellar shells, and it is further processed by chemical, physical and collision mechanisms in the interstellar medium. One of the aims of "Solid state astrophysics" is to simulate - as far as possible - processes of cosmic dust formation in the laboratory. In the past years, our group has produced cosmic grain analogues with various chemical composition by means of different laboratory techniques in order to investigate their morphological, structural and optical properties and to interpret astronomical observations. However, a more appropriate simulation of actual conditions in space can be achieved in 0-g conditions.

Recently, we have started a research program for the production of dust samples in low-gravity environment by means of experimental methods similar to those already employed in the laboratory. These experiments will provide useful information about condensation processes active in space; they will allow us to perform optical and morphological investigations on "isolated" grains and/or clusters which are not easily obtained on the ground.

In this paper we will present a program for our future experiments and some preliminary results about theoretical computations on nucleation processes in 0-g conditions.

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APPENDIX C

Manuscript of the paper presented by F.T. Ferguson at the AIChE 1990 Annual Meeting in Chicago, IL November 16, 1990

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Microgravity Nucleation of Refractory Materials: Modeling of Transport Processes in the Nucleation Chamber

by

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Microgravity Nucleation of Refractory Materials: Modeling of Transport Processes in the Nucleation Chamber

L.U. Lilleleht¹, F.T. Ferguson², J.A. Nuth, III³, and J.R. Stephens⁴.

Abstract

A two-part mathematical model has been developed to describe the transport processes in a nucleation chamber designed for condensation of refractory vapors in a microgravity environment. The model solves the transient diffusion equations for temperature and concentration fields in cylindrical coordinates using finite differences and the alternating direction implicit method. Vapor supersaturation ratios are then computed from the evolving concentration profiles thus permitting one to estimate the conditions at the location in the chamber where nucleation is observed in experiments.

Introduction

The process of nucleation and condensation of refractory vapors has importance not only to the astrophysicist for gaining insight to the origins of interstellar or cosmic dust [1,2], but it can also have more immediate practical applications. Thus, for example, the uniform suspensions of monodispersed ultrafine particles produced by such a process are expected to have special catalytic effects when consolidated into "nanophase" materials [3]. Additionally, it can also lead to a better understanding of how pollutant particulates are formed in combustion processes. We have therefore undertaken a program to study the nucleation of refractory vapors and the subsequent growth and coalescence of the solid particulates.

In 1935 Becker and Doring developed what has become to be known as the Classical Theory of Homogeneous Nucleation [4]. This theory has been shown to work quite well with relatively volatile materials, such as water and alcohols. However, the limited data that are available on the nucleation of refractory vapors do not appear to agree with the classical Becker-Doring theory [5]. Of course, such experiments are much more difficult to conduct, particularly when precise thermodynamic measurements of nucleation conditions are needed.

Hale has recently proposed a Scaled Theory of Homogeneous Nucleation which uses the critical point data and scaled quantities to develop an essentially materialindependent expression for conditions which will produce a flux of clusters at the rate of 1 per cm³ per second [6]. The results of this are encouraging, as they compare

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favorably with data from experiments with volatile materials as well as with some refractory data [7]. More experimental observations are needed, however, to validate this or develop other nucleation theories.

Most of the nucleation data with refractory materials have been obtained either in shock tubes or by the gas evaporation method. In the shock tube apparatus metallic vapor is produced from the dissociation of a volatile organometallic precursor compound as the shock passes through the tube. The vapor cools, becomes supersaturated and condenses, with the entire event taking place in microseconds [8]. In the gas evaporation method the refractory material is heated at some reduced pressure under an inert atmosphere. When a sufficiently high temperature is reached, significant amount of vapor is produced which is then allowed to diffuse into a lower temperature region where it becomes supersaturated and condenses [9].

