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The report presents fifteen papers from a workshop on microgravity science and applications held at the Jet Propulsion Laboratory in Pasadena, California, on December 3-4, 1984. The workshop and panel were formed by the Solid State Sciences Committee of the Board on Physics and Astronomy of the National Research Council in response to a request from the Office of Science and Technology Policy. The goal was to review the microgravity science and applications (MSA) program of NASA and to evaluate the quality of the program. The topics for the papers are metals and alloys, electronic materials, ceramics and glasses, biotechnology, combustion science, and fluid dynamics.

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Microgravity Science and Applications

Report on a Workshop

Microgravity Science and Applications

Report on a Workshop

December 3-4, 1984
Pasadena, California

Panel on Microgravity Science and Applications

Solid State Sciences Committee
Board on Physics and Astronomy
Commission on Physical Sciences, Mathematics, and Resources
and
National Materials Advisory Board
Commission on Engineering and Technical Systems
National Research Council

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NOTICE: The project that is the subject of this report was approved by the Governing Board of the National Research Council, whose members are drawn from the councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. The members of the committee responsible for the report were chosen for their special competences and with regard for appropriate balance.

This report has been reviewed by a group other than the authors according to procedures approved by a Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

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PREFACE

The National Aeronautics and Space Administration (NASA) has had a strong interest in carrying out materials-related research and development in space since the early 1970s. Exploratory experiments in crystal growing, bioprocessing, and containerless processing were conducted as early as 1973-1974 on Skylab missions. Much of this work was poorly conceived and controlled, and the results were of dubious quality. Yet NASA continued to have high hopes for space processing of exotic alloys, for manufacturing of perfect crystals, and for purifying important biological materials, although interest in space processing was limited to those materials that were of sufficient value or importance to offset the cost of spaceflight and extraterrestrial processing.

In the late 1970s, the National Research Council's Committee on Scientific and Technological Aspects of Materials Processing in Space (STAMPS) reviewed the NASA materials processing programs in detail. The STAMPS report, issued in 1978, was negative. It was critical of the planning of the experiments performed, and no evidence for economically justifiable processes for producing materials in space was found. Nonetheless, the report concluded that there was an opportunity for meaningful science and technology developed from experiments in space "provided that problems proposed for investigation in space have from the outset a sound base in terrestrial science or technology, and that the proposed experiments address scientific or technical problems."

After the STAMPS report was issued, the NASA program was substantially modified in response to the report's recommendations. The content of the program was changed: more scientific content was incorporated, and a substantial ground-based program was added. The program name has been changed to the NASA Microgravity Science and Applications (MSA) Program.

Recently the Office of Science and Technology Policy asked the National Research Council's Board on Physics and Astronomy (BPA) to carry out an assessment of the state of health of the present MSA program. In response, the BPA, working with its Solid State Sciences Committee (SSSC) and the National Materials Advisory Board (NMAB), initiated a plan for a workshop that would provide such an assessment. Brian R. T. Frost of the NMAB and A. I. Schindler of the SSSC cochaired the workshop. The present report summarizes the workshop and gives assessments and recommendations made by the workshop panel.

The Panel cochairmen acknowledge the help and cooperation of Donald C. Shapero, Staff Director of the Board on Physics and Astronomy, and the BPA staff; Richard E. Halpern, Director of the NASA MSA Program, and the NASA program staff; Don Rea, Assistant Director of the Caltech Jet Propulsion Laboratory, and Dan Elleman and his staff at JPL.

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Panel Report

INTRODUCTION

A Workshop on Microgravity Science and Applications, sponsored by the National Research Council's Solid State Sciences Committee (SSSC) and National Materials Advisory Board (NMAB), was held at the Jet Propulsion Laboratory, Pasadena, California on December 3-4, 1984. Approximately 75 scientists, representing industry, academia, and government, participated. The Workshop originated in a request from the Office of Science and Technology Policy to the SSSC to assess the prospects for materials research and industrial processing in the microgravity environment. The Workshop had the following goals: (1) to review the research program of the Microgravity Science and Applications (MSA) Program of the National Aeronautics and Space Administration and (2) to evaluate the quality of the program. The prospects for developing industrial processes in the microgravity environment were considered in the discussions and as part of the overall evaluation.

The two-day Workshop was structured to provide the participants with an overview of the entire MSA program. The program was divided into the following six major topics, which parallels the organization of the NASA Microgravity Science and Applications program:

- Metals and Alloys
- Electronic Materials
- Ceramics and Glasses
- Biotechnology
- Combustion Science
- Fluid Dynamics

Each topic was discussed for about 2 hours. Each session began with an 1-hour overview in which the general content and primary scientific objectives of the MSA program in the area of the topic to be covered by the session were addressed. The overview also addressed to a limited extent the applications potential and opportunities for commercialization. Following the overview presentation an outstanding feature of the topic for the session was presented in detail and in technical depth. For those sessions for which theory was a critical element, a presentation of the theory was also given. Each session concluded with a discussion period in which Workshop participants were invited to discuss critically the material presented. A complete agenda and summaries of the talks given at the workshop are reproduced in Part II of this report.

The Panel consisted of two experts for each session, one to act as session chairman, the other as discussion leader. Following the Workshop adjournment, each pair of panel experts was asked to provide a concise assessment of the work presented in their session addressing the quality of the program and, as appropriate, the opportunities for commercialization. In examining the quality of the program, experts were asked to assess how well the topic was understood in unit gravity and the necessity for conducting pertinent research in a microgravity environment. In considering the opportunities for commercialization the experts were asked to consider competing processes in unit gravity and the value added to materials by the microgravity processing. These assessments are given in the following Sections.

GENERAL CONCLUSIONS

The conclusions and recommendations are given with the realization that they are made solely on the basis of a two-day Workshop. Clearly not all the work being carried out in the Microgravity Science and Applications (MSA) program could be presented, nor was there time for in-depth questioning of the speakers during the discussion period. However, the Panel assumed that the speakers emphasized the more important aspects of the program and highlighted the major successes. With these caveats in mind, some general conclusions can be drawn concerning the program.

1. There has been a considerable restructuring of the program following the STAMPS report (Materials Processing in Space, Report of the NRC Committee on Scientific and Technological Aspects of Materials Processing in Space, National Academy of Sciences, Washington, D.C., 1978). The program has been significantly changed by incorporating more scientific content and by adding a substantial ground-based program for the baseline effort.

2. The quality of the science being carried out is mixed; some topical areas are outstanding, such as that of metals and alloys and fluid dynamics, while some are less developed or are of dubious quality such as parts of glasses and ceramics and of combustion science.

3. Long-range opportunities for commercialization appear to exist, but care should be taken that they are not oversold or inappropriately accelerated.

4. In many cases theoretical modeling in zero g has not been done and would be useful if added to the program.

5. A clear necessity for research being carried out in the microgravity environment is not apparent for parts of the program.

6. Opportunities for materials processing in space should not be viewed as the only justification of the program; in addition to an improved understanding of the science, a considerable return on investment is likely to result from improved technology based on research and development carried out in micro g, but implemented in unit g.

7. Impartial, in-depth peer review of the program and its components should be carried out on a periodic basis.

8. The balance of the present program is questionable and should be reviewed now and periodically in the future by an advisory committee.

9. The hardware for carrying out materials research in space should be viewed and organized as a national user facility such as the National Synchrotron Light Source at Brookhaven.

10. It is difficult to assess long-term potential for commercialization of the MSA program because comprehensive economic studies and cost data were not requested and were not available at the Workshop.

11. No attention is being given to complementary high-g research; at present the research is carried out at only two values--unit g and micro g. Higher-g research should be added, e.g., using centrifuges.

12. A substantial part (26 percent) of the program dealing with Centers of Excellence and the Microgravity Materials Science Laboratory was not reviewed. NASA should treat its Centers of Excellence in the same way that the National Science Foundation treats its Materials Research Laboratories with respect to both site selection and periodic review.

13. Consideration should be given to sponsoring research in some new areas, e.g., welding and tribology.

14. Communication and coordination between various sectors of NASA, e.g., between MSA and advanced technology, could be improved in the interests of optimizing the total program.

PRINCIPAL CONCLUSIONS AND RECOMMENDATIONS OF THE TOPICAL ASSESSMENTS

The highlights of the Panel assessments have been excerpted for each of the six topics of the Workshop, and here we present the principal conclusions and recommendations. The full assessment is provided in the following section.

Metals and Alloys

- The program has improved since the STAMPS report.

- Theory and experimental work in solidification science are of a high standard and should receive increased support.

- Impartial scientific review of critical program areas should be carried out periodically.

• Efforts should be made to bridge the gap between the NASA science- and technology-driven microgravity programs.

• Some important opportunities have been missed, e.g., in friction and fusion welding.

Electronic Materials

• A greater than unit-g experimental capability should be added to the program.

• Further steps need to be taken to ensure better coupling between the theoretical and experimental components of the program.

• Crystal-growth studies should emphasize larger crystals, novel nucleation methods, containerless processing, and full characterization.

• Microgravity facilities should be treated as a national facility with its own program committee to allocate space, establish priorities, and recommend improvements.

• Additional material characterization and property measurements should be included in the program.

Ceramics and Glasses

• The cost of in-space processing probably precludes general commercialization for the near future. However, there are a few products that appear to be commercially viable at this time (for example, glass spheres and fibers).

• Gravity effects are probably less important in the processing of crystalline ceramics than for glasses or metals because of the nature of ceramic processing.

• The program is overly biased toward containerless processing and glasses.

• The ceramics and glass program is substantially underfunded; it should be increased to above the critical size.

• The Discipline Working Group should be expanded to include individuals with more ceramic insight and should include more than principal investigators.

• The basic research content of the program is too small and should be increased.

Biotechnology

• Further studies on the effects of a gravitational field on processes in mammalian cells are warranted.

• The zero-gravity protein crystallization experiments are of high interest, and the forthcoming experiments to test further the advantages of microgravity on biomolecule crystallization are appropriate.

• The media pronouncements on the commercial prospects for free-fluid electrophoretic separation processes are probably premature. However, many of the results are unavailable because of the claimed proprietary nature of the work.

• Work on electrophoretic separations and cellular processes in low gravity should be carefully evaluated by the two new university bioprocessing centers and by the National Institutes of Health in order to place them in the context of the overall national life-science program.

• Cost/benefits should be carefully evaluated along with potential environmental and safety problems that may occur in space.

Combustion Science

• Overall the program appears to be technically sound, but the necessity of microgravity conditions was not fully demonstrated.

• Novel ideas seem to be missing, and the program seems largely to consist of perturbations of existing ideas and research.

• There should be a coordinated computational effort, especially in support of reactive fluid mechanics and in zero-g modeling.

• It is recommended that additional work be started in the field of turbulence and/or turbulent combustion.

Fluid Dynamics

• Overall, this program consists of high-quality basic research on fluid physics and fluid processing that tests the fundamental laws of condensed-matter physics.

• Large-scale scientific computation is helping to resolve many of the issues in this field of study.

• The combustion program should interact more closely with the fluid-dynamics program.

• Experiments should also be conducted at high-g values, e.g., in a centrifuge.

• Additional drop-tower tests should be carried out that would lessen the need for Shuttle experiments; in some cases computer-aided mathematical analysis is preferable to space experiments.

TOPICAL ASSESSMENTS AND RECOMMENDATIONS

Metals and Alloys

Considerable progress has been made in redirecting the microgravity science programs based on the recommendations of the 1978 STAMPS report. Good examples can be found in the programs addressed at the Plenary session. However, it is our impression, based on the more comprehensive documentation provided in the Metals and Alloys Overview (see Part II, Chapter 2), that many of the programs fall short of the high standards of excellence clearly demonstrated in the two oral presentations.

Both theory and experiments relating to dendritic solidification are elegantly integrated in the work carried out at the National Bureau of Standards, Rensselaer Polytechnic Institute, and the Institute for

Theoretical Physics in Santa Barbara. The theoretical work seems to be breaking completely new ground in scaling laws and has generated widespread interest in the scientific community. This seems to be opening new doors to theoretical modeling of interface migration phenomena, which hold promise for predictive capabilities. The coupled experimental work is providing definitive tests of the theoretical predictions, and it appears that both theory and experiment are evolving in a synergistic manner.

The experimental work is particularly well done in that it addresses the problem of autonomous processing, which minimizes the need for the human element in conducting the experiments. It is our impression that the instrument design is sufficiently flexible that it can be readily adapted to perform additional experiments, for example, investigations on the effects of trace additions of solutes on dendrite morphology and growth rate and possibly on the effect of imposed thermal pulses (for example, with a laser) on the characteristics of dendrite growth. As emphasized later, it may also find utility in basic studies of fusion welding.

Another bright prospect for coupling theory and experiment is in the area of containerless processing. A good start has been made in defining the major effects of undercooling of small particulates under reproducible, controlled conditions. Extremely large undercoolings have been demonstrated in the NASA-Marshall drop tube, and the influence of critical environmental factors has been documented. Even more dramatic has been the enhanced retention of metastable phases in drop-tube tests compared with splat-quenching tests. This represents a good example of the basic science work that is a necessary prerequisite to taking the next logical step of processing in microgravity environments. The microgravity environment and high-vacuum conditions offer the potential for even more effective undercooling of freely suspended droplets and possibly even bulk samples. In this regard, it would be interesting to investigate the potential for creating shaped, bulk amorphous solids by undercooling of ready-glass-forming alloys.

These two areas of research, namely convective influences on solidification growth processes and containerless undercooling phenomena, in our judgment offer the greatest opportunities for generating new science as well as providing for technological opportunities. In contrast, we see fewer prospects for dramatic science breakthroughs in several of the other projects now under way. In particular, we see no obvious merit in the proposed investigations on solidification of complex multicomponent alloys, for example, particle-strengthened alloys, cast irons, and superalloys. Some of these projects are illustrative of the cut-and-try philosophy that generally prevailed before the invaluable input of the STAMPS committee. Since we were not fully appraised of the detailed rationales of these programs, we are not in a position to judge them. However, we recommend that an impartial committee, which truly represents the solidification community, be established to provide another reassessment of the programs in question.

With regard to the potential payoffs, there is no doubt that we will expand the knowledge base in the area of solidification science, and the programs deserve continued support on that basis. However, it is our understanding that NASA is interested in commercial aspects of this work, and this is particularly difficult to evaluate. Fundamentally we see two aspects to the problem. First, does it make sense to process in space for potential marketing on Earth? We were presented with cost estimates of the total manufacturing costs in space to be as high as $10,000 per lb of material processed. Although this is an approximate number, it gives a feeling for the product value that would be needed to justify space processing on a commercial basis. At this time, only exotic semiconductor materials that have recognized limitations in ground-based processing seem likely candidates. Second, are there any obvious incentives to develop processes in anticipation of engineering needs in space fabrication, for example, in the construction and maintenance of a space station? A good example of an opportunity area is in the joining of similar or dissimilar materials. In a high-vacuum environment, friction welding would be a strong candidate for joining, yet there appears to be nothing in the research program that addresses critical science issues concerning friction and wear, which is also relevant to continuous and reliable operation of machinery in space. Another area of interest is fusion welding, which involves complex heat and fluid-flow interactions and internal stress effects. In order to develop a good science program relevant to fusion welding one could build on the excellent start made in solidification science studies. In our view, elegant experiments could be conducted using the equipment being developed for in-situ observations of dendritic growth. For example, a hot wire or laser beam could be used to weld a surface of succinonitrile crystals. This seems to be a reasonable approach, since all the relevant thermophysical properties are well known for this model material. To add a more practical dimension to the fusion welding studies, it would be appropriate to carry out a few experiments on deep-penetration-laser or electron-beam welding of candidate materials of construction. It is quite likely that these experiments will uncover new science opportunities that have not yet been recognized. For example, does the plasma cavity spontaneously collapse as it does under 1-g conditions? We believe that this kind of work should be encouraged, whether it is done under the auspices of the MSA group or the complementary technology group. In addition to consideration of joining problems in space, attempts should be made to identify other materials-related needs relevant to maintenance and construction of space platforms and hardware.

Conclusions

Basic Science

1. A good start has been made in implementing the recommendations of the STAMPS report.

2. Theory and experimental work in solidification science have set a new standard of excellence in the field.

3. Shortcomings exist in other programs that are less firmly based on defining critical science issues or uncovering new science opportunities.

Opportunities for Industrializing

1. Near-term payoffs are clearly knowledge-based, but the new insights gained could conceivably lead to rich payoffs in the industrial sector within a few decades.

2. Materials processing in space will remain an expensive proposition for the foreseeable future.

3. Good prospects exist for near-term payoffs by concentrating on developing capabilities for construction and maintenance of space platforms, with metallurgical joining perceived as an area of real opportunity.

Recommendations

1. The areas of established excellence in solidification science should be given the highest priority for funding. If possible, an effort should be made to augment these activities.

2. An impartial committee of experts in the solidification field should be established to re-evaluate the status of other programs currently funded but that seem to fall short of the high standards of the work in solidification science.

3. Efforts should be made to bridge the gap between the science- and technology-driven microgravity programs. Areas of profitable collaboration and interaction exist in the area of materials joining.

Electronic Materials

Crystal growth is a process of importance in electronics, and optical, laser, and biological sciences and technologies, as crystals of controlled composition and specified properties are essential components in many current and prospective products. The processes operative in crystal growth are at present understood only qualitatively. The subject is in a practical sense largely empirical, for present knowledge cannot be used quantitatively to develop new processes or produce new crystals. Industries requiring crystals would benefit from a substantial improvement in the scientific understanding of crystal growth, for both single and multicomponent systems.

The use of gravity as a variable in both theory and experiment allows a systematic study of the basic mass-transport processes in crystal growth. Diffusive and convective processes are competing mechanisms for transport to the surface of the growing crystal.

Convection is more important at higher gravitational forces, and diffusion at low gravitational forces. At atmospheric pressure both processes are in general operative, and it is correspondingly difficult to characterize and study the transport processes associated with crystal growth.

The limit of purely diffusive transport can be attained at very low g. Theoretical and experimental study of this limiting case is of basic importance to the understanding of crystal growth, including interface stability, dendrite formation, eutectic structures, and multicomponent systems in general. Nevertheless, for maximum scientific understanding, g should be used as an (essentially) continuous variable, rather than a two-point (0 and 1-g) variable.

The use of zero-g crystal-growth processes should allow significant improvement in crystal characteristics. For example, with containerless processing it should allow crystals of much greater size to be produced. This may well offer considerable technological promise in the long term.

It is generally believed that the use of zero-g processes, perhaps with containerless processing, should allow crystals of much greater perfection to be produced than are possible at 1 atm. Compositional homogeneity should be attained over much smaller distances, and imperfections such as dislocations, stacking faults, and grain boundaries should be much less frequent. There are indications in the present (limited) results that this may be a general pattern. Full compositional and structural characterization should be carried out for all crystals produced at different g's, as seems to be the program intent. These characterizations should be correlated with property measurements that bear on the expected end use of crystals.

It should be noted that zero-g growth and containerless processing may allow novel scientific experiments. For example, by introduction of nucleation centers (seed sites) it should be possible to undertake controlled studies of nucleation and crystal growth. Different geometries of seeding sites would lead to polycrystalline aggregates of crystals of individually controlled orientation.

Finally, from a broader perspective, we believe that the microgravity capability should be considered as a national resource, akin to other national facilities such as the synchrotron and neutron-scattering facilities. It would be useful, in the light of this analogy, to establish a program committee that would screen and evaluate proposals as is done at the national facilities.

Recommendations

1. It is recommended that procedures be instituted to ensure close interaction between the theoretical and experimental components of the program.

2. We recommend that a greater than 1-g experimental capability be added to the program, to allow consideration of g as a continuous variable. This might best be carried out at the Microgravity Laboratory at NASA Lewis.

3. It is suggested that large crystal size be made a specific goal of the program.

4. It is recommended that crystal-growth studies be followed by full characterization and property measurements. The latter should facilitate transfer of scientific know-how to industry.

5. Novel nucleation experiments that exploit the microgravity environment and containerless processing capabilities should be considered.

6. It is suggested that a program committee be established for the entire microgravity program.

Ceramics and Glasses

Of the three attributes of space (microgravity, low pressure/high vacuum, and unlimited volume), the area most difficult to achieve in ground-based research is that of microgravity, at least for extended periods of time, i.e., in excess of a few seconds. This, plus the fact that many ceramics and glass processing operations are strongly dependent on gravity effects, such as the fining of glass, casting processes, and crystal nucleation and growth, argue strongly for the investigation of phenomena under microgravity conditions to aid in the fundamental understanding of these processes. In the case of glass and ceramics research, the increase in fundamental understanding appears to be the paramount justification for in-space, microgravity experiments at present and for the foreseeable future. The primary objective is to obtain an improved understanding of gravity effects in glass and ceramics processing for virtually all glass and ceramic materials. The cost of in-space processing ($10,000-$250,000/lb--workshop estimates) may preclude the possibility of commercial in-space processing for the immediate future.

At present, there is an aura of excitement about new applications of ceramics and glasses such as heat engine components and fiber-optic communications. Consequently, there is a great deal of research related to the processing and properties of materials for these applications. In particular, ceramic powder processing is receiving a great deal of renewed research effort and development activity. Therefore, one must ask how can microgravity in space processing contribute to improved ground-based processing of ceramics and glasses? There will be some discussion of this below under Program Content. However, the point can be made that gravity effects, in general, are probably less important in the processing of crystalline ceramics than either glasses or metals since the newer, higher-technology ceramics are produced by all solid-state processes in which gravity and convective flows play a minimal role. Nevertheless, there are areas in which microgravity processing can be employed to develop better fundamental understanding.