Donn et al. have used this evaporation- condensation method to study SiO, Mg + SiO, and Ag [10,11]. They encountered some problems with this method arising from the necessarily high temperatures and temperature gradients. Radiant heat transfer interferes with accurate temperature measurements at the point of nucleation, and high temperature gradients produce severe convective currents which may affect the nucleation conditions. We have therefore modified the gas-evaporation apparatus of Donn et al. with the objective of minimizing these difficulties. Convective currents were reduced by locating the highest temperature region at the top of the chamber, and a mathematical model of transport processes occurring in the chamber was developed to predict conditions at the visually observable point of nucleation [12]. Also, the new nucleation chamber was built so that it could be operated aboard NASA's reduced gravity research aircraft, KC-135, in order to reduce the convection even further. This aircraft is capable of flying parabolic trajectories producing a series of 20 to 25-second windows of microgravity at the peaks of the arches. Between these periods of weightlessness, however, the g-forces can reach 1.8 to 2 g's in the troughs.

New Nucleation Apparatus

The new nucleation chamber consists of an approximately 60-cm diameter and 60-cm high vacuum chamber constructed of aluminum. The lower and upper sections of the chamber are separated by a plate assembly supporting the furnace region as shown in Figure 1. On the plate separating the two halves of the chamber, and nestled within a set of resistance heaters, are small crucibles of the material to be studied. The heaters are covered with insulation and then the top section of the chamber. Vapor leaves the crucibles diffusing downward into the lower half of the chamber through a spout which can be closed by a mechanical shutter. The lower section is provided with four viewing ports equidistant around the circumference. An essentially twodimensional fan of laser light is passed through one of the ports to illuminate a thin, vertical cross-section of the viewing region and the condensed particles, if present, as evidenced by the smoke cloud. This image is then captured by a video camera at a 90-degree angle to the light fan.



Figure 1: Cross-Section of the Nucleation Chamber

During a typical experiment the crucibles are charged with the refractory material, the top section of the chamber is bolted to the bottom, and the chamber is pumped down to remove oxygen. The system is purged with an inert gas, such as argon, and then pumped down to somewhat below the desired operating pressure. With the shutter closed, the crucibles are heated, usually considerably above the melting point of the metal to generate sufficient vapor pressure for the experiment. Once the desirable temperatures have been reached, which for Mg has been approximately 950°C, the shutter is opened allowing the metal vapor to enter through the spout and to diffuse into the lower-temperature region. On the KC-135 flights the shutter is opened soon after microgravity conditions have been established, and closed again when g-forces become significant. The location of the smoke cloud is captured by a video camera and monitored on a television screen. Temperatures and accelerometer outputs are recorded using a computerized data acquisition system.

Thermocouples are installed to measure temperatures on the chamber surfaces. However, they may not be placed where the smoke cloud is expected to form because of reading errors due to radiation and their possible interference with the homogeneous nucleation process there. Temperatures at the smoke cloud interface must therefore be obtained from mathematical models solved with thermocouple measurements in less critical areas as boundary conditions.



Figure 2: Geometry of the Chamber Viewing Region and the Boundary Conditions for the Model

Mathematical Models

The degree of supersaturation at the point of nucleation cannot be measured directly in this experiment and, therefore, has to be estimated from some model. Both the temperature and concentration fields are computed assuming molecular diffusion mechanism only [12]. In cylindrical coordinates, the equation for the transient diffusion of a property, f, in the r - z coordinates is given by:

$$\frac{\partial f}{\partial t} = D \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial f}{\partial r} \right) + \frac{\partial^2 f}{\partial z^2} \right]$$
(1)

where D is the diffusion coefficient. The geometry of the nucleation chamber viewing region for which this equation is solved is shown in Figure 2. Solutions of Equation 1 are obtained by numerical methods using finite differences, because of the rather complicated geometry and boundary conditions. Only one half of the viewing region needs to be modeled because of symmetry. The region to the right of the chamber centerline is divided into a 31 \times 31 gridwork, and the resulting finite difference equations are solved using the alternating direction implicit method.

Temperature Profile

As heat transfer in the viewing portion of the chamber is assumed to be by conduction only, the equation for the temperature is:

$$\frac{\partial T}{\partial t} = \alpha \left[\left(\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) \right) + \frac{\partial^2 T}{\partial z^2} \right]$$
(2)

where α is defined as the thermal diffusivity. Only the steady state temperature profile is used in this work. There are two options for obtaining the steady state profiles: (i) solve the steady state equation directly, or (ii) solve the unsteady equation for long times. The second approach is used here. The thermal diffusivity of the argon gas is calculated from an approximate expression fit to published data. The expression for the thermal diffusivity is:

$$\alpha_{Ar} = 8.32 \times 10^{-3} \frac{T^{1.73}}{P} \tag{3}$$

where P is the total pressure in torr and T is the absolute temperature in kelvin.

Temperature Boundary Conditions

The temperatures at all points, other than the centerline, are evaluated either from an one-dimensional conduction model or obtained from experimental measurements. The boundary conditions used in the temperature model are shown in Figure 2. At the centerline, where r = 0, the term (1/r) "blows up." To remedy this, the limiting form of this term is used. By L'Hospital's rule:

$$\lim_{r \to 0} \left(\frac{k}{r} \frac{\partial T}{\partial r} \right) = \frac{k \frac{\partial^2 T}{\partial r^2}}{1} = k \frac{\partial^2 T}{\partial r^2}$$
(4)

As a result:

$$\frac{\partial T}{\partial t} = \alpha \left[2 \frac{\partial}{\partial r} \left(\frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(\frac{\partial T}{\partial z} \right) \right]$$
(5)

This equation is then used in determining the temperatures along the centerline.

Concentration Profile

Another major goal of this project is to determine the concentration profile as it evolves while the refractory material vapors diffuse into the viewing region of the chamber. Unlike the temperature profile, which is assumed to have reached and to maintain a steady state prior to the opening of the shutter, the concentration profile requires a transient solution of the diffusion equation. This calculation makes use of the already established temperature field within the viewing chamber to evaluate the various temperature-dependent physical properties needed to find the partial pressure and from that the supersaturation ratio throughout the viewing area.

Since it is not known a priori at what conditions and where condensation will occur, we cannot account for it in our calculations. As a result, we assumed that the vapor always remains as vapor throughout the lower region, even after the onset of nucleation and the appearance of the smoke front. Thus the condensation process is essentially ignored, and the present model can therefore be expected to apply only up to the appearance of the smoke cloud. The equation for the transient diffusion of vapor in the chamber is given by:

$$\frac{\partial C}{\partial t} = D_{AB} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right) + \frac{\partial^2 C}{\partial z^2} \right]$$
(6)

where C is the concentration of the vapor A and D_{AB} is the diffusion coefficient of A through B. In this model, it is assumed that both the inert, carrier gas and the refractory material vapor behave as ideal gases. The concentration and partial pressure are related by:

$$C = \frac{P_p}{RT} \tag{7}$$

where P_p is the partial pressure of the refractory material. Calculation of the concentration is simplified by letting C be normalized. Therefore, the initial conditions for the concentration at the the spout area is saturation or C = 1.0, and at every other point within the chamber C = 0. The diffusion coefficient, D_{AB} , for the refractory material/inert gas system is estimated from:

$$D_{AB} = 1.858 \times 10^{-3} \frac{T^{1.5} (M_A + M_B)^{0.5}}{P \sigma_{AB}^2 \Omega_D (M_A M_B)^{0.5}}$$
(8)

From the temperature and concentration profiles, the supersaturation ratio can be calculated. The supersaturation ratio, S, is defined as the ratio of the local partial pressure of the vapor to the equilibrium vapor pressure:

$$S = \frac{P_p}{P_{eq}} \tag{9}$$

where P_p is the partial pressure of the vapor and P_{eq} is the equilibrium vapor pressure.

Concentration Boundary Conditions

Concentration boundary conditions used in model calculations are also shown in Figure 2. The vapor pressure of the refractory material is assumed to equal the equilibrium vapor pressure of the material at the temperature of the spout area, and the normalized C = 1.0. The same type of limiting form of the PDE is required at the centerline as was used for the temperature program to avoid problems with singularities. Finally, at all of the solid surfaces it is assumed that the flux of vapor is zero. In other words:

$$\left. \frac{\partial C}{\partial n} \right|_{wall} = 0 \tag{10}$$

where n is the coordinate direction normal to the wall.