Finally, many of the projects included under the Metals and Alloys Program are really generic in character and can be applied to the processing of ceramics and glass as well. Examples are the theoretical and experimental studies on the nucleation and interface stability of growing crystals and Ostwald ripening of immiscible metallic liquid phases. The only reason that they are included in that program rather than in the Ceramics and Glass Program is that the program was proposed by metallurgists, who, in general, are working with metallic systems. However, such generic studies could equally well have been performed in appropriate ceramic systems and, therefore, might be considered to be related to the Ceramics and Glass Program.

Program Content

The Ceramics and Glass Program actually contains much that is not really basic research on ceramics and glass. In some regards, the program could have been labeled Containerless Processing, since it appears that a substantial portion of the program is actually devoted to the engineering details of the acoustic levitation apparatus and techniques for making spherical hollow shells. It would seem that the latter might also be included under Fluid Mechanics. As a result, the content of scientific basic research devoted to ceramics and glass is rather minimal, perhaps even too small to be of critical nucleus size. There simply are not enough scientists working in the area to foster adequate communication with each other to achieve a fruitful level of interchange. Furthermore, the program, such as it is currently constituted, is heavily biased toward containerless processing and glasses.

The reasons for this are not obvious. Certainly, the program does not have a great deal of visibility in the ceramics community. As a result, the ceramics proposal pressure may not be sufficient to produce a larger and more diverse high-quality program. On the other hand, there simply may not be many areas in ceramics processing, other than those in glasses, for which gravity effects are important or in which the absence of gravity might be used to better understand the fundamentals of processing. That this need not be the case can be illustrated by the following example. There are many efforts under way to produce small, monodisperse ceramic powders by gas-phase reactions. Clearly, such reactions are strongly convection dependent, and the fundamental mechanisms of reaction kinetics and particle growth could ideally be studied in a microgravity environment. Another point worth noting is that the Discipline Working Group in this area seems to have just begun to focus its activities whereas those in some of the other areas appear to be more highly focused.

In summary, the following features of the Ceramics and Glass Program appear evident to the reviewers: (a) There is virtually no basic ceramics research in the program. (b) Much that is included under the program is really not glass and ceramics research. (c) It is biased toward glass and containerless processing. Experiments have not been so strongly focused on microgravity-controlled processes as may be desirable.

(d) The program lacks visibility in much of the ceramic community and appears to need stronger direction and more focused organization.

Quality of Science

Based on the presentations and the information provided, the quality of the science within the Ceramics and Glass Program is extremely difficult to judge. The formal presentations addressed the technical detail for only a small portion of the program, namely that related to the containerless processing of glasses to study crystallization kinetics. To these reviewers, it is difficult to justify the microgravity studies to address only containerless-processing experiments for nucleation studies. Cleverly designed ground-based experiments may accomplish some of the same objectives. The sol-gel studies are excellent, but many researchers are doing sol-gel processing of glass and ceramics, and it is not clear why this program, with its very limited budget, should also continue to study sol-gel processing. The bubble and surface-tension studies are conceptually quite sound but could not be evaluated. Perhaps a better understanding of bubbles in glass will be one of the promising areas of research. Certainly it has tremendous implications for the fining of earth-processed glasses.

The ceramics and glass program needs more funding, visibility, guidance, breadth and should not be used as an engineering vehicle for testing containerless systems.

Recommendations

1. Separate the real ceramics and glass research from generic instrument development. Interact with metals and alloys efforts where appropriate.

2. There needs to be an increase in the funding for ceramics and glass research, because the present program is below the critical size. This should be done only if high-quality research programs can be identified.

3. Expand the ground-based program and obtain more definitive results prior to in-flight experiments to prevent perceived high failure rate.

4. Increase the membership of the Discipline Working Group to include members with more ceramics insight. It should include more than the Principal Investigators. This group should establish priorities for the basic problems and concepts to be studied.

5. Make the program more visible to the ceramics community to increase proposal pressure and broaden the program.

6. Reduce the tendency to fly experiments prematurely.

Biotechnology

As the NASA manned spaceflight program moves into the era of routine Shuttle missions and permanent space stations, NASA activities in life sciences and applications are changing from being almost totally occupied with space medicine (health and safety of man in space; accommodation to zero gravity and readjustment to unit gravity) to having a substantial complement of experimentation on the cellular and physiologic basis for human response to zero gravity, on cellular interactions with the gravitational field, and on applications of microgravity in biotechnology. The great increase in opportunities for experimentation afforded by the Shuttle mid-deck and Spacelab, the ad hoc nature of almost all previous life-sciences activities aboard spacecraft, coupled with a substantial improvement in experiment planning and support studies on the ground, suggest that life-sciences research in space is really just beginning.

This workshop addressed the life-sciences activities with the Microgravity Science and Applications Program of NASA. The life-sciences segment of the program contains three components of particular interest and high level of activity: (1) studies of effects of microgravity on cellular and physiologic processes, (2) investigations of the utility of a microgravity environment to enhance crystallization of biomolecules, and (3) investigations of the utility of a microgravity environment to enhance separations of biomolecules, organelles, and cells. The number of experiments concerned with these topics carried out in microgravity has so far been relatively small.

Two sets of experiments carried out recently in conjunction with Shuttle flights have indicated large dependences of some important processes in cultured mammalian cells on the gravitation field, which may have implications for space medicine as well as for understanding control mechanisms in cellular biology. In one set of apparently well-controlled studies, Cogoli and Tschopp observed the virtual disappearance in the microgravity environment of the well-characterized in vitro stimulation of human T lymphocyte division by concanavalin A. Additional observations in ground-based work of a monotone increase in ConA-stimulated lymphocyte activation at $g > 1$ compared with $g = 1$ appear to confirm the gravitational-field dependence. In another study, Grindeland and Hymer observed the virtual shutdown (<5 percent) of growth hormone production in cultured rat pituitary cells kept near 0 g in space compared with controls at 1 g. At the same time, production of prolactin, another hormone, was similar for the two treatment groups. It has also been claimed that Russian workers have observed a 20-fold increase in interferon production by human leucocytes during spaceflight.

These apparent effects of gravity-field alteration on cellular function require and deserve further study. Several features can be ascribed to such studies at this time. The microgravity portion of the experiments is relatively simple, has low volume requirement, and is, therefore, not an expensive addition to, for example, a Shuttle mission. The Cogoli and Tschopp results indicate that it may be possible to screen for gravity effects by performing $g > 1$ experiments on the ground. Finally, at this time there are no well-founded working hypotheses to guide mechanistic inquiries. However, expanded cataloging and characterization of cellular processes as a function of the gravity field seem warranted, with special attention paid to controlling all other conditions. Besides possibilities for new understanding of control mechanisms in cellular biology, such studies may reveal clues about some observed human responses to reduced gravity. The T lymphocyte observations may have implications for immunologic studies.

It has been suggested that, in one standard approach to protein crystallization, crystal growth is limited by movement of growing crystals out of the region of fastest growth because of sedimentation or flotation.

In confirmation of this model experiments on Spacelab 1 demonstrated the production of larger, more perfect crystals of two proteins, lysozyme and p-galactosidase, compared with crystals grown at 1 g. Several groups of workers will be flying experiments mid-deck on forthcoming Shuttle missions in order to test the scope of improvement in biomolecule crystallization in microgravity.

The three-dimensional structures of biomolecules provided by x-ray crystallographic techniques have been seminal contributions to molecular biology and the central question of structure-function relationships. Indeed, the approach continues to yield information of fundamental importance, for example, the recent discovery of a novel structural form of DNA by Rich and co-workers. With modern diffractometers and computers, x-ray crystallography has reached the point at which, given sufficiently good crystals of appropriate size, quite large structures can be solved rather quickly. In recent years, advances in molecular understanding and in computer-aided graphical techniques have facilitated the design of small molecules for binding to specific sites on proteins and nucleic acids. Given the structure of the appropriate macromolecule, compounds can be designed to mimic, for example, enzyme substrates or hormones. Many pharmaceutical companies are taking this approach to drug design.

Since the limitation in x-ray crystallographic structure determination is often the availability of suitable crystals, the zero-gravity protein crystallization experiments have apparently evoked excitement in both academic and industrial circles. The forthcoming experiments designed to test the scope of the advantage of a microgravity environment on biomolecule crystallization are, therefore, of high interest. Should these studies demonstrate a general utility for microgravity in crystallization, it is not out of the question that some sort of microgravity crystallization service venture would evolve. At this time, however, it is not possible to predict the commercial viability of such an enterprise. It should be pointed out that biomolecular crystallizations can be carried out in an automated fashion within Shuttle mid-deck lockers so that they are relatively inexpensive procedures.

The enhancement of free-fluid-based separation methods for purification of biological materials in the microgravity environment of orbiting spacecraft has been the area of biotechnology that has been most active in the NASA MSA Program. The main reasons for this are (1) the perception that commercial-scale purification of the products of genetic engineering especially for human medical use, is an unsolved problem for which microgravity enhancement may well be the solution; and (2) the substantial activity that the McDonnell-Douglas Astronautics Corporation has been carrying out in conjunction with Shuttle missions, in part via a joint endeavor agreement with NASA. These activities in biomaterial separation technology development have received a great deal of media coverage replete with hyperbole of the sort "new medicines produced in space." This kind of premature pronouncement has provided some of the reason that a good deal of controversy surrounds these activities.

To gain some perspective, first consider the purification of some human polypeptide (protein) hormone produced by a bacterium into which the gene for the hormone was incorporated by recombinant DNA techniques. Assuming that the bacterium does not "secrete" the hormone, one ends up having to separate the desired hormone from the other several thousand proteins and other compounds contained in the bacteria. Besides the challenge provided by the necessity for high purity for products for human in vivo use, other difficulties are provided by the desired fermentation volume, typically 1000 gallons, and the need to achieve rapid separation of desired protein products from enzymes present in the mix that can destroy or modify the products. While a host of separation methods have been worked out over the years for both analytical purposes and for laboratory-scale preparations of proteins and other biological macromolecules, none of them are without problems at commercial scale-up levels. While problems of high costs are most common, there are also many technical problems associated with scale-up. Consequently there is at present a great deal of activity associated with improvement of commercial-scale biochemical separation procedures. Substantial progress has been made in scale-up of gel-filtration chromatography and of high-performance liquid chromatography (HPLC), especially affinity HPLC. Such techniques have made it eventually possible to purify efficiently all sorts of proteins and protein complexes at the laboratory scale, and there is every reason to believe that the same approaches will succeed at the commercial scale after further development.

While gel electrophoresis has no peer with respect to resolution of biomolecules for analytical purposes, electrophoretic methods become difficult at the laboratory preparative scale, and no such approach appears to be in use at the commercial scale. Convective flows in free-fluid electrophoretic procedures become more of a nuisance as the volume is increased because cooling becomes less efficient. The concentration of material to be separated is, of course, very much higher for preparative purposes compared with analytical purposes. At such high concentration, zone sedimentation of materials in regions of density instability is a common problem. Both convective flow and zone sedimentation should be absent in a microgravity environment, and it was hypothesized that preparative-scale free-fluid electrophoretic techniques would exhibit tremendously increased effectiveness in microgravity. A variety of experiments carried on by different groups of workers and flown on Apollo-Soyuz, Skylab, and several Shuttle missions (STS-4 through STS-8) have verified that free-fluid electrophoretic methods can be more effective in the microgravity environment. In one recent experiment of the McDonnell-Douglas group it was claimed that a 730-fold increase in throughput over that possible at unit gravity was achieved for the separation of ovalbumin and rat serum albumin in continuous-flow electrophoresis, a particularly convenient electrophoretic method. The enhancement was due to the use of a severalfold larger chamber, possible because of the absence of convection and the use of protein concentrations about 100-fold higher than those normally used at unit gravity. In collaboration with Ortho Pharmaceuticals (a subsidiary of Johnson & Johnson) the McDonnell-Douglas group has also made at least one trial purification of protein product of pharmaceutical interest. Because of the proprietary nature of the work no details were made available. It was reported, however, that the product had become "contaminated" at some point, and quality testing was uncertain.

Separations of subpopulations of cells is of great importance for research, for clinical purposes, and, with the advent of the newer biotechnologies, also for commercial purposes. Electrophoresis in microgravity should allow separations based primarily on the charge properties of the cells without interference from sedimentation. Several experiments aboard spacecraft have indicated that this conjecture holds true. However, there is yet no clear demonstration that the microgravity techniques offer a distinct advantage over procedures at unit gravity.

While it is clear that the microgravity environment can greatly enhance certain preparative-scale separations by electrophoretic methods, it is not at all clear at this time that this approach can compete with alternative ground-based technologies. Many important questions remain with respect to the logistics and economics of routine commercial use of such an approach. At this time it is certainly not possible to use the biotechnology activities at microgravity as a major justification for the U.S. space program. The question now is, where do we go from here? The NASA life-sciences programs have been criticized in the past for a number of reasons including insufficient review by peers outside microgravity research. There is currently a Microgravity Biotechnology Discipline Working Group within the Universities Space Research Association that performs some program assessment. Outside reviewers are used both for program evaluation and for review of research proposals, of Principal Investigators, and of technical feasibility of projects. However, the Discipline Working Group may not be adequate to assess the relationship of the program and of particular projects with the whole of biotechnology and biomedical research. The two university bioprocessing centers recently mandated by Congress (University of Arizona and University City Science Center, Philadelphia) are charged, among other things, with evaluation of microgravity biotechnologies in comparison with unit gravity technologies. This structure should provide significant further assurance that the money being spent in the microgravity program is not better spent in other biological and biomedical areas. One wonders why NASA does not also commission the National Institutes of Health to evaluate the program and to suggest areas of needed research. This would serve two purposes: the microgravity program would be understood in terms of the overall life-sciences objectives, and much apprehension in the academic life-sciences community about competing for federal research funds with the relatively expensive microgravity program might be allayed.

Much of the microgravity electrophoresis work is being carried out by McDonnell Douglas Astronautics under the aegis of a joint endeavor agreement with NASA. As some point in the program there needs to be some sort of cost/benefit accounting both because of the importance being attached to the electrophoresis program and the criticisms that have been voiced. It has been stated by one critic that the total cost to the U.S. taxpayer of all the microgravity electrophoresis work exceeds by far the government monies that supported the development of the artificial heart or of magnetic resonance imaging.

Design plans are fast being put together for a permanent space station in part justified by a prediction of commercially viable microgravity materials processing. It is imperative at this point that NASA carefully evaluate the environmental needs, outside of microgravity, for any biotechnologies contemplated, especially as pertains to product contamination, apparatus recycling, worker safety, and turnaround times. These are difficult issues on the ground and would appear to be a major challenge for a closed system 500 miles from the surface of the Earth.

Combustion Science

The combustion-science component of the program is somewhat at a disadvantage with respect to the other five components reviewed because there have been, as yet, no space experiments dealing with combustion research. In several of the other areas, notably that dealing with crystal growth, past experiments under microgravity conditions have provided new and exciting results, and current research is directed appropriately at understanding the new and often unexpected data. In contrast, in the combustion-science area the researchers can still only speculate. Furthermore, combustion represents such a broad and complex subject that the limited support currently available can only serve to scratch the surface of the topic.

The reviewers noted several central issues in combustion science that await resolution through microgravity combustion research:

1. Premixed gaseous flame propagation and excitation;
2. Premixed particulate cloud-flame propagation and extinction;
3. Smoldering and transition to open flaming;
4. Combustion of individual drops and particles;

5. Radiative ignition of condensed-phase fuels;
6. Large surface combustion and extinction;
7. Autoignition in premixed gaseous media.

Important to each of these issues is the role played by buoyancy. It was argued convincingly that natural convection, arising from the buoyancy of hot combustion products, clearly plays an important role at g = 1. The combustion-science program projects seem to favor an approach of simply trying a lot of known combustion experiments in the absence of gravity. This way individual mechanisms can be isolated and studied in simpler geometries with fewer wall effects.

This approach is not unreasonable, but it also could lead nowhere. The importance of the lack of natural convection may be overstated. In most practical combustion environments, natural convection is already much less important than forced convection, driven either by the expansion of the hot combustion products or by air circulation. Therefore, in some combustion problems natural convection is often neglected, and the opportunity to carry out experiments in zero gravity would be of little benefit. From the material presented at the review session, there appears to have been relatively little pre-analysis of the influence of low gravity.

Computational fluid dynamics and reactive flow modeling have advanced to the point where many of the technical issues can and should be treated computationally. Most models can assume g = 0 more easily and accurately than they can g = 1. In the final analysis we can only claim to understand a given physical and chemical mechanism quantitatively after we demonstrate that we can compute its behavior, so modeling will be needed eventually in any case. We did see a considerable amount of analysis during the flame-spread technical presentation, using primarily asymptotic methods, but no computer modeling efforts were discussed. It was unclear whether this was not discussed because of a lack of time or if such modeling is not a part of the program.

Fire and combustion are multidimensional reactive flow problems, a field in which computational physics techniques have grown rapidly in recent years. The presentations at this workshop suggest that it would be advisable to enhance the interaction between the combustion-science component and the fluid-mechanics component of NASA's microgravity science and applications program. Perhaps the program office should look into providing the program participants access to Class VI supercomputers. At present, the lack of a coordinated modeling component is seen as a significant limitation and weakness of the combustion-science component of the overall microgravity program.

The overall program review itself was quite general, rather vague, and nonspecific. The material that the panel was given to work with in forming this assessment was limited and probably does not do justice to the program. Certainly the records of the seven Principal Investigators are superior, but a discussion of their programs and the role that microgravity is playing in their research would have been most beneficial to our review process. Of the subjects summarized, a good case was not made for the study of cool flames in space, nor were good reasons (i.e., a possible mechanism or predictions based on numerical modeling) given for why g = 0 laminar flame propagation is any different from downward flame propagation at g = 1. However, it was clear that for nonplanar geometries, extended periods of microgravity seem indicated to study slow phenomena such as smoldering, flame propagation near the flammability limits, and minimum ignition energies. For example, ignition due to the absorption of radiation appears to be one area where enormous differences can be expected owing to the lack of buoyant convection. At conditions of g = 0 with stagnant air, a weak light bulb may be enough to start a fire, a result that is very different from that encountered with free convection. These and other specific topic areas were presented clearly, and the case for investigating them in microgravity conditions was made well. From the titles of the other research programs not summarized at the workshop, it was assumed that most of them fell into the category of trying well-characterized earthbound experiments in a zero-g environment. The presentation on flame spread in zero g was effective, leading us by implication to believe that the technical content of the entire program is in good shape.

However, novel ideas seem to be missing, and the program seems largely to consist of perturbations of existing ideas and research. Separating physical mechanisms and simplifying complex problems like those encountered in combustion is almost always a good idea, and the ability to eliminate buoyancy effects in zero g will provide valuable insights, but it is somewhat disappointing not to see a venturesome component in the program somewhere.

Fire safety in space was indicated as one major application area, and the reviewers tend to agree with this assessment. Fire in space creates some new problems, and rules of thumb applicable at g = 1 may be incorrect in space. However, the fire-safety problem in space has some aspects that are similar to those encountered in other limited living space environments such as submarines. We would encourage those concerned with fire safety in microgravity environments to become familiar with current developments in these areas, which, at first glance, might appear to be unrelated to space conditions.

Since turbulent combustion is of such importance in both current theoretical research and practical applications, we suggest that this topic be considered for possible inclusion into the microgravity programs. Specifically, this would include variable-density turbulence studies, both with and without reactions. Turbulent flows are difficult to study convincingly with existing fluid mechanics computer models, and the effects of buoyant convection complicates earthbound experiments, so the potential benefits of turbulence studies in microgravity conditions are substantial. Most important, since turbulent combustion is a dominant process in all practical combustion systems, the eventual economic benefits of anything that is learned would be enormous. This is, however, a long-range goal. Still, we believe that this is such an important combustion field that it should be represented in the microgravity research program.

The program appears, from the limited material presented, to stress basic processes and fundamental knowledge at the expense of applied or potentially commercial projects. We believe that this emphasis on fundamental science is correct at present in the case of combustion science. Knowledge gained in space is likely to have a major impact on processes on Earth without the high and recurring cost of space application or processing. A fraction-of-a-percent improvement in combustion efficiency realized as a result of microgravity research would be worth billions of dollars each year, far more than we can foresee resulting from the implementation of some combustion process that can be carried out only under microgravity conditions.

In conclusion, we found it difficult to assess the quality of the program, other than the specific research of the investigators who were present at the workshop. We would encourage the person presenting the overall review at future workshops to present a more complete survey of what other researchers are doing. Overall, we believe that the program is technically sound and likely to provide useful and interesting material on combustion. However, we also believe that the necessity of microgravity conditions was not fully demonstrated. Of course, until some actual experiments are carried out in space under microgravity conditions, the conservative approach being followed is probably satisfactory. We believe that the lack of a coordinated computational effort, especially in support of the reactive fluid-mechanics parts of the program, is a serious problem and should be addressed as soon as possible. With a field as broad as combustion it is impossible for a small number of programs to cover everything, and it would be inappropriate for NASA to attempt to do so. The range of technical areas spanned by the existing investigators is reasonable for an effort of this size. However, because of its importance in both applied and theoretical combustion, we recommend that an activity in the field of turbulence and/or turbulent combustion be considered as a worthwhile and appropriate addition to the overall effort.

Fluid Dynamics

The following conclusions and impressions are based on the presentations by Labus and Lipa in the session on fluid dynamics or on related presentations in earlier sessions and on our own independent knowledge of various researchers engaged in the fluid-dynamics program. Overall we find that this program consists of high-quality basic research in aspects of fluid physics and fluid processing. Although nothing in the program has obvious commercial value, the researches supported seem likely to lead to better understanding of various process mechanisms in the light of which terrestrial processes can be improved or developed.

A major part of the research activity in the program can be characterized as the testing of the fundamental laws of condensed-matter physics. Examples are attempts to determine experimentally critical exponents of the heat capacity of helium at its lambda point and of the transport coefficients at an ordinary critical point, ellipsiometric studies of adsorption and wetting transitions, and investigations of the physical meaning of the failure of the classical law of capillary hydrostatics to admit the existence of free surfaces under certain conditions. Microgravity experiments should play a crucial role in these investigations.

The results of the lambda-point and critical-point studies will either increase our confidence in the universality of the scaling laws, predicted by modern theory (the renormalization group) or challenge theoreticians to develop a new modified theory of phase transitions in condensed matter. The high-resolution thermometer developed to study the lambda transition is an experimental tour-de-force and will most likely provide new capability in studying other significant scientific problems such as finite system-size scaling and, tricritical-point behavior in multicomponent fluids. Such spinoff is a favorable measure of high-quality science. The research on free surfaces will probably lead to a better understanding of the limitations and necessary extensions of the classical laws of capillarity.

Process analysis is the second main type of research activity supported by the fluid-dynamics program. Here no new laws of physics are likely to be invoked. What is entailed, rather, is the identification of dominant physical mechanisms, the choice of the appropriate physical principles to use in analyzing a particular process, and the development of the mathematical and computational machinery needed to understand the basic phenomena occurring in the real world of two- and three-dimensional geometries and nonlinear processes. Examples of research topics in this area are gravitationally driven fluid convection in crystal growth phenomena and interfacial tension-driven flows. Large-scale scientific computation can resolve many of the issues posed in this area. (Examples of commercially available, improved crystal-growing furnaces designed with the aid of insights provided by NASA-supported modeling of solidification processes were cited.) However, microgravity experiments will be useful to verify the results of the complicated calculations, to give empirical insights into real systems whose geometries and/or processes cannot yet be handled by state-of-the-art supercomputers, and actually to produce microgravitationally grown crystals should they be desired. One use that seems promising (and of possible commercial interest) is to produce good crystals of large molecules, such as proteins, for analytical purposes. The research on interfacial tension-driven flows is especially important, since, at low gravity, tension becomes a primary mechanism of fluid concentration gradients.

To the extent that we can judge (together we are familiar with more than half of the investigators and their projects listed by Labus in his overview) the Principal Investigators in the fluid-dynamics program are first-class researchers and are carrying out high-quality research on the NASA projects. Most research is reasonable in the context of the microgravity sciences, although, for example, the surface-tension work is unlikely to profit directly from microgravity capabilities and such generically labeled projects as "transport processes research," "mass transport phenomena," and "geophysics fluid flow" might well lack focus on the questions that microgravity experiments can best answer.

It appears that for the most part there is reasonably good interaction among the groups studying problems with overlap or common elements. An exception to this may be the combustion groups, which could probably profit from closer interaction with those studying convective fluid flow in crystallization and drop-dynamics processes.

Finally, it is worthwhile to raise a few questions that we feel should be considered in guiding future research in the fluids-dynamics program.

Questions

1. For which gravitationally related problems would <u>increasing</u> g, with the aid of centrifuge, prove appropriate in gaining insight before, as a supplement to, or even in place of microgravity experiments?

2. How many of the significant microgravity issues can be essentially resolved by studying matched density systems or by doing drop-tower experiments (perhaps in an expanded drop-tower facility) instead of on the Space Shuttle?

3. Where theory is well understood, when are space experiments to be preferred to appropriate computer-aided mathematical analysis?

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II
Workshop Proceedings

AGENDA

WORKSHOP ON MICROGRAVITY SCIENCE AND APPLICATIONS
December 3-4, 1984

First Day

SESSION I		
Opening Session		Brian Frost, Cochairman
0800	Welcome	Robert Parks, JPL Director
0815	Introduction and Workshop Objectives	Al Schindler, Cochairman
0830	General Program Philosophy and Commercialization Opportunities	Richard Halpern
0930	Discussion	Arden Bement
1000	Break	
SESSION II		
Metals and Alloys		William Nix, Chairman
1015	Overview	Robert Naumann
1115	Theory	Sam Coriell
1145	Dendritic Growth Studies	Martin Glicksman
1215	Discussion	Bernard Kear
1245	LUNCH	
SESSION III		
Electronic Materials Session		Herb Johnson, Chairman
1345	Overview	Roger Crouch
1445	Fluid Flow Modeling	Robert Brown
1515	Crystal Growth in Space	Harry Weidemeier
1545	Discussion	Praveen Chaudhari
1615	Break	
SESSION IV		
Glasses and Ceramics		Dennis Ready, Chairman
1630	Overview	Robert Doremus

1730	Spherical Shell Technology	Dan Elleman, Taylor Wang
1800	Discussion	Richard Bradt
1830	ADJOURN	

Second Day

SESSION V
Biotechnology — Angelo Lamola, Chairman

0830	Overview	Dennis Morrison
0930	Separation of Growth Hormone	Wes Hymer
1000	Discussion	Norman Anderson
1030	Break	

SESSION VI
Combustion Science — Jay Boris, Chairman

1045	Overview and Theory	Abe Berland
1145	Flame Spreading	R. Altenkirsch
1215	Discussion	C. Westbrook
1245	LUNCH	

SESSION VII
Fluid Dynamics — Michael Fisher, Chairman

1345	Overview	Tom Labus
1445	Specific Heat of Helium, Theory	John Lipa
1530	Discussion	Ted Davis

OPEN SESSION

1600	Panel Discussion	Al Schindler, Brian Frost Panel Cochairmen
1700	ADJOURN	

CLOSED SESSION

1700	Panel Meeting
1830	Working Dinner
2030	Panel Meeting Adjourns

1. GENERAL PROGRAM PHILOSOPHY AND COMMERCIALIZATION OPPORTUNITIES

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INTRODUCTION

The purpose of this paper is to present an overview of the current program at the National Aeronautics and Space Administration (NASA) in microgravity science and applications. However, before presenting the current program, it is important for the reader to become familiar with the background from which the current program sprang.

The Microgravity Science and Applications Division is an outgrowth of the former Materials Processing in Space (MPS) activity at NASA. This activity began in the early 1970s and might be characterized as a haphazard, shotgun approach to experimentation. Early experiments were carried out on the Apollo program in electrophoresis, crystal growth, and solidification. These activities continued through the Apollo-Soyuz era. Unsubstantiated claims, talk of building factories in space, making better ball bearings, and so on, led to a National Academy of Sciences review in 1977.

This review on the Scientific and Technological Aspects of Materials Processing in Space (STAMPS) reported in 1978 that "...to date, the NASA program ... has been weak." Scientific and technological results [from these missions] were sometimes shallow, incomplete, or inconclusive." Perhaps the one exception taken by the Committee was in the area of combustion, where the Committee indicated that a strong program was taking shape and that NASA should support this activity. The report also emphasized that NASA should undertake for "approximately the next five (5) years" a strong program of ground research to better understand the parameters of the experiments that would ultimately go into space.

The result of this report was that NASA instituted a change in the management of the Materials Processing in Space Program and encouraged the new management to follow the recommendations of the Committee. A ground research program involving scientists at universities, as well as within the NASA organization, was undertaken. Experiments in microgravity began using drop tubes and drop towers as well as airplanes flying Keplerian trajectories. These activities provided from 4 to approximately 20 sec worth of zero-g time and enabled researchers to study small sample results while pursuing model and parameter activities. A program of sounding-rocket flights was also instituted, which provided approximately 5 minutes of microgravity time for experiments. In addition to the scientific results from these early experiments, engineering tests of flight hardware, particularly in the area of containerless processing, were facilitated. It should also be remembered that in 1978, we were being told that the Space Shuttle would be available for experimental use in 1980. This resulted in an unfortunate and premature desire to develop sophisticated and expensive generic hardware. Since, as we know, the Shuttle did not become available until much later, yearly budget cuts were instituted that ate into both the hardware preparation and the ground research program.

The current program is, then, the outgrowth of these early years of activity. The Shuttle is available for experimental use, and it is my belief that the 6 years between 1978 and 1984 have been used prudently in preparation for the forthcoming activity. It must be noted, however, that the Shuttle is not yet truly operational, and the number of experiments actually flown to date is small.

Having provided the background, it is appropriate to look at the goals and objectives of the Microgravity Science and Applications Division's program. Simply stated, the objective of the program is to facilitate the use of the microgravity environment of space in order to better understand basic phenomena of physics and chemistry, as well as of materials and material processing. It is our further objective to apply this basic research in order to develop practical applied usage. This objective has three goals: first, the goal of knowledge of phenomena and how these phenomena differ in the microgravity environment as compared with the unit-gravity environment. Second, to produce new and novel products, unique to and requiring zero g. Third, to provide experimental information to enable changes in processes on Earth and, thus, to provide more efficient and economically viable applications.

NASA Headquarters provides the overall direction and guidance for the Microgravity Science and Applications Program. Implementation of the program is carried out by five NASA Centers: the Langley Research Center, the Lewis Research Center, the Jet Propulsion Laboratory, the Johnson Space Center, and the Marshall Space Flight Center. Activities in both basic and applied research are carried out at these locations. At the same time, research activities that involve university scientists, as well as industrial researchers, are monitored through these Centers after selection by peer review through NASA Headquarters.

The Division has established Centers of Excellence at various locations throughout the United States that provide focus for certain types of research. These Centers include the Materials Processing Center at the Massachusetts Institute of Technology (MIT), a theoretical focus for the program at the Institute of Theoretical Physics (ITP) in Santa Barbara, a program of fundamental theory and measurements at the National Bureau of Standards (NBS), and finally, two new Centers in biotechnology, one at the University of Arizona and a second in Philadelphia at the University City Science Center, representing a consortium of 12 universities in the Pennsylvania area.

The approach that the Division is taking starts with suggestions of new ideas and concepts from multiple sources, including our own in-house NASA research work, university researchers, and industrial researchers. On approval of the concept, ground-based activities are undertaken and flight instrumentation development begins only at that time when a researcher believes that sufficient information is available to justify the expenditure for flight equipment. Results from flights may either prove or disprove certain theoretical work that has been conducted and, of course, may lead (and is almost expected to lead) to new experiments. The Shuttle allows us a continuing program of experiment parametric changes and reflight of equipment. This is the analog of the laboratory activity of many material scientists on Earth.

One of our current issues, however, is the infrequent opportunity to experiment in space with Shuttle payloads. More will be said about this later. From these space and ground research activities may come areas that are commercially interesting. I must emphasize the use of the word "may" because it needs to be made explicitly clear that NASA is a research and development agency, and we expect that industrial interests will decide from the results of our research what is commercially viable, and not the other way around.

THE GROUND RESEARCH PROGRAM

This workshop is being held to review in some detail NASA's current scientific base in the area of fundamental physics and chemistry investigations as well as in materials and materials processing. Therefore, this paper will not dwell at great length on this area; however, there are several points that should be brought out as part of the overview. The first is that we are currently working (as noted above) at 5 NASA Centers and, in addition, at more than 50 universities throughout the United States. All the proposals for research are peer reviewed for scientific significance, relevance to the need of the microgravity environment, and the desire of the investigator for eventual spaceflight. Budgets and engineering feasibility are also examined prior to grant approval.

We have established six working groups to help focus the research activities of this Division. These six groups are Metals and Alloys, Glasses and Ceramics, Biotechnology, Electronic Materials, Combustion Science, and Fluid Dynamics and Transport Phenomena. The purpose of these groups includes, but is not limited to, the evaluation of the specific research areas and determination of the rationale for microgravity experiments; the incorporation of fundamental experiments in both physics and chemistry into the total program; and, the establishment of priorities for fundamental issues within each discipline. Membership on the working groups includes researchers from government, from universities, and from industry. The working groups have been deliberately kept small in order to provide an environment conducive to hard decisions and recommendations. It is my plan to establish, within the next year, an oversight committee of senior members to help guide the total Division activity.

In addition to these working groups reporting directly to the Division, the Division receives critical review from an internal NASA advisory board known as the Space Applications Advisory Committee (SAAC), which has established a Microgravity Science and Applications Subcommittee headed by Robert Sekerka of the Carnegie-Mellon University.

The Space Science Board of the National Research Council is currently reviewing a request from the Administrator of NASA to establish a subcommittee of that Board also to provide oversight.

Finally, in the area of ground research, it should be noted that we have available and are currently using drop-tube facilities and drop-tower facilities at the Jet Propulsion Laboratory. Additionally, we are providing low-gravity experimental time aboard the KC-135 airplane, as well as a Learjet, which is available at the Lewis Research Center.

THE FLIGHT PROGRAM

Having once satisfactorily completed a ground research program, an experimenter proposes an experiment for spaceflight. A wide variety of instrumentation, largely based on sounding-rocket technology, is currently available for use by the researchers. These instruments include general-purpose furnaces, acoustic levitators, directional solidification furnaces, and electrophoresis equipment, both for continuous flow and isoelectric focusing. Additionally, an experimenter may propose the building of unique instruments in order to provide the experimental data desired. This latter situation is largely true in the areas of combustion science and fundamental experimentation in physics and chemistry, as other papers in this volume will describe. We also have under consideration a second-generation set of equipment that has been proposed for development through the normal proposal route.

The current flight program is based on the use of the Space Transportation System (Shuttle). The Shuttle provides the capability of performing experiments in an environment of approximately 10^{-4} g for periods up to 10 days. Within the Shuttle, several different locations, each with a different capability for experimentation, exist. One of these locations is called the Middeck Locker Facility. Experiments of approximately 10 X 17 X 20 in^3 and weighing up to 60 lb can be accommodated. Additionally, larger multiples can be accommodated through the use of adapter plates. The Middeck Facility is characterized by easy installation of the equipment, access to the equipment up to a day before launch, and a very simple interface. Conversely, however, the Middeck has available only about 300 W of power, and cooling is either by the cabin air or by special cooling apparatus within the equipment.

Larger equipment can be carried in the Shuttle Orbiter Bay; here several options are available. The first and simplest is the use of a Materials Experiment Assembly (MEA), which is a self-contained housing capable of carrying three experiments 17 in. in diameter, 36 in. in length, and weighing a maximum of 200 lb each. The MEA provides 32 kWh of energy through the use of batteries and can reject up to 1000 W of heat. The MEA was successfully flown as part of the STS-7 payload in the summer of 1983 and will be flown again as soon as possible.

A more advanced carrier, known as the Materials Science Laboratory, is currently being built and is expected to be flown in 1985. This carrier will provide 103 kWh of energy by plugging into the Orbiter power directly. It will have active cooling available to it, its own recording system, as well as providing a standard interface for experiments. It will be capable of carrying a total payload weight of some 3000 lb, which may be divided into three instruments or any combination thereof. A second version of this carrier is currently being planned to allow more frequent flight opportunities starting in 1987 and through the advent of the Space Station.

Another option available to experimenters is through the use of the Spacelab equipment, either within the module or on pallets that require automatic equipment. Spacelab-1, which flew in November 1983, carried a variety of materials processing experimentation provided by the European Space Agency. Spacelab-3 flew in April 1985, and carried a number of U.S. instruments that allowed research in the area of vapor crystal growth, solutal crystal growth, and drop dynamics. These instruments are characterized by large size and relatively high cost. At present, no further pieces of equipment of this type have been proposed or are being planned.

What is in the planning stage, however, is a series of Spacelab flights named the International Microgravity Laboratory. The concept of this series is to utilize instrumentation that is available, not only in the United States but in Europe and Japan as well. Because money has been a constraint of the program (and it is anticipated that this will continue), the concept of the international use of existing equipment has generated a great deal of enthusiasm. The first International Microgravity Laboratory is expected to be flown in 1987, with flights thereafter on approximately one-year intervals.

NASA has established an ambitious flight program, which, if vigorously pursued, is expected to lead to the discovery of new and exciting results. However, we are seeing a continuation of Shuttle operational problems that cause postponement of experimental flights. This has resulted in a delay of data becoming available to our researchers and is adding a financial burden to an already constrained budget. While every effort is being made to remedy this situation, lack of flight data poses a serious threat to the program.

THE APPLICATIONS PROGRAM

While this workshop is devoted primarily to a review of the science base of the Microgravity Program, it must be noted that the applications part is an integral area of activity. Applications in space, otherwise known as the Commercial Use of Space, has been cited by President Reagan as an area of great interest to the United States. As a result, the NASA Administrator recently established a staff office reporting directly to him to handle all commercial activities and, primarily, to act as NASA's interface with the industrial community. One important commercial area is expected to be that of materials and materials processing.

There are three types of arrangements that NASA has currently undertaken in cooperation with an industrial partner. These are the Joint Endeavor Agreement (JEA), the Technical Exchange Agreement (TEA), and the Industrial Guest Investigator (IGI).

JEA involve low-gravity experiments with specific commercial goals, using equipment built and funded by the commercial partner. NASA typically provides launches into space and may use the commercial equipment for its own research. The commercial data may remain proprietary. In any event, there is no exchange of funding, with the partners each paying their own share of the project.

TEAs allow commercial investigators to work with NASA investigators in focused areas of applied research, with essentially no additional investment on the part of the commercial partner. This research usually occurs before a space experiment has been determined necessary. Under this type of arrangement, the commercial partner may use NASA ground-based facilities (drop tubes, drop towers, aircraft, and laboratories) to support the decision whether to proceed with the next step. Again as in the JEA, each partner pays his own cost.

IGI pursue topics of mutual interests with NASA researchers at NASA facilities, with their salaries and expenses paid for by their commercial sponsors.

At present, within the area of Microgravity Science and Applications, we are working on three JEAs: (1) with McDonnell Douglas in the area of continuous-flow electrophoresis, (2) with Microgravity Research Associates in the production of gallium arsenide crystals using electroepitaxial growth, and (3) with the 3M Corporation in the area of diffusive crystal growth.

Possibly because of the large investment in equipment required for the JEA, we have seen, over the past year, only a single new JEA negotiated. At the same time, we have, or are currently negotiating, 17 TEAs, a fivefold growth over the past year. The conclusion that may be drawn from this growth is one of cautious interest on the part of the industrial community.

At present, we have no arrangements for IGIs, but we are negotiating with two large pharmaceutical companies for such a program in the area of protein crystal growth.

THE FUTURE PROGRAM

During the summer of 1984, the President announced his decision to embark on the development of a Space Station. The future of the Microgravity Science and Applications Division is inextricably tied to this development. One element of that Space Station will be a Microgravity Laboratory that is expected to provide experimental equipment for both scientific and applied research. We are currently involved in the Space Station Phase B studies, which will determine the requirements of such a laboratory for microgravity research in the 1990s and beyond. Initial study activities have indicated a need for specific generic-type activities as well as individual research work. Total power expected to be available is on the order of 60 kW. G levels are expected at 10^{-5} or better. Long durations of extreme low gravity for the growth of ultrapure crystals will become a reality. It is our plan to fly third-generation equipment on the Space Station including equipment for diagnostics. However, until the results of our current flight program become available, this design will be deferred. Development of the Space Station is expected to begin in 1987, with the initial operating capability in 1992/1993.

In preparation for a Microgravity Laboratory as part of the Space Station, we have established at the Lewis Research Center a Materials Science Laboratory, which we expect to develop into a ground-based prototype. This laboratory, which is expected to begin operations in 1985, will contain duplicates of our current flight equipment as well as diagnostic equipment. Flight equipment will be upgraded to meet the requirements of the researchers using the facility. This facility will be open, at no charge, to interested NASA investigators, as well as to the university and industrial communities. As the design of the Space Station progresses, this laboratory will evolve into a ground-based test bed that will enable us to check design as well as enhance the Space Station laboratory itself. It will provide a preview for those proposing investigations as to capability and possibilities aboard the Space Station.

SUMMARY

In summary, the Materials Processing in Space Program, which began in the 1970s, has been reorganized into the Microgravity Science and Applications Program. This program is predicated on a strong scientific, ground-based, research activity evolving into a focused flight program that will return data of a fundamental and applied nature. The NASA Center involvement is strong and committed. Finally, a program of Shuttle flights is planned, but the ultimate vehicle for microgravity experimentation in a low-g environment will be the Space Station.

2. OVERVIEW: METALS AND ALLOYS

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INTRODUCTION

The research sponsored by the Microgravity Science and Applications Program in Metals and Alloys may be divided into eight categories: coarsening and stability of two-phase mixtures, solidification of supercooled melts, alloy segregation, scaling laws, thermophysical properties, metals deposition from solutions, convective interactions, and technological applications. These categories were suggested by the Metals and Alloys Discipline Working Group headed by Marty Glicksman. This working group's function is to help NASA promote the use of microgravity research to enhance fundamental understanding of the processing and properties of metals and alloys, to communicate to NASA and the research community the importance of microgravity research in metals and alloys, and to identify and help to establish a sound ground-based science program that will stimulate appropriate innovative flight experiments. This working group is in the process of identifying key issues in the above categories and evaluating the existing research programs to determine how well these issues are being addressed. This process will result in recommendations to the Director of the Microgravity Science and Application program in terms of priorities and research areas that should be strengthened. Because of this ongoing activity, this synopsis will follow the format developed by the working group. It will not be possible to discuss each activity, so only the most significant results in each category will be mentioned.