Figure 3: Temperature Profile

Results

Temperature Profile

Figure 3 is a plot of the isotherms for the case where the spout area is at 1000K and the bottom plate of the viewing region is at 300K. All other wall temperatures are estimated using a simple, one-dimensional conduction model along the walls. The isotherms have also been normalized such that they range from 1.0 at the spout to 0 at the bottom of the chamber. The computed isotherms tend to be hemispherical about the spout, and there is a large temperature drop in this area. Much of the viewing region of the nucleation chamber is still fairly cool. The need to include radiant heat transfer in the temperature model was also explored. Even though the temperature field was affected at some distance from the vapor source, near the spout the differences were minimal as far as the early development of the concentration field was concerned. This was the basis for assuming transport by molecular diffusion mechanisms only.

Concentration Profile

Although the model is solved for the concentration profile, the quantity of real interest is the supersaturation ratio, S. Since the supersaturation ratio depends on the local equilibrium vapor pressure, the S-profile requires the knowledge of the temperature field. Figures 4 and 5 are plots of the supersaturation ratio for magnesium vapor in an argon atmosphere of 200 torr. Because refractory materials have very low vapor pressures and hence high numerical values of supersaturation ratios, the contours shown in Figures 4 and 5 are actually lines of constant base 10 logarithms of the supersaturation ratio, i.e. log(S). These plots, and all subsequent supersaturation data, are based on the temperature profile of Figure 3. The first plot represents the log(S) at 1 second after opening the shutter, and the second plot is 2.5 seconds



Figure 4: Log(S) for Mg vapor after 1 second



Figure 5: Log(S) for Mg vapor after 2.5 seconds

Maximum Log(S)									
	40 torr			200 torr			400 torr		
Metal	1s	2.5s	5s	1s	2.5s	5s	1s	2.5s	5s
Mg	7.53	9.42	10.9	4.42	6.14	7.54	3.32	4.85	6.18
Zn	5.73	7.31	8.53	3.18	4.58	5.74	2.30	3.52	4.61
Sn	10.4	12.9	14.9	6.29	8.55	10.4	4.80	6.96	8.58
Pb	9.36	12.3	15.4	4.73	7.26	9.38	3.12	5.37	7.32

Table 1: Ambient Pressure Effect on Log(S) at Source Temperature 1000K

after the opening.

Portions of log(S)-contours also appear to be somewhat hemispherical in shape. But unlike the monotonically decreasing temperature profile, the supersaturation goes through a maximum between the vapor source and the bottom plate. Saturation conditions at the vapor spout give S = 1.0, while S = 0 at the bottom plate because the vapor has not yet reached there. Between these two extremes, though, the concentration is sufficiently high and the temperature low enough to generate high supersaturation ratios.

The Effect of Ambient Pressure

The effect of ambient pressure is illustrated in Table 1. To simplify interpretation of the results, only the maximum achievable supersaturation ratios at different total system pressures are listed. Since we are planning to investigate the behavior of other metals such as zinc, tin and lead as well, some expected results for them are also presented. The supersaturation profiles for these metals are similar in shape to those of magnesium, but they have very different supersaturation ratios because of their different equilibrium vapor pressures and diffusion coefficients. This table was compiled from the centerline results. The maximum supersaturation ratio increases for each material as the ambient pressure in the chamber is decreased. This increase may be attributed to two factors. First, as the chamber pressure is decreased, the resistance to diffusion decreases. Secondly, at a constant source temperature the relative concentration of the refractory vapor itself is higher because of the lower total pressure.

The Effect of Crucible Area Temperature

Table 2 shows the effect of increasing the crucible or source temperature from 1000K to 1250K. In both cases the temperature of the bottom plate is 300K. The chamber temperature profile for the 1250K source is not shown, but it should be very similar in shape to that of Figure 3. Again, only the maximum values of the log(S) are given. As expected, the maximum supersaturation ratios have increased at higher crucible temperatures. Most of this increase is probably due to the fact that the hotter crucible temperature produces a greater concentration of vapor in the spout area. The diffusion rate of the vapor is also higher at elevated temperatures.