COARSENING AND STABILITY IN TWO-PHASE MIXTURES

Several early flight experiments attempted to form fine dispersions from monotectic systems and found that almost complete phase separation occurred even in the absence of buoyancy-driven sedimentation and convection. From the theory of critical-point wetting developed by Cahn, we now understand the instability observed in these early experiments. Potard (CENG, France) demonstrated that this massive phase separation could be avoided by choosing a crucible material that was preferentially wet by the majority phase. However, droplet migration in a thermal gradient from thermocapillary flows and possibly other effects still caused considerable agglomeration, and uniform, fine dispersions cannot be formed until these effects are understood and controlled.

Gelles on STS-7 attempted to measure the amount of thermal migration by holding a two-liquid-phase Al-90 wt. percent In mixture in a thermal gradient. Contrary to expectations, the Al-rich minority phase droplets migrated to the cold end, which implies that either the interfacial energy increases with temperature or that there are some solutal or other effects that are larger than the thermocapillary effect.

CONTAINERLESS STUDIES OF NUCLEATION AND UNDERCOOLING

A number of undercooling experiments have been conducted using the 100-m evacuated drop tube at the Marshall Space Flight Center. Nb alloys have been undercooled in excess of 0.23 T_M, and metastable phases such as the A-15 superconducting phase have been obtained in bulk samples. However, the recalescence heating causes transitions back to equilibrium phases unless the samples are rapidly quenched.

Collings (Battelle-Columbus) rapidly solidified a series of austenitic, fcc-structured, nonmagnetic stainless steel samples both by the hammer and anvil method and by impacting a quench plate after free fall in the drop tube. The rapid solidification traps some of the metastable bcc-structured, delta-ferrite magnetic phase. However, Collings found that the drop-tube samples of Nitronic 40, which undercooled before quenching, contained 86 percent of the delta-ferrite phase, whereas the hammer and anvil samples contained only 40 percent. This indicates that metastable phase formation can be promoted by deep undercooling before quenching.

Progress is being made in developing techniques to eliminate heterogeneous nucleation sites from melts. Spaepen (Harvard) has found, for example, that a molten oxide flux, Ba_2O_3, is more effective than vacuum melting in the preparation of bulk metallic glass from $Pd_{40}Ni_{20}P_{20}$.

A microgravity experiment is being prepared by Glicksman (RPI) to test his theory of dendrite growth at low undercoolings. At small degrees of undercooling, convection plays a significant role in dendrite growth, and an unambiguous test of the theory is difficult in unit gravity.

Johnston (MSFC) has compared dendrite growth in microgravity with that in unit gravity for both transparent model systems ($NH_4OH + H_2O$) and in metals. She has found that the secondary arm spacing tends to be larger in microgravity. The reason for this is not yet understood.

An Au-Ge sample solidified on Skylab was reported to have a superconducting transition at 1.5 K, whereas the ground control sample exhibited no superconductivity. Chu (U. Houston) recently remeasured the Skylab sample and found no superconductivity. However, he found that splat-quenched Au-Ge had two superconducting transitions at 1.6 K and at 2.4 K. These superconducting phases are metastable, and the 2.4 K phase exists only for a few hours at 300 K. However, the 1.6 K phase persists for several weeks. Additional experiments are in progress using the MSFC drop tube and drop tower to obtain more information on this peculiar set of results.

ALLOY SEGREGATION

Larson (Grumman Aerospace) obtained an unusually high magnetic coercivity on his MnBi-Bi sample that was directionally solidified on the Apollo-Soyuz flight. This high magnetic coercivity was later found to be the result of a much finer eutectic rod structure obtained on the flight sample as compared with the ground control sample. Subsequent experiments on sounding rockets have confirmed this result, which is contradictory to the Jackson and Hunt theory of eutectic solidification.

Frazier (MSFC) has developed various transparent organic systems that are analogs of monotectic metallic systems. One such system, succinonitrile/H_2O, can be made isopycnic at selected temperatures by adding amounts of D_2O. Using such systems, phase separation can be observed directly and the various mechanisms studied. One interesting result found by Kaukler (UAH) was that the interface of this system, when solidified on a thermal gradient stage of a microscope, resembles a batch of growing mushrooms. The stalks do not follow the heat flow but tend to twist about. They are capped by a bubble of liquid, which occasionally pops off and moves rapidly away, driven by Marangoni convection. The exact nature of this behavior is still being analyzed, but it is suspected that this is the result of the fact that the solid's critical wetting temperature is below the monotectic temperature. This would prevent the formation of a stable trijunction between the two liquid phases and the monotectic solid.

SCALING LAWS

In addition to the previously described work of Glicksman, who is attempting to verify scaling laws for dendritic growth, and by Larson, who discovered a departure from the well-known λv^2 = constant law of Jackson and Hunt, an effort is being made by Langer and others at the Institute for Theoretical Physics (ITP), Santa Barbara, to apply various pattern formation theories to the dendrite-formation problem. This institute is fostering interactions between theoretical physicists and material scientists in order to identify problems of common interest in which some of the powerful theoretical approaches developed for other branches of physics may be applied to problems in materials science. Progress is also being made in advancing the Ostwald ripening theory to account for finite volume fractions of second-phase material and to couple this with nucleation theory to describe the precipitating phase formation and structure in alloys.

Interactions between the National Bureau of Standards (NBS) and visiting scientists at ITP have also resulted in the identification of a unique structure in rapidly quenched Al-14 percent Mn. This material has long-range directed bonds with icosahedral symmetry, which does not allow the formation of a regular structure but instead forms a quasi-periodic structure that is neither amorphous nor crystalline.

THERMOPHYSICAL PROPERTIES

The Microgravity Science and Applications Program has prompted many efforts to measure thermophysical properties. These efforts are motivated by the opportunity to make unique high-temperature measurements using the containerless methods possible in a microgravity environment and by the need for such measurements to support the development of other experiments to be done in microgravity.

Margrave (Rice) and Bonnell (NBS) have developed electromagnetic levitation methods coupled with electron-beam heating and drop calorimetry to obtain heats of fusion, heat capacities, and enthalpy functions for various refractory materials. They have reported the first direct measurements of liquid W and have found that heat capacities for liquid Ag and Ga exceed the 3R equipartition value and are constant over a wide range of temperatures. Why this is the case is not yet understood. Trinh (JPL) has developed containerless techniques for obtaining viscosity and surface tension of molten materials. This would be especially important for highly corrosive melts whose surfaces would be contaminated by contact with crucible walls.

Cerzairliyan (NBS) using a liquid bridge technique, obtained dynamic measurements of selected thermophysical properties of molten metals. Rods of the material of interest are melted by the passage of a heavy direct current during the low-g parabola of a KC-135 flight. Thermophysical properties, such as heat of fusion, heat capacity, and electrical resistivity, are obtained as a function of temperature. In the first test the liquid zone collapsed at a superheat of 50 K. Whether this collapse was the result of a g spike or a hydrodynamic instability is not clear. Additional tests are planned with the objective of extending the superheat to 200 K which can be attained during a low-g parabola on the KC-135.

Hardy (NBS) has measured the surface tension of pure molten silicon with sufficient precision over a wide temperature range to obtain, for the first time, an accurate determination of the variation of surface tension with temperature. This property is an essential parameter for the prediction of Marangoni convection in float-zone processes. Stroud (Ohio State) has developed techniques for computing thermophysical properties, including surface tension, from first principles by combining the statistical-mechanical theory of the liquid state with an electronic pseudo-potential theory of electrons in metals. The theory can be extended to include impurity effects on surface tension and to obtain solid-melt interfacial energies. This latter property is difficult to measure experimentally and is crucial in estimating maximum undercooling and for testing the various homogeneous nucleation theories.

Various techniques have been developed for measuring diffusion coefficients and thermal diffusivities for alloy systems to be used in future microgravity experiments. These properties are needed to design various directional solidification and crystal-growth experiments. A laser pulse method has been used to measure the diffusivity of $Hg_xCd_{1-x}Te$ in the melt over a variety of x values. This measurement is difficult because the vapor pressure at the melting point is on the order of 100 atm. An order-of-magnitude increase in diffusivity was found to occur as the material melts. This increase can seriously alter the heat flow and warp the solidification interface as the heat carried by the melt must be transferred to the ampoule wall.

DEPOSITION OF METALS FROM IONIC SOLUTIONS

This appears to be a promising area for microgravity research since thermo-solutal convection is virtually always involved in such processes. Currently, the only active program in this area sponsored by the Microgravity Program is the work of Sadoway (MIT). Sadoway is using laser schlieren optical techniques to correlate flow patterns with cell electrical performance and plating morphology. Transport-limited deposition can be achieved using flat electrodes arranged so that the solutal gradient stabilizes the convective flow. Such a configuration generally produces a rough dendritic surface since metal ions attach at the first available site, usually the highest point. This is analogous to dendritic growth in undercooled melts. Laminar flows along the surface tend to produce dendrites that grow in the direction of the flow. Turbulent flows tend to produce powdery deposits with entrained electrolyte.

CONVECTIVE INTERACTIONS

Brown (MIT) has analyzed the effects of convective flows produced by radial thermal gradients in Bridgman-type directional solidification of binary alloy systems. Even with a solutally stable layer formed by the rejection of a denser component at the interface, he shows that significant radial segregation will result unless the thermal Rayleigh number is below 102. This means that such segregation is virtually impossible to avoid in Earth's gravity.

Coriell and co-workers (NBS) have studied the nature of such flows and their interactions with the solidification interface. They have shown that strong coupling can occur between such flows and the interface shape. Linear stability analysis for flows parallel to an interface has predicted instabilities that agree with experimental results of Glicksman (RPI), who first observed these coupled modes in a vertical column of molten succinonitrile surrounded by a solid annulus of the solid.

Pirich (Grumman Aerospace) has shown that the result of Larson's space experiment with MnBi/Bi eutectic can be essentially duplicated by applying a strong magnetic field. This tends to confirm that lower convection is indeed responsible for the finer rod formation observed by Larson. What remains as a puzzle, however, is that experiments with convection agree with the Jackson and Hunt theory, which is based on diffusion as the only transport mechanism, whereas experiments with lower convection (magnetically dampled or in reduced gravity) deviate from the theory.

TECHNOLOGICAL APPLICATIONS

Curreri (MSFC) has directionally solidified various cast iron compositions during the multiple low-gravity parabolas flown by the KC-135. The low-gravity time available allows the solidification of only 1-2 mm of material in low g followed by several millimeters solidified in 2 g. This is coordinated with Deere and Company's research program to study the influence of convection and other processing parameters on the graphite morphology that determines the iron's structural properties and machinability. These experiments are being done as part of a Technical Exchange Agreement between Deere and NASA, and a JEA is being developed to extend this research to Shuttle experiments.

Deere is also working with the NBS to obtain accurate thermal properties on various types of molten cast iron to use with their thermal modeling effort. With new sophisticated computer techniques, heat flow and solidification may be computed for complicated castings, which are important for the prediction of their physical properties. The needed thermophysical data may require low-gravity experiments to eliminate convective effects.

Johnston (MSFC) and Curreri have also investigated the solidification of MAR-M 246 in the directional solidification furnace flown on the KC-135. This complex alloy has a large number of components with a wide variety of densities. Macrosegregation is significant during solidification in Earth's gravity. They are finding interesting changes in microstructure in the portion solidified in low g such as variations in dendrite arm spacing and in the amount and morphology of the carbide formation. A Shuttle experiment is being developed to extend the available low-gravity time so that enough sample can be produced to determine if these are transient or steady-state effects and to obtain sufficient sample for evaluation of mechanical properties.

Hellawell (Michigan Tech) has carried out experiments to investigate the origins of the long vertical columns of solute-rich melt that arise in castings in order to determine what controls their spacing and diameters with the hope of developing better control strategies for inhibiting this effect. Using transparent analog systems ($NH_4Cl + H_2O$) and metallic alloy systems (Pb-Sn), Hellawell has found that the channels do not originate from the dendritic growth front as might be suspected, but instead appear to result from liquid-phase perturbations ahead of the growth front. Formation of such channels can be inhibited by slow rotation about an inclined axis. The inhibiting mechanism is still being analyzed.

3. METALS AND ALLOYS - THEORY

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During the first-order phase transformation, the motion of the interface between the two phases depends on the transport of heat and solute. For alloy solidification or crystal growth from the melt, the parent phase is a fluid and the transport is by both diffusion and convection. For metallic alloys, thermal diffusivities, κ, are four orders of magnitude greater than solute diffusivities, D, so that fluid flow has a much greater effect on solute distribution than on the temperature field. The solute distribution at the crystal-melt interface directly determines the solute distribution in the solidified material since diffusion rates in the solid are generally extremely small.

The general problem of mathematical modeling of fluid flow during solidification is challenging since it involves the nonlinear Navier-Stokes equations and Stefan problem, i.e., the boundary conditions on the temperature, concentration, and fluid flow fields are specified at the solid-liquid interface whose position is unknown. The temperature, concentration, and fluid flow fields are not only coupled through the convective and buoyancy terms in the field equations but also through the boundary conditions at the solid-liquid interface. These boundary conditions directly couple the temperature and solute field, since the melting point depends on composition, and indirectly couple the fluid flow field to the temperature and concentration field, since the position of the interface is determined by these fields. Further, the role played by the solid-liquid surface energy requires calculation of the mean curvature of the solid-liquid interface. For metals and semiconductors, the Prandtl number ($Pr = \nu/\kappa$ where ν is the kinematic viscosity) is of the order of 10^{-2} and the Schmidt number ($Sc = \nu/D$) is of the order of 10^{2}; these parameters indicate widely different length and time scales for mass, momentum, and energy transport and may cause additional numerical problems.

Directional solidification of a binary melt to form a single-phase solid at constant velocity V is most amenable to theoretical analysis and is a good approximation to directional solidification experiments in which there is uniform relative motion of the sample and its thermal environment. Since the solute concentration of liquid is usually quite different from that of a solid with which it is in equilibrium, there is either rejection or preferential incorporation of solute by the solid at a moving crystal-melt interface. This results, for a quiescent liquid, in an exponential concentration gradient that extends ahead of the interface with a characteristic distance of D/V. The temperature gradient is also exponential with characteristic distance κ/V; however, κ/V is usually sufficiently large that the temperature gradient is essentially constant within each phase.

If the crystal-melt interface is planar and there is no fluid flow in the melt, then at steady state the solidified material will have a uniform solute distribution. Even in the absence of convection, it is well known that for sufficiently high solute concentrations the crystal-melt interface will be unstable[1] and develop into a cellular or dendritic interface with lateral solute segregation (microsegregation). For small solidification velocities (typically less than 10^{-2} cm/s), an excellent approximation to the stability criterion is the modified constitutional supercooling criterion, i.e., the planar interface is stable if

$$G^*/(mG_c) > 1$$

with

$$G^* = (k_S G_S + k_L G_L)/(k_S + k_L),$$

where k_L and k_S are thermal conductivities of liquid and solid, respectively, G_L and G_S are unperturbed temperature gradients at the interface in the liquid and solid, respectively, m is the change in melting temperature with solute concentration, and G_c is the unperturbed solute gradient at the interface in the liquid. The product mG_c is always positive, even though solutes can either raise or lower the melting temperature. Basically, positive temperature gradients are stabilizing while the solute gradient is destabilizing. Another stabilizing influence, not evident in the equations, is the solid-liquid surface free energy, which tends to minimize interfacial area. At small velocities, the surface energy has a negligible effect on the stability criterion but plays an important role in determining the wavelength λ at the onset of instability; typically these wavelengths are in the range 1-100 μm and decrease with increasing velocity.

Recently, there has been renewed theoretical interest in the nonlinear aspects of the morphologies that occur during directional solidification. For interface shapes that do not differ too much from planarity, the free boundary problem may be handled by means of linear stability analysis[1] and nonlinear expansion techniques.[2] High-speed computers have made possible numerical calculations of nonplanar interface morphologies that differ significantly from planarity.[3,4] Since cellular spacing are generally orders of magnitude smaller than sample dimensions, computational domains with periodic boundary conditions corresponding to wavelengths of anticipated cell spacings are used. In general, it is possible to find a family of steady-state solutions (interface shapes, temperature, and concentration fields) for a range of wavelengths. The fundamental problem of wavelength selection has been long-standing and is just beginning to be understood.[5]

Steady-state two-dimensional interface shapes and interface concentrations are shown in Figure 3.1. The three calculations[4] correspond to bulk alloy concentrations that exceed the critical concentration for instability by 0.5, 4.0, and 13.0 percent. The cell groove deepens, and accompanying solute segregation increases rapidly as the bulk concentration is raised. Recent work by Ungar and Brown[6] successfully follows the evolution of cells to groove depths of up to 15 times their wavelength. A nonlinear stability analysis of three-dimensional cells has recently been carried out by Sriranganathan et al.[2] in an attempt to understand the complex morphologies that have been observed experimentally. For example, in lead-antimony alloys[7] the interface morphology at the onset of instability depended on crystallographic orientation; nodes (circular wells of liquid penetrating into the crystal) developed when the growth direction was near [100] or [111], while elongated cells developed near [110]. At higher degrees of instability, regular (hexagonal) cells developed.

It is well known that horizontal temperature and concentration gradients give rise to buoyancy-driven fluid flow.[8] In order to minimize these horizontal gradients growth should be in the vertical direction. For a normal liquid, which expands on heating, growth vertically upward will ensure that in the absence of solute the liquid density will decrease with height. However, even for this growth configuration, since container walls are not perfectly adiabatic, the differences in thermal conductivity between crystal, melt, and container and the latent heat released on freezing will give rise to horizontal temperature gradients, resulting in fluid flow. The nature of this flow in prototype crystal-growth systems is beginning to be understood through extensive numerical modeling.[9] For vertical growth in the absence of horizontal temperature and concentration gradients, a motionless solution will formally satisfy the fluid-flow equations; however, such a solution is not necessarily stable and convective instability may occur.

When both the temperature and solute fields individually cause the density to decrease with height, there is no possibility of convective instability. However, even in this case, there can be undesirable convective effects. During measurements of cell spacings in Al-6 wt. percent Cu alloys, McCartney and Hunt[10] observed that the macroscopic interface was highly curved. Since the curvature could be eliminated by using the ternary system Al-Mg-Si, which has a very low value of the temperature derivative of the liquid density along the liquidus line, convection appears important in the stably stratified Al-Cu system. Presumably, convection due to horizontal temperature gradients interacts with the solute gradient to cause a nonuniform solute concentration at the interface.

If a solute that is rejected at the crystal-melt interface decreases the density of the melt or a solute that is preferentially incorporated at the crystal-melt interface increases the density of the melt, then the solute field in the absence of a temperature gradient will cause the liquid density to increase with height during growth vertically upward, and there is the possibility of convective instability. The unperturbed static density profile due to both temperature and concentration variation is of little utility in predicting the onset of convection since one can have a statically unstable density profile without convection and a statically stable density profile with convection. The latter case of convection, even though the liquid density decreases with height, is a feature of double diffusive convection and arises owing to the difference in diffusivities of the two diffusing species, viz., solute and heat.[11] The results of linear stability analysis[12] of combined morphological and double diffusive convective instabilities are shown in Figure 3.2 for the growth vertically upward of lead containing tin with a temperature gradient in the liquid of 200 K/cm for three different constant gravitational accelerations. The system is unstable for tin concentrations C_∞ above the curves. The upper line with negative slope represents the onset of morphological instability and is essentially independent of the gravitational acceleration. The three curves labeled with values of the gravitational acceleration represent the onset of convective instability and are very sensitive to the gravitational acceleration. For tin concentrations below the dashed-dot curve the static liquid density decreases with height. Extremely small amounts of solute can cause convective instability in experiments on Earth, e.g., at a growth velocity of 1 μm/s the critical tin concentration is 3.2×10^{-4} wt.

percent. Additional calculations for a growth velocity of 1 μm/s show that a vertical magnetic field of 1 tesla increases the critical concentration by an order of magnitude.

The behavior of the convective stability curves shown in Figure 3.2 can be qualitatively understood by recognizing that the relevant length scale is the diffusion distance D/V. The thermal Rayleigh number Ra is then proportional to $G_L(D/V)^4$ and the solutal Rayleigh number is proportional to $G_c(D/V)^4$ or, equivalently, $c_\infty(D/V)^3$. For fixed G_L and large growth velocities the thermal Rayleigh number becomes very small and stability is determined by a balance of the destabilizing solute field and the stabilizing viscous forces; instability occurs when the solutal Rayleigh number exceeds about 10.[13] For small growth velocities the thermal Rayleigh number is large, and stability is determined by a balance of the destabilizing solute field and the stabilizing thermal field; the solutal and thermal Rayleigh numbers are roughly comparable.

The coupling between morphological and convective modes of instability is rather weak except near the concentrations and velocities at which the morphological and convective curves cross in Figure 3.2. In the vicinity of this crossing, instabilities that are oscillatory in time may occur.

In order to determine the extent of solute segregation and the nature of the flow field caused by double diffusive convection during directional solidification, the time-dependent nonlinear differential equations for fluid flow, concentration, and temperature have been solved numerically in two spatial dimensions for small Prandtl numbers and large Schmidt numbers.[14] For slow solidification velocities, the thermal field has an important stabilizing influence: near the onset of instability the flow is confined to the vicinity of the crystal-melt interface. Further, for slow velocities, as the concentration increases, the horizontal wavelength of the flow decreases rapidly--a phenomenon also indicated by linear stability analysis. For a narrow range of solutal Rayleigh numbers and wavelengths, the flow is periodic in time.