	Maximum Log(S)							
	\overline{T}_h	=1000	Γ.	$T_{h} = 1250 \text{K}$				
Metal	1s	2.5s	5s	1s	2.5s	5s		
Mg	4.42	6.14	7.54	5.56	7.28	8.68		
Zn	3.18	4.58	5.74	4.13	5.53	6.70		
Sn	6.29	8.55	10.4	13.1	16.2	18.7		
Pb	4.73	7.26	9.38	6.55	9.10	11.2		

Table 2: Source Temperature Effect on Log(S) at 200 torr Argon Pressure

Let us now, for example, consider the case of magnesium. By increasing the temperature 250K, the maximum logarithm of the supersaturation ratio along the centerline is increased by 1.14 at 2.5 seconds. However, by decreasing the pressure from 200 torr to 40 torr (Table 1) the maximum logarithm of the supersaturation ratio at 2.5 seconds is increased by 3.28. Adjusting the chamber pressure is relatively easy and can be accomplished much more quickly than adjusting the crucible area temperature. Therefore, pressure adjustment should be the preferred means of controlling the operation of the nucleation chamber.

Summary

A new design of a cylindrical chamber to investigate the nucleation and condensation of refractory vapors is described. Mathematical models have been developed to predict the fully established temperature field and the time-dependent supersaturation ratios within this nucleation chamber. These programs solve the two dimensional, transient diffusion equations in cylindrical coordinates using finite differences and the alternating direction implicit method.

Several conclusions can be drawn from the results of running these two programs:

- There is a large temperature drop near the vapor inlet to the chamber.
- The total chamber pressure has a significant effect on the supersaturation profile.

Since the chamber pressure is much easier to adjust than the crucible area temperature, pressure adjustment will provide the most convenient way of increasing or decreasing the maximum attainable supersaturation ratio. This will be especially important for the KC-135 flights since there is little time to adjust parameters between successive sets of parabolic arches.

References

- B. Donn, "Some aspects of Condensation in Clouds", Astrophysics and Space Science, 65, 167 (1979).
- [2] B. Donn, R. Khanna, J.A. Nuth, D. Stranz, and A.B. Anderson, "The Formation of Cosmic Grains: An Experimental and Theoretical Study", Surface Science, 106, 576 (1981).
- [3] J. Eastman and R.W. Siegel, "Nanophase Synthesis Assembles Materials from Atomic Clusters", Research & Development, 31(1), 56 (1989).
- [4] F.F. Abraham, Homogeneous Nucleation Theory: The Pretransition Theory of Vapor Condensation, Academic Press, New York (1974).
- [5] B. Donn and J.A. Nuth, Astrophys. J., 288, 187 (1985).
- [6] B.N. Hale, "Application of a Scaled Homogeneous Nucleation-Rate Formalism to Experimental data at $T \ll T_c$ ", Phys. Rev. A, 33, 4156 (1986).
- B.N. Hale, P. Kemper, and J.A. Nuth, "Analysis of Experimental Nucleation Data for Silver and SiO Using Scaled Nucleation Theory", J. Chem. Phys., 91, 4314 (1989).
- [8] H.J. Freund and S.H. Bauer, "Homogeneous Nucleation of Metal Vapors 2: Dependence of the Heat of Condensation on Cluster Size", J. Phys. Chem., 81, 994 (1977).
- [9] K.L. Day and B. Donn, "Condensation of Nonequilibrium Phases of Refractory Silicates from the Vapor", Science, 202, 307 (1978).
- [10] K.L. Day and B. Donn, "An Experimental Investigation of the Condensation of Silicate Grains", Astrophys. J., 222, L-45 (1978).
- [11] J.A. Nuth, K.A. Donnelly, B. Donn, and L.U. Lilleleht, "Experimental Studies of the Vapor Phase Nucleation of Refractory Compounds. III. The Condensation of Silver", J. Chem. Phys., 85(2), 1116 (1986).
- [12] F.T. Ferguson, "Modeling of Transport Processes in a Nucleation Chamber", M.S. Thesis, University of Virginia (1990).

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