The steady-state stream function and solute field as a function of position for a semiconductor melt (Schmidt number of 10 and Prandtl number of 0.01) is shown in Figure 3.3. The crystal-melt interface, assumed planar in the calculations, is located at the bottom of the figures, and the interface concentration varies about 60 percent along the interface. For this calculation, the thermal Rayleigh number is large and the solutal Rayleigh number exceeds the critical solutal Rayleigh number for the onset of convection by only 30 percent so that the flow is confined to the vicinity of the crystal-melt interface. The maximum flow velocities are relatively small. For example, for a lead-tin alloy at a growth velocity of 2.0 μm/s and a liquid temperature gradient of 200 K/cm, the maximum flow velocity was 17.0 μm/s when the bulk concentration exceeded the critical concentration for instability by a factor of 4.3. However, this flow velocity is sufficient to cause a 30 percent variation in the solute concentration at the crystal-melt interface.

We discuss briefly a few experiments that demonstrate the effects of convection during directional solidification. Boettinger et al.[15] studied the growth of lead-rich lead-tin off-eutectic alloys. For growth vertically upward, the solute field is destabilizing and substantial variation in composition in the growth direction (macrosegregation) was found. A vertical or horizontal magnetic field of 0.1 Telsa did not reduce the macrosegregation, but downward solidification in small (3 mm) diameter tubes virtually eliminated the macrosegregation. Sample and Hellawell[16] have recently investigated the mechanisms of channel segregation during alloy solidification vertically upward. Channels containing high solute concentrations develop from positions near a dendritic growth front when the solute is less dense than the melt, and plumes rise to the top of the bulk liquid. Recent experiments by Schaefer[17] on the directional solidification vertically upward of succinonitrile containing ethanol have shown an interesting interaction between thermal convection due to radial temperature gradients and the solute field. For growth conditions near the onset of morphological instability, a macroscopic pit develops in an otherwise slightly curved crystal-melt interface. The flow is basically down near the container walls and upward near the axis of the tube. The solute concentration is increased near where the flow turns upward, and the macroscopic pit occurs in this region. Morphological instability then takes place in the pit, and the pit disappears leaving a region of cellular growth in its place.

Except for a very limited range of parameters, the double diffusive and morphological instabilities that occur during directional solidification are rather weakly coupled. In particular, the short-wavelength morphological mode is essentially unaffected by the possibility of convection in the quiescent liquid. Owing to very different length scales for convective motions and for cellular interfaces, it will be extremely challenging to model the effect of convection on cellular morphologies. Various simplified models for the effect of a fully developed flow on morphological instability have been reviewed recently.[18] Delves[19] has calculated the effect of forced flow parallel to the crystal-melt interface on the onset of morphological instability. The effect of such a flow on combined double diffusive and morphological instabilities has recently been treated for Couette flow.[20] Such a flow does not affect perturbations with wave vectors perpendicular to the flow. For perturbations with wave vectors parallel to the flow, the onset of morphological instability is somewhat suppressed and double diffusive instability is greatly suppressed. When instabilities occur, they are oscillatory and correspond to traveling waves. A complicated flow field could be approximated in the vicinity of the crystal-melt interface by a simple flow such as a Couette flow. This would simplify the modeling of the interaction between fluid flow and the crystal-melt interface. However, Glicksman and colleagues[21-23] have recently demonstrated coupling between hydrodynamic and morphological instabilities for which such approximation is clearly incorrect.

We conclude with a brief description of this coupled instability. A long vertical cylindrical sample of high-purity succinonitrile was heated by an electrical current passed through a long coaxial heating wire, so that a vertical melt annulus formed between the coaxial heating wire and the surrounding crystal-melt interface. The outer radius of the crystal was maintained at a constant temperature below the melting point of the material. This arrangement permits the temperature to decrease monotonically from the melt to the solid across the crystal-melt interface, and consequently the interface would be morphologically stable in the absence of fluid flow.

The thermal gradients in the melt induce buoyancy forces that cause the fluid to flow upward near the heating wire on the axis and downward near the crystal-melt interface. When linear stability analysis is used to calculate the critical Grashof number for instabilities of the axisymmetric flow occurring between two vertical infinite rigid coaxial cylinders held at different temperatures, it is found that for a Prandtl number of 23 (corresponding to succinonitrile), the flow is unstable to an axisymmetric perturbation above a Grashof number of the order of 2000, and the resulting wave speed of this perturbation is comparable with the maximum in the characteristic unperturbed flow velocity.

In contrast, the experimental observations with a crystal-melt interface indicate an asymmetric helical instability at a critical Grashof number of about 150 with a wave speed 2 orders of magnitude less than the unperturbed flow velocity. Linear stability analysis reveals that the instability is due to a coupling between a basic hydrodynamic instability in the buoyant flow and the deformable crystal-melt interface. The crystal-melt interface lowers the critical Grashof number of an analogous rigid-walled system by an order of magnitude; furthermore, the hydrodynamic mode that is actually destabilized by the interface is not the least-stable mode in the rigid-walled system. The calculations show that the instability may be regarded either as a large alteration of a basic hydrodynamic instability by the crystal-melt interface or as a significant modification of the morphological stability of the interface by the presence of the buoyant flow. The instability is sensitive to the form of the flow field, e.g., it does not occur for Couette or Poiseuille flows.

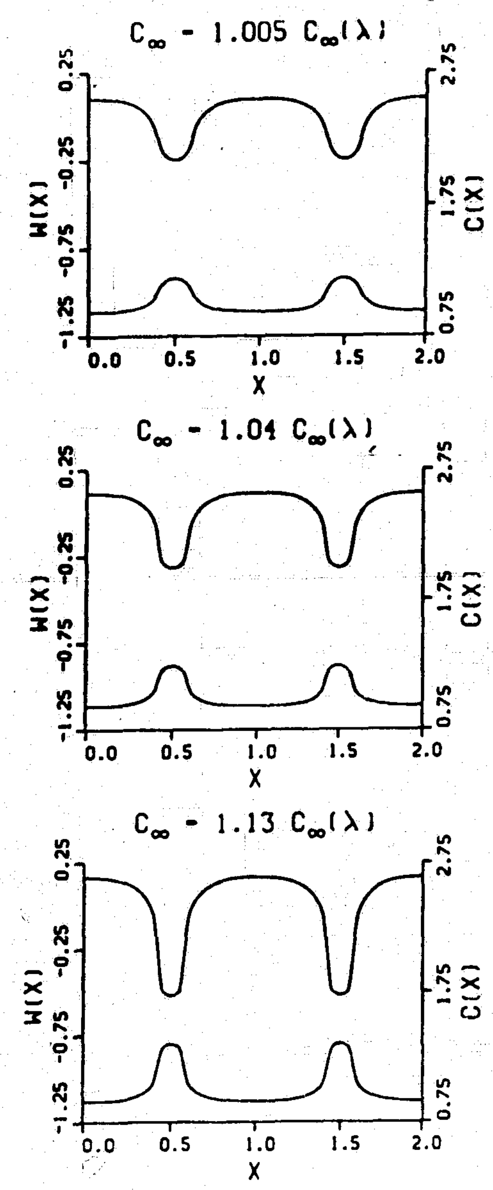

FIGURE 3.1. Plots of interface shape and distribution of solute in the solid for a given wavelength as the bulk concentration is increased. The top curve in each plot is the interface shape, the bottom curve is the concentration of solute. Two wavelengths are shown.

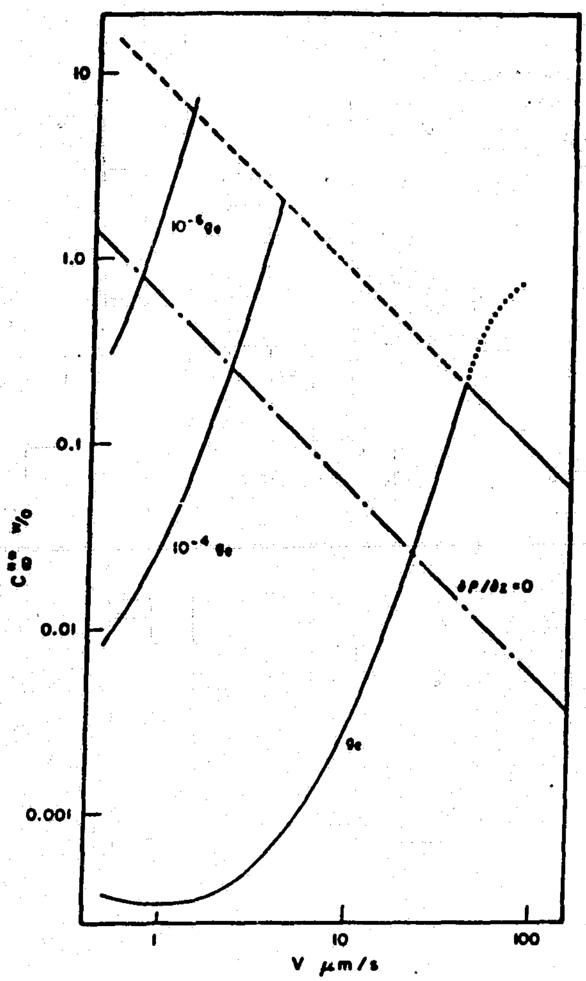

FIGURE 3.2. The critical concentration c_∞^{**} of tin in lead above which instability occurs as a function of the velocity V of directional solidification for a temperature gradient G_L in the liquid of 200 K/cm. The solid curves represent gravitational accelerations g_e = 980 cm/s^2, 10^{-4} g_e, and 10^{-6} g_e. The upper line with negative slope represents the onset of morphological instabilities; the nearly parallel dashed-dot line labeled $(\partial\rho/\partial z) = 0$ represents the neutral density criterion. The dotted extension of the curve labeled g_e corresponds to oscillatory instabilities.

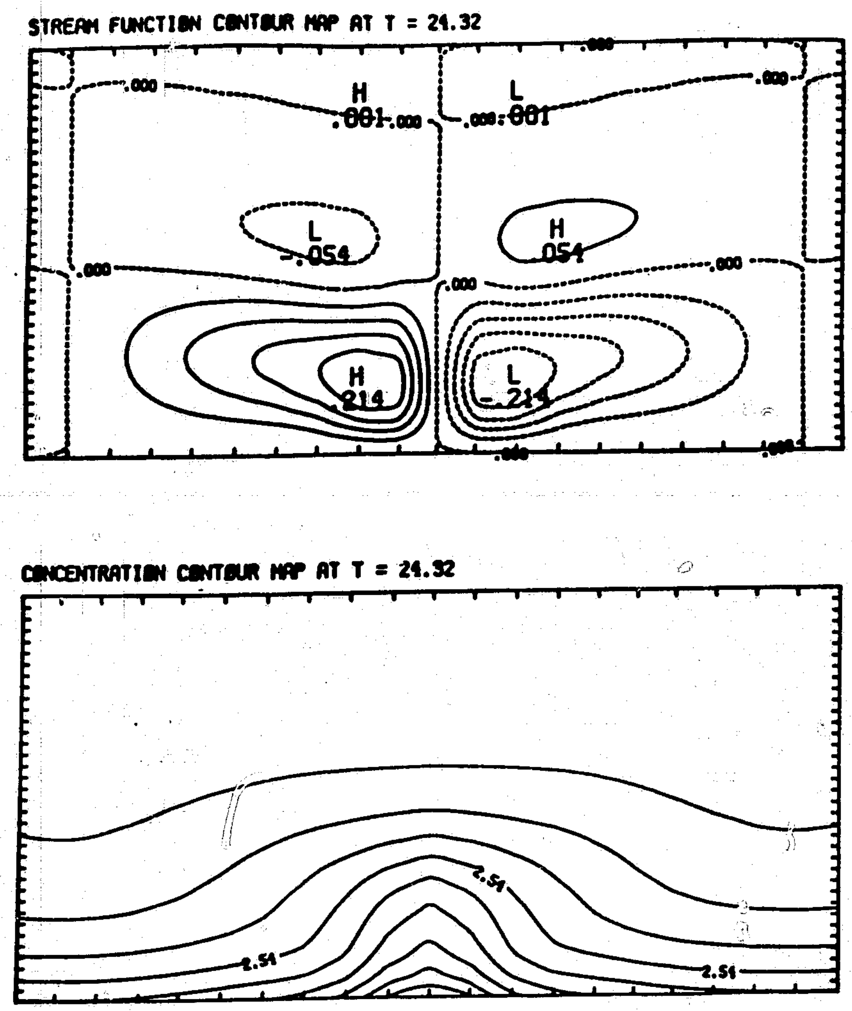

FIGURE 3.3. The steady-state stream function and concentration as a function of position for a bulk concentration 29 percent above the critical concentration for the onset of convection. Parameters used in the calculation are typical of semiconductor melts. In the concentration plot, the increment of (dimensionless) concentration between contours is 0.37. The maximum flow velocity is about 5 times the crystal growth velocity of typically 20 μm/s.

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4. INFLUENCE OF MELT CONVECTION ON SOLID-LIQUID INTERFACE UNDER TERRESTRIAL AND MICROGRAVITY CONDITIONS

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ABSTRACT

The solidification of metals as well as other more general crystal growth processes results in the formation of chemical and thermal inhomogeneities within the melt. In the presence of gravity such inhomogeneities lead to density gradients, which in turn are responsible for convection. When the crystal growth morphology is dendritic, as is often the case in casting, then the convection patterns become three dimensional and complex. The underlying kinetic and morphological behavior of dendritic crystals is known to be governed by heat conduction and solute diffusion--both spatially isotropic transport processes. Convective transport normally leads to anisotropic redistributions which change the fundamental crystal growth behavior. Under microgravity conditions the convection will be reduced, permitting purely diffusive transport to dominate the solidification.

When convection-free transport occurs, then the kinetics and so-called morphological "scaling laws" simplify, thus allowing a quantitative check on the interfacial responses such as sidebranch development, tip stability, and microsegregation. The availability of relatively long-duration microgravity now makes feasible achieving diffusion-limited dendritic growth, even at small thermodynamic driving forces. Finally, it should be noted that at small driving forces, the dendritic growth occurs at relatively large size-scales, allowing far greater ease of microstructural observation than can be achieved under terrestrial conditions.

BACKGROUND

It is now generally recognized that when materials solidify into supercooled or supersaturated melts, they freeze in an unstable manner, resulting in a branching, tree-like crystal form termed dendritic. Virtually all metals and alloys, and many other substances, including fused salts and certain organic systems, freeze dendritically.

Dendritic solidification is scientifically interesting because it exemplifies a simple case of "pattern formation," wherein a structureless molten phase spontaneously gives rise to a "pattern" or microstructure. This pattern results primarily from the diffusive redistribution of latent heat and/or solute. A remarkable feature of the redistribution is that diffusion occurs as a spatially isotropic process, and yet an elaborate, highly anisotropic, pattern results. See Figure 4.1.

As latent heat flows away from an advancing dendrite, the resultant temperature gradient alters the local density of the surrounding molten phase. Similarly, as solute is rejected from the solid-liquid interface, the resultant concentration gradient independently alters the density. These inhomogeneities in local density are acted upon by gravity to induce a body force within the solid-liquid system. One result is the prompt flow, or convection, of the spatially inhomogeneous fluid and stressing of the solid. This is not to imply that were gravity eliminated all internal force fields would vanish. Indeed, even in the absence of gravity, streaming flows must occur in the melt to compensate for the volume change on freezing. Nonetheless, the major source of convective motions is buoyancy-driven flows that are proportional to the gravitational acceleration. The bulk motions of the convecting melt alter the diffusion fields surrounding the dendritic crystals. The distortion of the local thermal gradients and the solutal gradients changes the growth rate and pattern of development. For example, under terrestrial conditions, one may observe a significant spatial anisotropy of the growth rate and microstructure, which would not occur under strict zero-g conditions. Figure 4.2 shows an excellent example of how dendritic morphologies change with respect to their orientation to the g-vector.

We plan to develop an experiment that permits quantitative measurement of dendrite growth under low-g conditions. Using modification of techniques developed in our laboratory for studying dendritic growth under terrestrial conditions we plan to design an autonomous version for use aboard the STS. Basically, dendritic growth will be "seeded" into quiescent melts at a variety of supercoolings under microgravity conditions. The axial growth speed and micromorphology will be carefully measured and correlated to the acceleration level. Under strict diffusion control, the axial growth speed, V_d, of a pure material should be given by the expression

$$V_d = \beta\,(\alpha L \Delta S/\gamma C \Omega)\Delta\theta^n, \quad (1)$$

where β is a known constant from theory; α is the thermal diffusivity of the melt; L is the molar heat of fusion; C is the molar specific heat; ΔS is the molar entropy of freezing; Ω is the molar volume; γ is the solid-liquid interfacial energy; and n is an exponent between 2 and 3, depending on the normalized supercooling, $\Delta\theta$. $\Delta\theta \approx (T_m - T_o)C/L$, where T_m is the melting temperature, and T_o is the supercooled temperature of the melt prior to seeding the dendritic growth.

Equation (1), which is derived from diffusion theory and interfacial dynamics, implies that the growth rate is a unique function of the supercooling and is completely predictable for a given material. Unfortunately, the physical constants appearing in Eq. (1) are usually not all known well--especially γ, the interfacial energy. These constants are, however, all carefully measured for succinonitrile (SCN), a BCC organic crystal with the molecular formula $NC(CH_2)_2CN$. SCN is a transparent material with a convenient melting point (58.1°C) and one which is easily purified by zone refining. Ground-based studies on SCN have been conducted by the author over the past six years. Specifically, the observed dendrite axial speed V has been carefully measured as a function of supercooling. Figure 4.3 shows the ratio of the observed speed, V, to the speed, V_d, calculated from Eq. (1). We note that the ratio of these quantities is close to unity from slightly above a supercooling of 1° to about 5°. Below supercooling of about 1°, the ratio rises in accordance with the approximate relationship that $V/V_d = \Delta T^{-1/3}$. At a supercooling of 0.1 K, the ratio V/V_d has increased by more than a factor of four from unity. Clearly, convection effects dominate dendritic growth at small supercoolings, and reducing gravity should profoundly influence the growth kinetics and morphology at small supercooling, i.e., $\Delta T < 1$ K.

A successful microgravity experiment should permit a definitive test of dendritic crystal growth theory at small supercooling. Such information would be of value scientifically because of the current emphasis to gain a deeper understanding of dendritic pattern formation. Specifically, work by Langer, Muller-Krumbhaar, Koplik, and other theorists is precisely directed at uncovering the fundamental mechanisms of dendrite growth. See news article in Science, June 8, 1984, page 1085. Microgravity experiments on dendrite kinetics should permit slowly growing dendrites of relatively large physical dimensions to grow under pure diffusional control, without the influences of convection present. Although, as demonstrated in Figure 4.3, almost pure diffusion control does occur under terrestrial conditions, but its range is restricted by convection (below about 1.5° supercooling) and by interface molecular attachment kinetics (above about 5° supercooling). In fact, the resolution of morphological details become almost impossible above about 2° supercooling, because the tip dimensions are well below 1 µm, and the tip speed is high. Work by U. Lappe at the Kernforschung Institute, Julich, West Germany, also corroborates the loss of time image resolution above about 2° supercooling. The result of this situation is that there is too small a range of supercooling over which both diffusion control and morphological resolution are possible, and consequently little assurance exists that the theory of dendrite growth can ever be tested adequately by Earth-based techniques. The development of a successful flight experiment would certainly extend the range of accessible supercoolings down to much lower levels by reducing convection. This should permit achieving one order of magnitude of supercooling for critically testing the predictions from theory. To date, other attempts to reduce convection (for example, increasing viscosity or reducing the length scale) have not proven to be feasible. Thus, reducing gravity is perhaps the most straightforward approach to achieve a pure diffusive dendritic crystal growth condition over relatively long durations for careful quantitative assessment.

EXPERIMENTAL DETAILS--GROUND BASED

The techniques for simultaneous kinetic and morphological measurement of dendritic growth were established in 1976 (Met. Trans. 7A, pp. 1747-1759). Basically, a large volume, ca. 30 cm^3, of material is melted and uniformly supercooled in a bulb or observation chamber. The entire chamber is immersed within a precision thermostatic bath, which maintains a steady temperature to ± 0.001 K. A seed crystal is released by de-energizing a control heater, which permits the growth of the seed into a capillary orifice, which terminates at the center of the observation chamber. Figure 4.4 is a schematic of a simple observation chamber used for achieving "free" dendritic growth. The term "free" implies that the path of crystal growth is unimpeded or influenced to any significant degree by the walls of the observation chamber. Interaction of the growth with container walls invariably leads to complicated growth forms under poorly defined transport conditions.

Once the dendrites grow a few millimeters away from the capillary orifice, they achieve a steady-state axial growth rate and a stable steady tip configuration. The displacement and time of the tip position yield the growth speed at steady state. The observation is terminated either when the dendrite reaches the walls of the observation chamber or, under slow growth conditions, when sufficient displacement-time data have been gathered. Many other technical details enter the experimental procedure, such as seed orientation, growth orientation relative to the local gravity vector, lighting conditions within the thermostat, camera angle and optics, thermometry, sample purity, or alloy composition. This method is capable of producing dendrite growth velocity data accurate to ±2 percent, including all random and systematic errors. The independent variables include the supercooling and alloy composition for binary melts.

For a pure material, which we will consider initially as a candidate for a flight experiment, only one independent variable occurs, viz., the supercooling, $\Delta T = T_m - T_o$. Inasmuch as ΔT results from the difference of two independent quantities, the accuracy to which ΔT may be set experimentally is the error sum on T_m and T_o. T_m is an intrinsic property of the material but is approached (asymptotically) as the purity approaches 100 percent. T_o is an arbitrary temperature below T_m, which is set and maintained with the thermostatic bath. Clearly, the accuracy of ΔT then depends on purity as well as on control of temperature (spatial and temporal) and the limits of error of its measurement. The experimental details have been carefully worked through under normal laboratory conditions. Temperatures may be set to an accuracy of ±0.003 K for the value of both T_m and T_o, although using differential techniques ΔT can be set meaningfully to 0.001 K. This represents a relative error or only 1 percent on a supercooling of 0.1 K. Since velocity depends on ΔT^2 at low supercooling, the measuremental precision in both velocity and temperature is well matched. As the supercooling is reduced below 0.1 K, the uncertainty in ΔT increases above that for the velocity measurements.

Pure diffusional crystal growth requires establishment of an undisturbed thermal field about the advancing dendrite. This "adiabatic" condition is easily met for rapidly growing crystals, where the diffusion length for heat is small. The diffusion length is α/V_d, which by virtue of Eq. (1) increases as ΔT^{-n}. Again, at small supercooling, the diffusion zone length will expand as ΔT^{-2}. At a speed of about 5 μm/sec, the diffusion length will approach the radius (2 cm) of the observation chamber, and some latent heat will flow out into the thermostatic bath. This violates the "adiabatic" condition and introduces some error into the experiment. Succinonitrile will achieve a velocity of 5 μm/sec at a supercooling of about 0.2 K. Thus, as the supercooling is reduced below about 0.2 K, some nondiffusive transport will occur, unless the sample volume is increased. Increasing the sample volume drastically increases the thermal time constants of the apparatus and soon becomes impractical. Clearly, a lower limit of about 0.1 K is set by practical specimen dimensions. This should provide an order of magnitude in supercooling (0.1-1.0 K) for a microgravity experiment. As explained earlier, under terrestrial conditions convection sets in at about 1 K, so microgravity conditions should provide an experimental "window" of opportunity for observing pure diffusive dendritic growth over about two orders of magnitude in the velocity.

EXPERIMENTAL DETAILS--FLIGHT

1. Thermostat: The major requirement for a quantitative study of dendritic growth in the microgravity environment is the development of an autonomous thermostat capable of providing a programmed thermal cycle to millikelvin precision with low power consumption. This development requires considerable departure from conventional observation thermostats, of the type shown in Figure 4.5. In conventional thermostats, the heating and cooling are performed in the observation chamber itself, with a mechanical impeller used to provide rapid stirring close to the sources of heating and cooling. The conventional and prototype flight thermostats use Tronac precision temperature controllers. A Tronac controller maintains temperature stability by adding heat to the thermostatic bath at a rate that equals the loss from all sources. This "make-up" heat is generated by the ohmic heating of a tubular stainless steel coil that has a resistance of approximately 0.15 Ω. Cooling is accomplished by flowing cooling fluid through the tubular heating coil. The heat added just compensates for the heat lost through the walls of the thermostat and through the cooling coil. Any difference between the set-point and the actual temperature sensed by the thermal probe is electronically integrated, and the amount of power applied to provide the make-up heat is proportional to the voltage of the stored charge in the error

integrator. Although constancy of temperature during steady-state operation is important, achieving spatial uniformity is of equal concern here. Temperature uniformity depends on the physical configurationof the thermostat and on the nature of the mixing of the bath fluid. The faster the fluid flows past the coiling coil, the better is the uniformity of temperature achieved. Transient conditions must also be considered to ensure good dynamic performance. The types of transient changes likely to be encountered include (1) changes in cooling rate, (2) sensing of local temperature inhomogeneities, (3) drift in line voltage, (4) thermal drift of the control unit itself. We briefly discuss these transients. Severe thermal transients may result when the cooling rate through the heater coil changes, especially when such changes occur with a time constant less than that of the controller. A strategy must be developed to avoid rapid changes in the temperature of the cooling fluid or its flow rate. Thermal inhomogeneities which pass the thermal probe may cause the controller to apply a correction erroneously when, in fact, the average bath temperature is correct. The most effective manner of avoiding such problems is to ensure rapid, turbulent, mixing, thus reducing the volume and magnitude of the spatial inhomogeneities. Set-point drift may be expected when the ambient temperature changes, thereby resulting in undesirable changes in the potentiometers, which are used to balance the electronic bridge circuits. Similarly, line-voltage fluctuations can raise or lower the power delivered by the controller to the thermostat. Both ambient temperature and line-voltage changes can be sensed in real time, their effects anticipated, and compensating actions taken by an appropriate microprocessor. Such a microprocessor would have to be incorporated in the final design of the control circuit for an autonomous experiment.

We have conceived and built a prototype autonomous thermostat, the major components of which are shown schematically in Figure 4.6. The thermostat departs from conventional design by using a control chamber separate from the observation thermostat. The unit constructed for test purposes uses a 10-liter thermostat working in conjunction with a 2-liter control chamber. The configuration shown in Figure 4.6 leads immediately to a volume (and weight) reduction of about 45 percent from conventional single-chambered units. The separate control chamber permits a relatively unobstructed view of the object of control and the opportunity for flexible lighting arrangements for optimal photomicrography. Mixing is accomplished by turbulent "jetting" of the thermostatic fluid into the observation chamber. A high flow rate is needed to achieve good mixing over most of the volume of the thermostat. We have shown that 10 liters/min provides reasonable uniformity without excessive pumping power. The thermostat thereby experiences a complete "turnover" of its fluid each minute. This time period is acceptable, provided that heat losses through the walls are kept to just a few watts. Wall insulation of 6-8 cm of closed-cell polystyrene foam board is adequate to achieve such low losses at a control temperature of about 60°C. The other major departure from conventional thermostats is our use of gas cooling to provide internal cooling of the mixing chamber. Our prototype uses a compressed-air cooling loop at 20 psig. Air is pumped through a finned aluminum heat sink to cool it prior to flowing through the cooling coil. The waste heat is discharged to the laboratory atmosphere. The prototype constructed shows promising overall performance. Input heating to the control coil has been reduced 75 percent from conventional application to about 25 W, of which only 5-8 W are lost through the walls of the observation thermostat. The main power requirement is the air compressor, which consumes about 300 W. An efficient turbocompressor would reduce this power requirement to more reasonable levels. We have achieved long-term working temperatures near the center of the thermostat that are stable to ± 0.75 mK. The control precision deteriorates away from the central region as the thermostat walls are approached, but design improvements to overcome this characteristic should be possible with further major developments of the prototype.

2. <u>Growth Chamber</u>: Several growth chambers have been constructed to check the possibility of achieving oriented crystal growth prior to kinetic and morphologic measurements. The advantage of oriented crystal growth is that if the dendrites will grow in a [100] crystallographic plane that is coincident with the focal plane of the object field of the camera, then sharp focus and clear profile views of the entire dendrite should be possible. In addition, stereographic

corrections to obtain true length, true orientation with respect to the gravity vector, and true velocity would be either unnecessary or small in magnitude. The two viewing cameras would then give two orthogonal plane views of the growing dendrites, which would be ideal for analysis. We have found that extreme care in handling the growth chamber is required to maintain the proper crystal orientations, once established. Several seeding devices are being studied. The most promising appears to be a seed chamber connected to a helical "choke" or grain selector which connects to the main observation chamber via a flexible channel. The "choke" eliminates undesirable crystal orientations, whereas the flexible channel permits small goniometric corrections to be applied to bring the {100} growth plane and <100> growth directions into coincidence with the object plane of the photographic system.

A second area requiring developmental activity of the growth chamber is achieving a sufficiently rugged design to withstand the rigors of flight aboard the STS. Clearly, the all glass laboratory models typified by Figure 4.4 are unacceptable for meeting these demands. We have designed and constructed a prototype launch-compatible crystal growth chamber, shown schematically in Figure 4.7. Clearly, the ruggedized growth chamber is more complex than the conventional laboratory design (cf. again Figure 4.4) and is composed of diverse materials, namely, glass, elastomers, metals, and welds. The stringent purity requirements discussed earlier can only be met and maintained over time by a judicious selection of materials for the crystal growth chamber. We are now carrying out long-term compatibility tests between some candidate materials and the model substance, succinonitrile. The unit we constructed as a prototype was machined from brass and nickel plated for corrosion resistance. A series of critical tests for presetting the growth orientation and recycling the growth chamber in a "hands-off" manner is being planned for August 1984. As the results from these tests and the compatibility among materials becomes clearer, additional chambers will be built and evaluated for flight application.

3. <u>Photographic Systems</u>: The overall photographic system envisioned in our conceptual design differs from that used in the normal ground configuration. On the ground, we use stereo microscopes with a tri-occular

camera port for recording images in the magnified condition. Working distances are relatively long, but become somewhat restricted as the magnification rises above about 10X. Figure 4.8 shows the preliminary concept for the camera locations around the thermostat. We plan to use long working distances with subnormal magnification on a 16-mm film format, instead of magnified images on a 32-mm or Polaroid film format. Clearly this requires much more photographic enlargement of the flight data as a postflight data processing operation, and the use of fine-grained films capable of recording high-resolution images. We plan to continue to test other films types and various developing methods to achieve the required resolution of about 300 lines/mm. It is important to resolve the dendritic type structure and the side branches with sufficient clarity that accurate fitting of these images with theoretically derived shapes is possible. It remains an open technical question as to what is the best compromise between the ease of long working distances and achieving the requisite resolution. At present, we are estimating the capacity of photographic requirements on the basis of 16-mm fine-grained film in 65-foot magazines (2000 frames). We anticipate the need for 1 frame per mm of crystal growth, or about 20 frames per growth cycle. A set of experiments involving 20 cycles would be reasonable (see Section 4 below), and this protocol would consume 400 frames per camera. If the images are formed at an original magnification of 0.25X, then photographic enlargement of 20-40X will be needed to produce analyzable images. The growth rates from 2 per minute to 12 per minute should suffice. This requirement is easily met with conventional cameras.

4. <u>Experiment Flight Profile</u>

The flight experiment requires two major phases for its execution, (1) "warm-up" and (2) data-acquisition cycles. We estimate that the thermostat and its contents can be brought up to the operating temperature range in approximately 8 hours. The warm-up phase is based primarily on the time a "cold" thermostat, at say 25°C, can be brought to the operating temperature range of 60°C. Once the energy is delivered to the thermostat, then relatively small modulations in the thermostat's heat content are needed to cycle the crystal growth chamber and acquire data. Specifically, we can anticipate the following thermal schedules:

		ΔT = 0.1 K	ΔT = 1.0 K
A.	Raise to 60°C from prior cycle	30 min.	30 min.
B.	Hold to totally melt chamber	30	30
C.	Achieve desired supercooling	60	60
D.	Hold for spatial uniformity	30	30
E.	Seed and grow dendrites	60	5
F.	Total data cycle	210 min.	155 min.

We see that data cycles will take approximately 3 hours each. If the decade of supercooling (0.1-1.0 K) is divided into 10 specific supercoolings (perhaps with a parabolic distribution to achieve uniform velocity increments) then for two experimental runs per supercooling we require at least 60 hours for all the data acquisition cycles. The two phases (warm-up plus data cycles) therefore will require about 68 hours of orbital time. Should the microgravity environment be seriously altered by such events as thruster firings or other activities aboard STS that require high accelerations, then the data acquisition cycles can be interrupted until a "quieter" phase occurs. Indeed, the entire autonomous sequence of events must be microprocessor controlled and be capable of "holding," pending unanticipated orbital events. As long as the seed crystal is protected from melting (perhaps by locating the seed outside of the thermostat) then the data acquisition cycles can be stopped or started arbitrarily. Even assuming that a serious condition interrupting the acquisition cycles does not occur, we would still require monitoring local accelerations on the growth chamber with a three-axis accelerometer.

The required experimental phases can be accomplished with only a modest requirement for on-board electrical power and total energy. Based on ground tests performed in our laboratory, and using reasonable "guesstimates" otherwise, we find the following power demands:

Thermostat	40 watts (average)
Pump	70 " "
Circulator	50 " "
Electronics	40 " "
TOTAL	200 watts (average)

This power schedule will require the total consumption of 13.6 kWH over the entire flight experiment. We do not anticipate difficulty in meeting these requirements for power and energy.

5. Summary of Flight Requirements

- Develop an autonomous millidegree thermostat for orbital application over periods of approximately 3 days
- Develop a method of repetitive crystal seeding for orientational control of the dendtritic crystals
- Design a launch compatible crystal growth chamber
- Provide data acquisition for:
  - Temperature
  - Interface Velocity
  - High-Resolution Photomicrography
  - Acceleration
- Flight hardware must satisfy design limits on:
  - Temperature
  - Power
  - Weight
  - Volume
  - Flight Safety

PREFLIGHT/POSTFLIGHT ACTIVITY

The flight experiment will be designed to be a self-contained autonomous package, requiring minimal involvement of crew and/or payload specialists. The various subsystems such as the crystal growth chamber, thermostat, optical system, and recording systems will be interrelated through a master control system consisting of a microprocessor and a sensor array. The sensors will provide information regarding the condition of the various subsystems, especially regarding temperatures, flow rates, logic configuration, acceleration, ambient data, time, consumables, safety, etc.

Prior to flight, the experiment must be placed in a stand-by mode, with the seed orientation carefully checked for optimal growth and illumination for microphotography. In addition, the condition of the system will be such that lift-off accelerations will not overstress the apparatus. Once in orbit, the system will be activated by a crew member. The microprocessor will interrogate the critical assemblies for verification that everything is in proper condition for thermostat heat-up and eventual melting of the crystal growth chamber. Over the course of the next 8 hours, final checks will be made so that data cycling phase can begin.

After completion of the pre-programmed thermal cycles, and recording the relevant optical, thermal, time, and acceleration data, the microprocessor will secure the experiment by cutting power to the thermostat and allowing the crystal growth chamber to freeze totally over the remainder of the STS mission. Film and electronically recorded data will be returned for development and replay, respectively. We expect a sequence on in situ photomicrographs showing the progressive growth of similarly oriented dendritic crystals growing freely within the chamber. Timing signals will permit a correspondence to be established between the data channels recording the supercooling and the growth photographs. Acceleration data will be similarly coordinated to determine any influence of residual acceleration or its power spectrum on the growth process. Of prime interest will be establishing the scaling behavior of dendrites growing at low supercoolings with comparisons to data taken under terrestrial conditions. For example, we will analyze the photomicrographs for the tip radius, tip shape, sidebranch amplification, axial growth speed, symmetry, and overall correspondence with predictions based on thermal diffusion theory. Deviations from pure diffusion behavior will be analyzed against the knowledge of in-flight accelerations or other departures from the planned experimental conditions.

It is planned to recover the crystal growth chamber and perform postflight melting-point determinations to verify that the sample has remained uncontaminated during the flight. This is a critical check, since the supercoolings programmed into the microprocessor depend on the melting point remaining at 58.082°C. This check could in principle be obtained during flight by appropriate internal thermometers within the growth chamber, but whether the added complexity of internal sensors, hermetic seals for the lead wires, and so on, are compensated for adequately by this added assurance remains uncertain at this time.

PROGRAM STATUS

The current program, which is supported as a NASA grant (NAG3-333), is in the second year of funding under contract administration and scientific liaison with the Lewis Research Center, Cleveland, Ohio. The ground-based research program and the current flight-oriented conceptual design activities are considered distinct but related tasks. Hence, over the last 1 1/2 years, two task "streams" have been pursued which have quite different aims. The work discussed here only includes the flight-oriented design tasks, whereas the ground-based research has been delineated for continuation under grant NAG3-333.

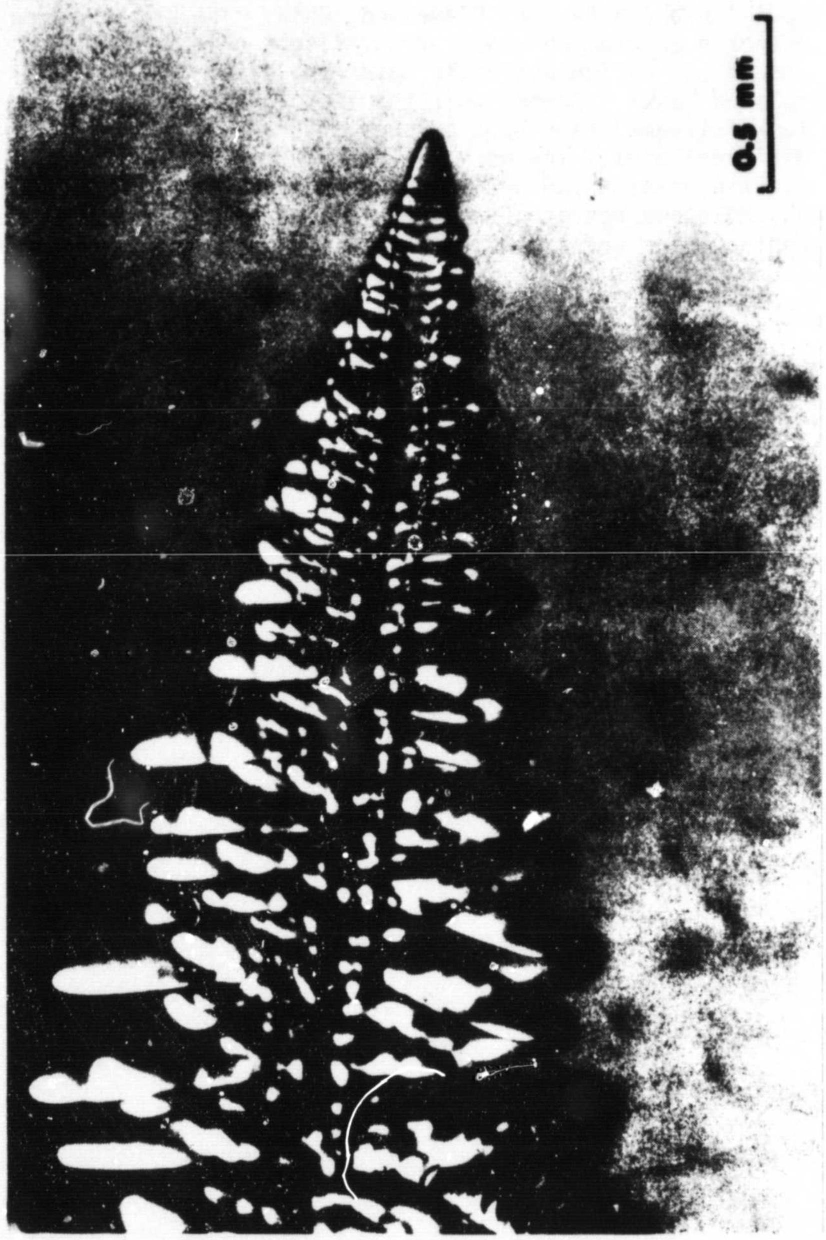

FIGURE 4.1. Anisotropic pattern of dendritic solidification.

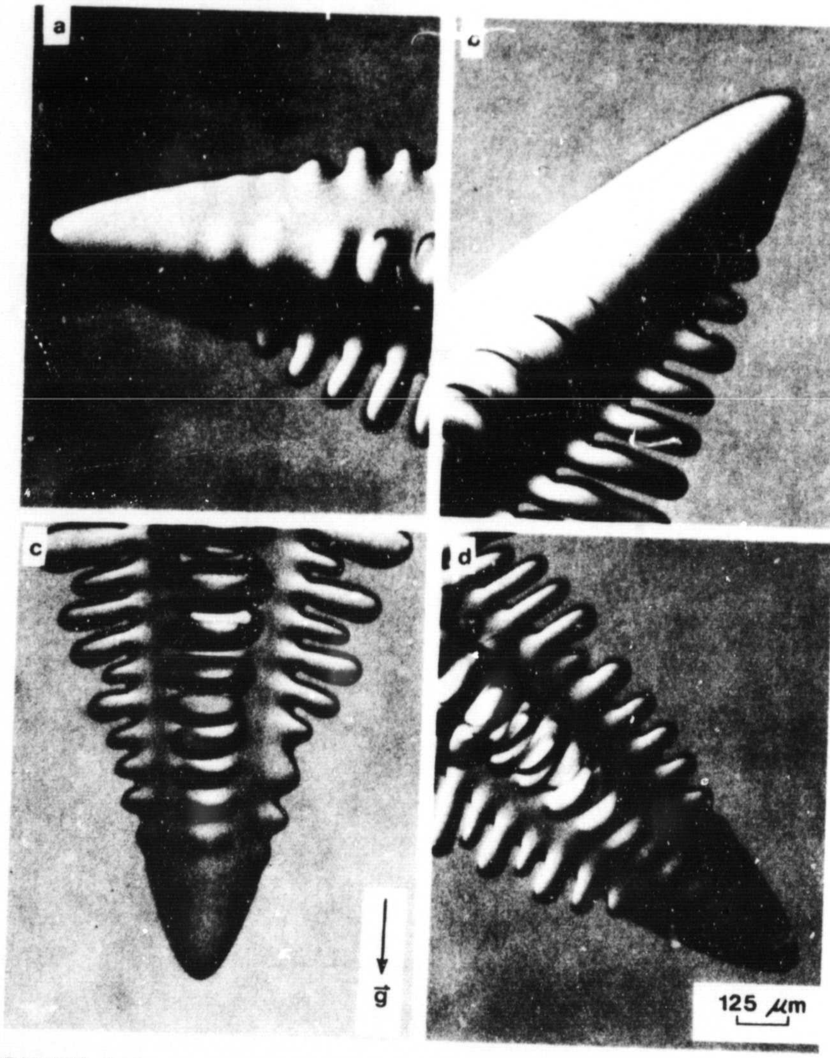

FIGURE 4.2. Influence of spatial orientation of dendritic growth axis on tip morphology.

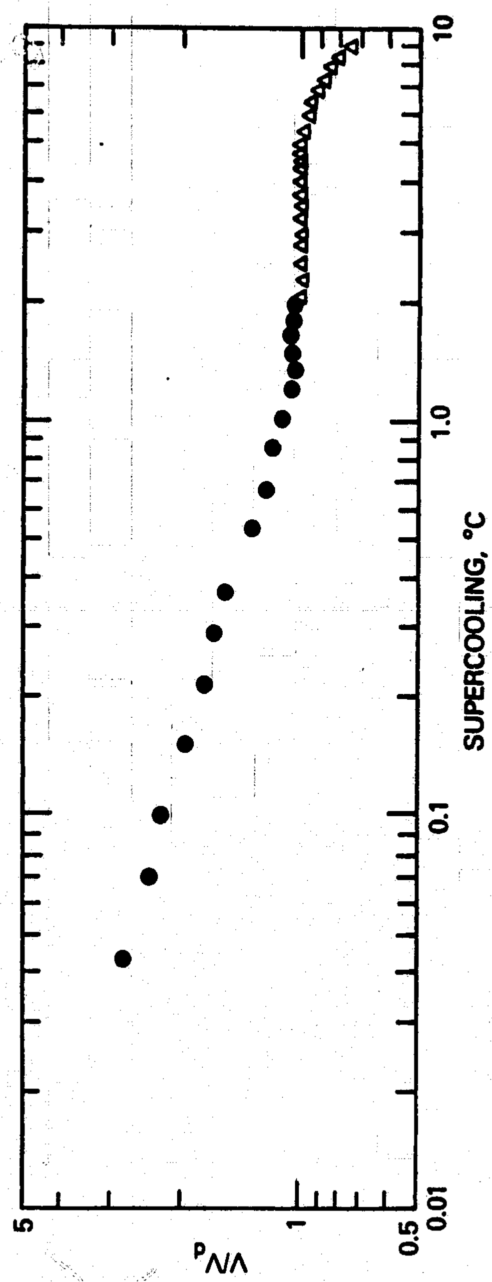

FIGURE 4.3. Dendritic kinetics (temperature vs. volume change).

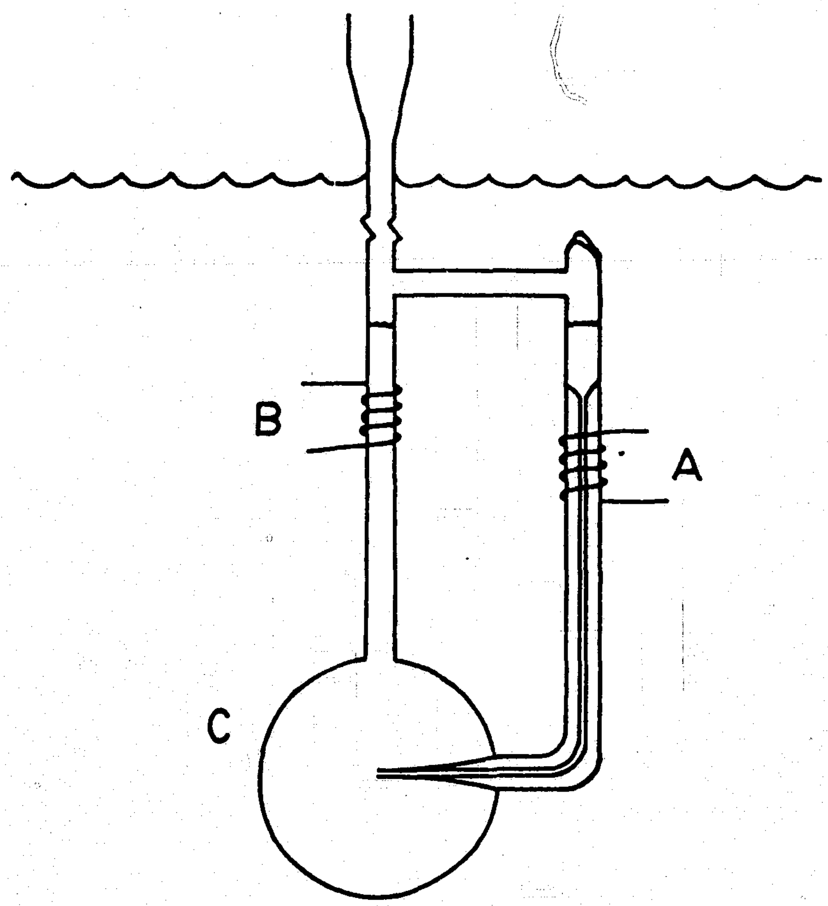

FIGURE 4.4. Schematic of observation chamber used for achieving "free" dendritic growth.

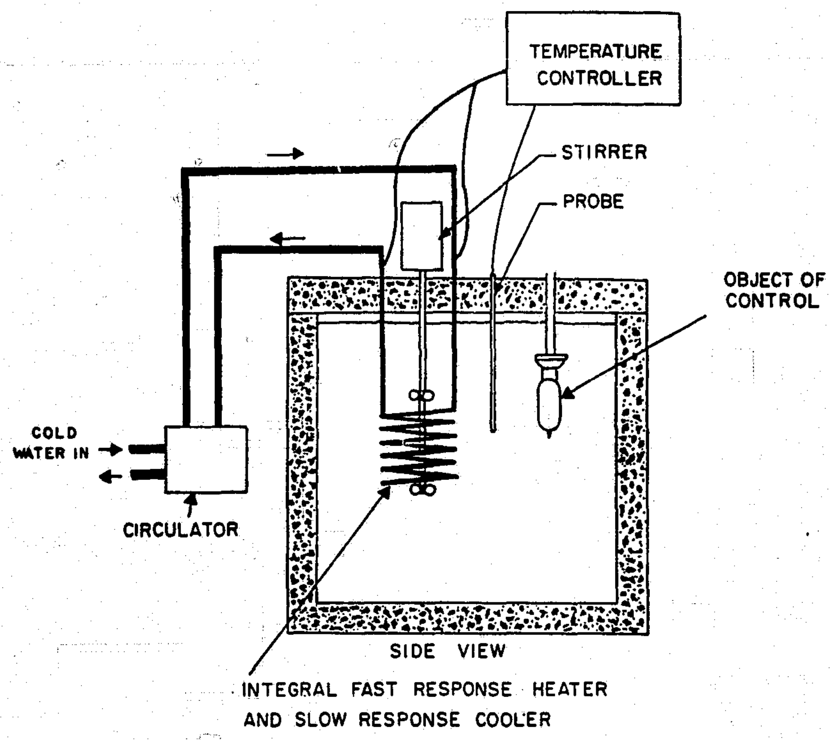

FIGURE 4.5. Conventional observation thermostat.

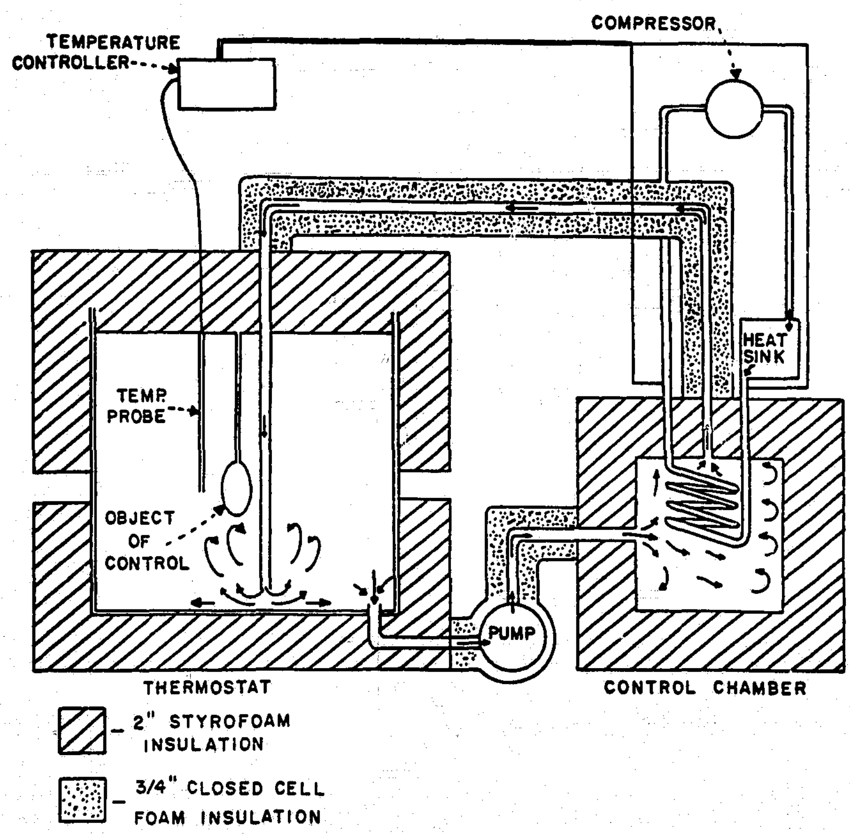

FIGURE 4.6. Schematic of thermostat for dendritic growth.

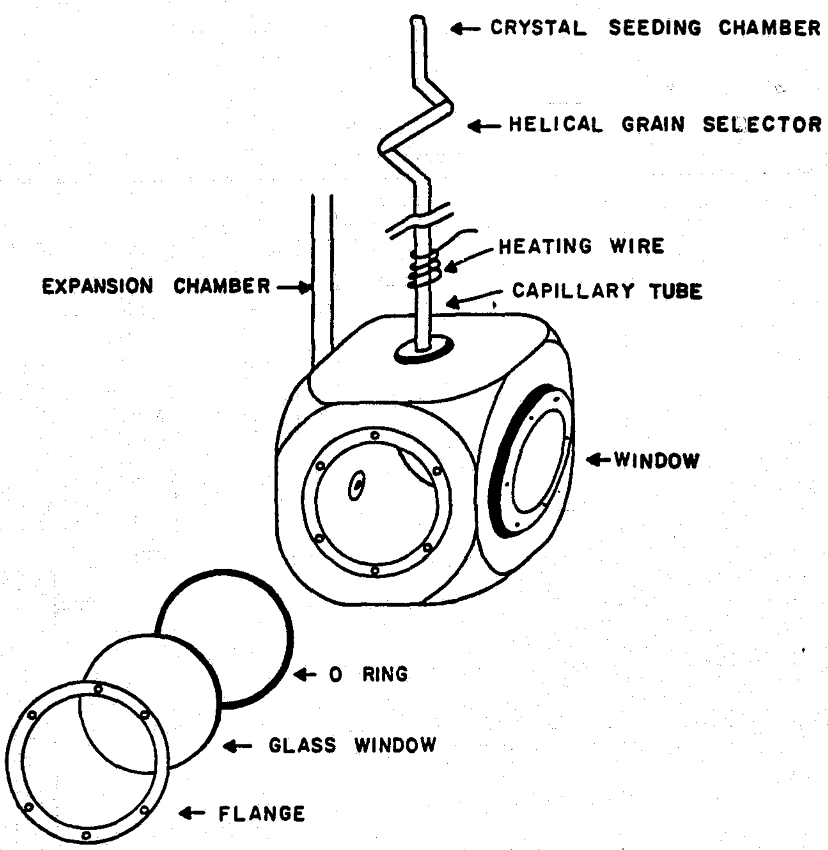

FIGURE 4.7. Prototype of crystal growth chamber (launch compatible).

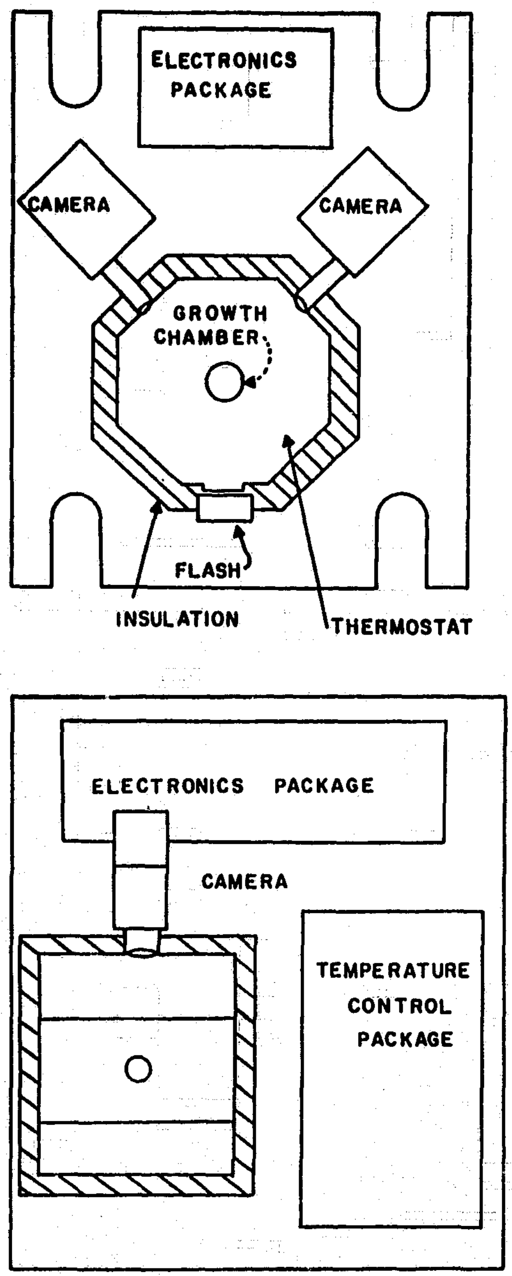

FIGURE 4.8. Preliminary concept for camera location around the thermostat.

5. ELECTRONIC MATERIALS RESEARCH IN THE MICROGRAVITY SCIENCE AND APPLICATIONS PROGRAM

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In the recent past, many of our nation's technological advances have been based, directly or indirectly, on improvements in our ability to process electronic materials; therefore, if one accepts the premise that advances in the science and technology of electronic materials are key elements to advances in the nation's technological future, then the questions relevant to a space research program become: (1) What materials systems are technologically important? (2) What are the technology-limiting problems? (3) What aspects of a space research program are likely to have an impact on solving these problems?

Concerning the first question it is well established that silicon (Si)-based devices are the foundation of the world's electronics industry as it exists today, and will remain the foundation for the foreseeable future. Other electronic materials of a steadily increasing technological importance are binary semiconductors such as GaAs, InP, CdTe, and others and alloyed ternary semiconductor crystals such as HgCdTe, GaAlAs, and PbSnTe. In addition, new classes of electro-optical and detector materials have increasing significance for futuristic, general device applications and current strategic applications.

The answer to the second question is not quite so easy. In bipolar devices, both yield and performance are affected noticeably by the homogeneity of the substrate material. In charge coupled devices (CCD's), the resistivity uniformity is very important. The influence of materials deficiencies, including process-induced contamination becomes increasingly problematic as the degree of device integration increases. In advance VLSI technology, which makes use of larger diameter wafers, the mechanical properties of the substrate become critical. In silicon, these mechanical properties are highly dependent on the density, distribution and state of the incorporated oxygen. A rigorous determination of the relationship between growth conditions and the state and distribution of incorporated oxygen and other impurities is inhibited by subtle effects arising from temperature fluctuations in the melt caused by gravity-induced convection. In compound semiconductors, one of the most important problems is the control of stoichiometry. The electronic properties of these materials appear, to a large extent, to be controlled by the relationships between stoichiometry and heat and mass transfer conditions prevailing during solidification. Gravity-induced convective flows constitute major perturbations on these conditions. In growth of multicomponent alloy systems, the situation is further complicated by the presence of more than one major component. Thus, without being specific as to the nature of the technology-limiting problems, it is established that the presence of buoyancy-driven convection impedes optimization, as well as understanding, of the solidification process. This lack of a detailed understanding complicates the establishment of correlations between growth conditions and the pertinent chemical and structural characteristics which directly affect the relevant electrical and optical properties that govern device performance and manufacturability. In the final analysis, it must be stated that gravity-driven, convective effects prevent the quantitative elucidation of the crystal growth process by which electronic materials are produced. This may be the barrier which prevents us from producing materials in which property requirements for projected device technology are met.

As reported by the STAMPS committee and as implied in the preceding paragraphs, the potential for long periods of reduced gravity is a facet of a space environment that will likely have the greatest impact on solving problems faced by the electronic materials community. The effects of reduced gravity which could be advantageous for the science and technology of electronic materials growth are: (1) an elimination or considerable reduction in buoyancy-driven convection, (2) an elimination or reduction of sedimentation, (3) a reduction in hydrostatic pressure, and (4) the possibility for containerless processing of large samples. In addition the reduced gravity environment could lead to new/novel growth apparatus and configurations. In order to utilize the potential of reduced gravity to gain a better understanding of solidification processes, experiments of several types can be identified. These include, in no priority order: (1) measurements of thermophysical properties such as diffusion coefficients, (2) studies of growth processes in materials with a miscibility gap where sedimentation of settling affects phase distribution, (3) containerless processing studies which allow reduction of heterogeneous nucleation sites and contamination from the crucible, (4) experiments to test the assumptions necessary in theoretical models of inherently complicated systems, (5) production of standards of electronic materials in which the properties are limited only by thermodynamic considerations, (6) studies of other convective driving forces such as surface tension which are normally masked by gravity-driven convection, (7) studies of critical point phenomena which are severely affected by density gradients which give rise to buoyancy driven convection, and (8) the possibility of producing limited quantities of materials with improved properties for special applications. A program in the space environment should concentrate on addressing fundamental problems leading to advances in materials science which, historically, have lead to subsequent advances in technology. In parallel, the program should maintain a vigil of technology-limiting problems in the commercial sector and explore the possibility of applications in the space environment which would directly impact these commercial limitations. This aspect of the program would include establishing a broad data base of past and projected scientific results, publicizing current efforts funded by the government, solicitation of inputs from the commercial sector for programs directly related to their specific requirements, and long-term planning which includes the capability for space processing of bulk materials in commercially required dimensions.

This paper will present a brief review of past experiments in space related to electronic materials, a summary of current European experiments related to this discipline, and a broad overview of experiments in crystal growth from the vapor phase, from the melt, and from solution.

HISTORICAL BACKGROUND

Electronic materials research has previously been done on Skylab and Apollo-Soyuz. The hardware for these experiments is described in by Naumann and Herring.[1] The results of these experiments have been summarized by Naumann[2] and are repeated here.

Skylab Experiments

Skylab offered the first opportunity to perform space processing experiments in a dedicated facility, although considerable constraints on apparatus and experiment development were imposed by the available development time and onboard power.

Crystal Growth Experiment

Some potential advantages of growing crystals in a microgravity environment were demonstrated by A. F. Witt et al. (MIT). Samples of InSb doped with Te (10^{18} atoms/cm^3) were grown on Earth by the conventional Czochralski technique. These were melted back over a portion of their length in space using a gradient furnace and allowed to regrow by lowering the furnace temperature. After a brief growth transient required to achieve steady-state growth conditions, very uniform dopant concentration was obtained along the axial direction. This indicates that diffusion-controlled growth conditions were indeed achieved. Also, the space-grown sample was completely free of the dopant striations usually seen in Earth-grown samples. These striations originate from growth-rate fluctuations which are generally believed to be associated with convective flows in the melt.

A novel crystal growth experiment, developed by H. U. Walter (University of Alabama, Huntsville), involved a quasi-containerless growth of InSb. A portion of an Earth-growth single crystal was melted back as was done by Witt and Gatos, except the crucible was larger than the rod diameter so that the melt could form a spherical shape. As the melt was solidified in the gradient freeze mode, an unanticipated combination of effects from volume change on freezing and contact angle between the melt and solid formed a tear-drop shape which ultimately encountered the end of very smooth crystallographic planes on the unconfined growth portion of the crystal. X-ray topographs indicate that this portion of the crystal is virtually free of defects. There are dopant striations, however, which are puzzling in light of the results obtained by Witt and Gatos. One possible explanation is that an unconfined melt may be subject to Marangoni (surface-tension-driven) convection which gives rise to growth-rate fluctuations similar to those produced by gravity-driven convection on Earth. Also, a melt confined only by its contact with a solid, which in turn is fixed to the spacecraft, will be affected more by small low-frequency accelerations (g-jitter) associated with crew motion than a melt constrained by a container. Oscillations produced by such residual accelerations could conceivably result in striations. Clearly, more research is needed to resolve such questions.

J. T. Yue and F. W. Voltmer (Texas Instruments) grew germanium crystals with various dopants by the same techniques used by Witt and Gatos. The objective of this experiment was to investigate the distribution of dopants in the space-growth samples. The results were somewhat inconclusive because crucible contaminants produced some unwanted doping. Also, the amount of crystal grown in space was not sufficient to achieve steady-state growth conditions. There appeared to be more uniform radial distribution of dopants in the space-grown sample. The Earth-grown control samples seem more subject to random compositional fluctuations, which are probably caused by time-dependent temperature fluctuations associated with gravity-driven convection. The space-grown samples had a more uniform composition in the interior but did show a marked increase in dopant concentration near the surface. The growing crystal in this experiment pulled away from the crucible wall (as did several other experiments) which indicates that the melt may also have had a free surface. This would permit Marangoni flow that could account for the observed radial dopant profile. However, a curved solidification interface imposed by the thermal conditions could also account for the observed effect.

The InSb-GaSb alloy-type crystal growth experiment developed by W. R. Wilcox (Clarkson College) did not have sufficiently high thermal gradient to growth rate in the gradient-freeze furnace to stabilize the growth interface against breakdown from constitutional effects, and polycrystalline growth resulted.

H. Wiedemeier (Rensselaer Polytechnic Institute) flew a closed-ampoule vapor crystal experiment in which iodine was used as a chemical transport agent to grow GeSe and GeTe. The crystals grown in space were generally larger with a more compact growth habit and smoother facets than the control samples grown on Earth. They also exhibited a much greater perfection as determined by etch pit analysis. A major surprise in the experiment was the fact that the total material transported from source to seed was substantially greater in space than when the growth ampoule was in the vertical stabilizing configuration (hot over cold) on Earth. The reasons for this anomalous transport rate are not clear; although, we now realize that considerable convection takes place even in a thermally stable system because of radial thermal and solutal gradients caused by wall effects. It is conceivable that convective cells driven by these effects actually inhibit diffusive transport in a thermally stable configuration in Earth's gravity.

Apollo Soyuz Experiments

The Apollo Soyuz Test Project (ASTP) offered an opportunity to repeat some of the more interesting Skylab experiments and to accommodate several new investigations. Unfortunately, the time to prepare these experiments was short and the resources offered by ASTP in terms of power, weight, volume, and on-orbit time were much less than on Skylab. Therefore, the experiments on ASTP in many cases were performed under more primitive conditions than on Skylab.

Crystal Growth Experiments

Witt and Gatos repeated their Skylab experiments with Te-doped germanium in a new furnace that was equipped with Peltier pulsing for interface demarcation. This allows the interface shape and position to be correlated with time and gives a direct measure of growth rate. Contrary to the Skylab experiment, uniform dopant distribution was not achieved and there were indications that asymmetries in the furnace module produced unequal radial heat flow. Similar effects may also have occurred in their Skylab experiment but were not so apparent because of the difference in segregation coefficients and the sensitivity of the technique used on the ASTP experiment.

Wiedemeier repeated his Skylab experiment using different materials and transport agents. $GeSe_{0.99}Te_{0.01}$ was grown using GeI_4 as the transport agent. Other samples included $GeS_{0.98}S_{0.02}$ with $GeCl_4$ and GeS with $GeCl_4$ and Ar. The Ar was added to vary the total pressure independently of the partial pressure of the transport agent. The results were similar to those obtained in Skylab, i.e., significantly enhanced growth rates as compared with what was then believed to be diffusion-controlled growth on the ground and improvements in growth habit and perfection suggestive of quiescent, diffusion-controlled growth conditions in the space-grown crystals.

European Experiments in Space

At a recent conference on Spacelab I results,[3] Eyer and Nitsche reported on the float zone growth in space of silicon in a double ellipsoid furnace. They were only able to obtain growth in one sample that showed striations which were similar to those found in samples grown in 1 g. No other characterization studies were made on the sample. They attributed the striations to Marangoni convection. An experiment by Benz on solution growth of GaSb was cut short owing to technical problems, but in the small amount of growth obtained, there was a region with no striations. A solution growth experiment by Nitsche on CdTe was also cut short by technical difficulties, and the sample was fractured by internal stresses due to the quenching effect when the furnace prematurely shut down. Kolker reported on the directional solidification of a silicon sphere. Although the surface of the sphere was accidentally coated by a black substance, which he attributed to an oil contamination from the furnace, he still observed striation patterns which he also attributed to Marangoni flows. Rodot did experiments on three PbTe samples in a gradient freeze furnace. The ampoule for the unseeded sample was ruptured during growth by what Rodot believes was crew activities. Although she reports the seeded samples grown in space appear to be better than those grown in a 1-g environment, she has not completed her analysis. Recommendations by the rapporteur at the conference were that Europeans needed theoretical support for space experiments and they need to do more ground-based analysis and characterization of the samples. His criticism was that by limiting the analysis to a search for striations, they neither discovered any new phenomena that may be occurring nor gained any understanding of the relative magnitudes of the convective driving forces in a low-gravity environment. In results from sounding rocket experiments in the German TEXUS program, Carlberg reported that in Ge(Ga, 1.5×10^{18} cm^{-3}) interface demarcation and spreading resistance measurements indicated a near diffusion control distribution of dopant and no striations in the low-gravity samples. Walter, in a different TEXUS experiment, started solidification on the ground and showed that striations were present in the sample under acceleration, but the flow apparently stopped or became laminar when the system was in low gravity as evidenced by the absence of striations. In summary, it appears that while the European program currently has more flight opportunities, the scientific base for their program is not so strong as for the U.S. program. The Germans will fly the D-1 mission in October of 1985. Most of the hardware related to electronic materials processing on that mission will be the same as on Spacelab 1. Experiments will include new materials such as PbSnTe as well as reflight of those which experienced technical problems on Spacelab-1.

Russian Space Program

The Russians have maintained a strong effort in electronic materials growth in space. While details are sketchy, they appear to have grown a wide range of materials similar to those in the U.S. program, i.e., Ge, GaSb, GaAs, PbTe, SnTe, and HgCdTe. However, the bulk of their experiments have been in Ge and the III-V materials. In general, their results are similar to those obtained in previous U.S. flights.[4] They report a suppression of the gravity-driven convection, near diffusion-controlled distribution of constituents, nonwetting of the ampoule walls, and up to four orders of magnitude reduction in dislocation densities. All of which have profound effects on the electronic properties of these materials.

Up to this time, it appears that the Russian efforts have been devoted to materials of a practical interest using a "fly and try" philosophy in which, "...the fundamental significance of experiments under microgravity is by no means a less serious argument in their favor than their possible practical application." L. L. Regel calls for experiments which are better based in theory and for better hardware for obtaining the data required for postflight analysis. She states, "...in better part of published papers authors are very optimistic about the practical application of the results. According to them, there is good reason to predict future development (even 'violent,' some of them say) of space technology and the organization in space of economically substantiated production of some unique semiconducting materials. Some of the authors are pessimistic in this respect. Still, preference should be given to optimists who strongly believe in future space production of semiconductor materials as well as some others."

In general, the crystal growth experiments performed in space up to the present have not produced spectacular breakthroughs, but they have confirmed that gravity-driven convective flows were sufficiently suppressed in space that other phenomena such as diffusion-controlled conditions and/or Marangoni-driven convection could be studied. A worldwide philosophy seems to have evolved that experiments should only be flown after extensive ground-based testing and theoretical analyses have been carried out. This is exactly the type of program recommended by the original STAMPS committee report and what NASA has achieved during the past five years.

CURRENT MSAD PROGRAM IN ELECTRONIC MATERIALS

To establish a firm scientific base for the furtherdevelopment of electronic materials, programs have been established to study the important processes occurring during the growth of crystals from the vapor phase, solution, and a melt. In addition, studies are being carried out using the quasi-containerless float zone technique. The work in each of these areas has a theoretical aspect, an experimental crystal growing aspect, and a supportive analytical aspect to determine properties and characterization techniques. All three of these aspects are required in order to optimize the subsequent flight experiments. Cooperation among the different subgroups, such as Electronic Materials, Metals and Alloys, and Fluid Dynamics, in the MSAD program allows for an interactive process which utilizes the capabilities of each subgroup to advance the overall program.

Float-Zone Processing

Float-zone crystal growth is a quasi-containerless technique which is especially useful for achieving high-purity, dislocation-free crystals from materials such as silicon which are highly corrosive in the melt. (Interaction between the crucible and melt result in undesired dopants.) Since the weight of the zone is supported by the surface tension of the melt, the zone size is severely restricted on some materials in low-g. In space it should be possible to grow large crystals in systems that have low surface tension to extend the zone length out to the Rayleigh limit, to achieve better thermal control at the growth interface, and to use a symmetrical radiant heating system to avoid the thermal asymmetries inherent in RF induction systems. Whether or not diffusion controlled growth is possible in the float-zone process is a scientific question that has not yet been settled. Vigorous stirring always takes place in this process on the ground because of the combination of buoyancy-driven convection, electromagnetic stirring associated with the induction heating, and surface-tension-driven (Marangoni) convection. The first two flows can be reduced or eliminated by using a quiescent heating system in a microgravity environment. Marangoni flow might possibly be controlled by altering the free surface either by growing a thin solid film such as an oxide that imposes no-slip boundary conditions on the melt, by flowing an inert gas along the surface to dampen the flow, or by introducing a surface-activecontaminant that lowers the surface tension of the melt.

Another scientific issue that must be resolved is the effect of low-level accelerations (g-jitter) on the float-zone process. Float zone is probably more susceptible to such accelerations than any other process because a free fluid surface is in contact with a solid surface which, in turn, is fixed to the spacecraft. Small vibrations that would not affect a fluid in a closed container will cause the floating zone to oscillate, especially if they are near the resonant frequency of the zone. How such oscillations affect the growth is not clear at this time.

Because of the complexity of the flow in a float-zone configuration, theoretical modeling of the entire system is not yet possible. Various investigators are studying different aspects of the problem, such as (1) boundary conditions required for a flat interface, (2) sensitivity of boundary conditions and thermal profiles to variations in materials parameters required for the calculations, and (3) coupling mechanism between shape of the melt domain and the magnitude of the convective velocity induced by variable surface tension, as modified by rotation, and as modified by surfactants. Experimental work under E. L. Kern and G. L. Gill is aimed at developing new float-zone techniques in silicon to minimize melt flow instability and to improve the resulting crystal homogeneity. They have recently reported promising results by using zone refining in a hot wall heater that has a significantly lower thermal gradient. Experimental work to evaluate techniques for dampening Marangoni convection is being done to better understand this phenomenon. S. C. Hardy, of the NBS, has measured the surface tension of silicon as a function of temperature and found the value to be quite sensitive to oxygen contamination.

Vapor-Phase Growth

F. Rosenberger (University of Utah) has extensively analyzed convection in closed tube, vapor crystal growth and finds that viscous interactions between the transporting vapor and the container wall create density gradients that are always convectively destabilizing; hence, it is not possible to achieve diffusion controlled vapor transport in 1 g. Further, he finds that the accepted models for diffusion-controlled vapor transport work as well as they do because they contain errors that self-cancel. Two experiments currently have flight status in this area. H. Wiedemeier is investigating vapor growth of alloy-type semiconductor crystals and will report in detail on this in a later paper. The other flight experiment is the vapor growth of HgI_2 by W. F. Schnepple and L. van den Berg. This is a physical vapor-phase transport process where a temperature gradient is used to drive material from a source to the growing crystal. The scientific objectives of this experiment are first to determine the effects of gravity on crystalline perfection and second to study the vapor transport rates and mechanisms. This experiment flew on Spacelab 3 in 1985 with L. van den Berg as a Payload Specialist. Ground-based tests in the flight hardware have been optimized to obtain the best possible crystals in 1 g, and this experiment is now awaiting flight.

In addition to its importance in understanding fundamental processes, this is also an area in which there is an ongoing, commercial effort by 3M. They have recently proposed an experiment to grow thin crystalline films of materials in order to evaluate the optical properties and crystalline properties of such films grown under purely diffusive growth conditions. They will also compare observed transport rates with the 2-D theory developed by F. Rosenberger. This is an excellent example of NASA's goals for encouraging industrial participation. Being aware of the theory developed by Rosenberger and the flight results of H. Wiedemeier, 3M entered into an agreement with NASA to fly a series of experiments of direct commercial interest to them in which they develop the hardware, and do the ground-based testing. NASA pays for the flight costs and shares in the data on nonproprietary materials flown in the 3M scientific package.

Solution Growth of Crystals

This is an area that has one experiment with flight status. R. B. Lal and co-workers at Alabama A&M University have developed an experiment for the solution growth of triglycene sulfate (TGS), a material used in room temperature infrared detectors. The objectives of the experiment are to develop a low-gravity, solution growth technique, to characterize the growth environment provided by the Shuttle and its influence on growth behavior, and to determine how gravity influences the properties of the resulting TGS crystal. Theoretical modeling for this experiment is being provided by W. L. Wilcox of Clarkson University. This experiment has been optimized in 1 g and flew on Spacelab 3 in 1985.

Ground-based tests on the solution growth of materials such as HgCdTe and $CdSnP_2$ are being carried out by K. J. Bachmann of NCSU. These materials are very difficult to grow in low g and K. J. Bachmann is investigating new growth techniques that may be possible in the space environment.

This is another growth technique in which 3M has an effort. They have proposed and flown experiments to evaluate the growth of organic crystals by allowing two reacting solutions to interdiffuse and form crystallites of the desired compound which has a low solubility in the solvent. On Earth, these crystallites, because of their density difference, will rapidly precipitate. However, in space, the particles will remain suspended and the larger crystallites will grow at the expense of the small crystallites by a process called Ostwald ripening. Extending the results of previous space experiments, 3M has grown crystals of various electro-optic materials (e.g., urea) to measure their nonlinear and electro-optical properties and to relate these properties to the growth habit and structure. In addition, growth rate and size distribution of crystals will be compared to theoretical models to validate the model. This is required to facilitate the design of future experiments utilizing this growth technique.

Melt Growth

Most industrial processes for bulk crystal growth in use today involve growth from the melt. These processes have evolved with little detailed understanding of the convective flows and how even small convective flows can have major effects on defect structure and compositional homogeneity. The MSAD program is largely responsible for heightening the awareness of the crystal growth community to the significance of these effects. R. A. Brown will present details of the theoretical status of this work in a subsequent talk. However, it must be pointed out that in a 1-g environment, buoyancy-driven convective flows will exist in a "vertically stable" configuration due to radial temperature gradients in the melt which are almost impossible to avoid in most semiconductor materials. Most systems of interest have more than one component; hence, thermal and solutal effects must be considered. S. Coriell and co-workers have shown that in a dilute solution if a less dense component accumulates at the solidification interface, a potential for solutal convective instability exists even when the concentration is so low that the combined thermal and solutal effects produce a density that decreases monotonically with height. For nondilute solutions, parameters such as solidification temperature and thermal diffusivity are sensitive to composition which means that the growth rate becomes a parameter which must be included in modeling studies.

The MSAD program has a wide base of experimental research in this area. H. C. Gatos and co-workers at MIT have been working on GaAs and related materials. Their initial work was in the area of liquid-phase electro-epitaxy (LPEE). In 1983, this work was chosen by Microgravity Research Associates (MRA) to realize bulk growth of GaAs in space. This work is now being sponsored by MRA in a joint endeavor agreement with NASA. Recently, this group has identified stoichiometry as a fundamental factor controlling structural and electronics properties in GaAs. They have shown correlations between microscopic stoichiometry fluctuations and the presence of defects in the crystal.

Working in close cooperation with the theorists, experimental analysis of the previous flight results and ground-based tests have shown the importance of furnace design on the control of interface morphology in crystals grown by the Bridgman technique. The growth rate can be continuously varying in an uncontrollable and totally unpredictable manner if great care is not taken to control both heat input to the sample and heat extraction from the sample. This has led to adoption of a multizone furnace configuration with an insulation region between the hot and the cold zones. A. F. Witt and co-workers at MIT have designed and constructed a furnace that is fully computer operated which allows the establishment of axial temperature gradients in germanium doped with gallium [Ge(Ga)] which are in good agreement with theoretical predictions and which allow excellent control of the interface morphology, except near the walls of the container. S. L. Lehoczky and co-workers at MSFC are studying the growth of the solutally stable mercury-cadmium-telluride (HgCdTe), and R. K. Crouch and co-workers at LaRC are studying the solutally unstable lead-tin-telluride (PbSnTe) system by the Bridgman technique. Since the thermophysical properties of these materials were not characterized, determinations of important parameters such as solutal diffusion rates, thermal diffusivities, specific heats, and densities as a function of temperature and composition have been measured. Lehoczky's group has found that the thermal diffusivities of the compounds of HgCdTe have significant discontinuities between the solid and the melt, which makes thermal modeling very difficult. In addition, the density of HgTe goes through a maximum at a temperature about 60° C above the solidification point. The other complications which must be considered in modeling the nondilute solutions of the compound semiconductor systems are the changing solidification temperatures and thermal properties of the melt as the composition near the interface changes owing to the difference in the equilibrium concentration in the solid and the liquid. Finally, the thermal conductivity due to the presence of the container must be taken into account. In Ge(Ga), which has a relatively high thermal diffusivity, this effect results in a curvature of the interface near the walls; however, in the compound semiconductor materials which have relatively low thermal diffusivities, this effect can result in major differences between the predicted and experimental interface shape.

SUMMARY

NASA has established a broad based program for understanding the effects of a low gravity environment on the growth and processing of electronic materials. This program covers the major growth techniques used to grow crystals. Through close interaction between the theoreticians and the experimentalists, there have been significant advances in understanding the effects of buoyancy-driven convection upon crystalline quality and morphology and the importance of furnace design has been established. These results have been reported and have heightened the awareness of the electronic materials industry. It is believed by the author, as a direct result of the work sponsored by this program, new growth

techniques have been developed which better control convection during growth and this has led to significant improvements in commercially available silicon and gallium arsenide. Although previous results from space experiments have not been spectacular, they have shown sufficient promise for improvement of materials to interest the commercial sector, as evidenced by the current participation of 3M and Microgravity Research Associates in the program. Recent results from the European space flights have shown the importance of convective forces such as Marangoni convection in processes which have free surfaces. The importance of solutal effects on convection and of crucible interaction with the heat and mass flows in crystal growth systems are just beginning to be understood. As progress continues in these areas and as more flight results become available, it is believed that even more dramatic improvements in electronic materials will result. In conclusion, NASA's establishment of a strong scientific basis for the Microgravity Science and Applications Program has had and will continue to have a direct, valuable impact on the technological advancements in the electronics industry in this country.

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6. STATUS OF MODELING OF CONVECTION, SEGREGATION, AND INTERFACE MORPHOLOGY IN MELT GROWTH OF SEMICONDUCTOR MATERIALS

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SUMMARY

Asymptotic analysis and newly developed numerical methods have lead to detailed descriptions of convection in directional solidification and its effects on macroscale and microscale segregation and interface morphology. Transitions in flow between diffusive and convective control of solute mixing have been established. Calculations distinguish between different experimental configurations for directional solidification of common semiconductor materials and compare well within laboratory measurements. Combinations of finite element methods and computer-implemented perturbation methods have predicted transitions to large-amplitude cellular interface morphologies.

TEXT

Prediction of the role of fluid convection on interface morphology, homogeneity, and defect structure of semiconductor crystals grown from the melt requires solution of the mathematical problems describing transport and phase change in solidification at a level of detail not attempted before the development of NASA's Microgravity Sciences and Space Applications Program. The purpose of this paper is to give my perspectives on the status of this modeling and on the implications of the results to the development of experimental programs both for space and Earth. The myriad of technical details and results comprising the analyses mentioned in the text are not presented but are available in the referenced publications.

Convection in the melt in a typical terrestrial crystal growth system is a combination of buoyancy-driven, surface-tension-driven, forced, and solidification-induced flows. The patterns of these motions influence the temperature and solute fields in the melt, the shape of the melt/solid interface, and, through the solidification process, the compositional uniformity and defect level in the crystal. Calculation of the flow and species fields in laboratory experiments involves numerical problems at the forefront of large-scale scientific computing. Calculations of melt flows in even simple geometries must account for nonlinear transitions in spatial and temporal pattern that are extremely important for correct prediction of solute segregation in the crystal. On Earth, such transitions in buoyancy-induced flows can lead to chaotic or turbulent convection and to random segregation patterns in the crystal. Experimental studies of surface-tension-driven flows on Earth and in space have shown similar transitions.

Even when the motion in the melt is steady, convective transport may cause detrimental dopant segregation across the crystal and unwanted curvature of the melt/solid interface. In an ideal growth system, solidification is unidirectional and the melt/solid interface is planar, so that dopants are uniformly distributed across the crystal. From this starting point, nonuniform convection caused by any source distorts the concentration field by stirring the melt. The melt will be well mixed and the dopant concentration will be uniform across the crystal only when this mixing is intense. This state is achieved on Earth in most industrial-scale growth systems, but at the expense of having turbulent flow and the resulting microscale fluctuations in interface structure and composition.

Intermediate convection levels achieved in small-scale systems on Earth and in microgravity experiments give less-than-perfect mixing and dopant segregation. Much of our analyses has been aimed at predicting segregation in realistic crystal growth systems and using numerical simulations to optimize these geometries.

Convection and the resulting solute transport also have important effects on the microscale transitions in melt/solid interface morphology important in the growth in most alloys and microstructured solids. Changes from planar, to cellular, and to dendritic interface morphologies can be described mathematically as nonlinear transitions in the same framework used for convective flows. We have taken this approach in our analysis of microscale structure.

Successful modeling of the complex transport processes in melt crystal growth has required the development of both mathematical tools and scientific collaborations. Some of our most important developments are listed below:

1. New numerical methods for solving the free- and moving-boundary problems arising in solidification systems have been formulated.[3,7,8,10,19]
2. Asymptotic analyses specifically designed for studying melt convection problems have been constructed for analyzing transport processes in idealized systems.[11,13]
3. Analytical and numerical methods for identifying and following nonlinear transitions in flow structure and interface morphology have been developed and applied in crystal growth phenomena on several length scales.[14,21]
4. Significant collaborations with experimentalists have been developed and have lead to accurate modeling of laboratory experiments and to feedback between calculations and furnace design.[1,2,5]

Each of these contributions is summarized below along with the applicability of these results to the NASA Microgravity Science Program.

The numerical calculations of convection and interface morphology on both microscopic and macroscopic scales are based on newly developed finite-element methods combined with a Newton iteration scheme for explicitly locating melt/solid and melt/gas interface shapes while computing field variables, e.g,. velocity, pressure, temperature, and solute concentration. These techniques were first developed for solving steady-state solidification problems[7,10] and have since been generalized to transient or moving-boundary problems[8,9]. The finite-element/Newton methods are the basis for powerful computer-implemented perturbation methods for tracking solution families as a parameter is varied, for detecting multiple steady and time-periodic solutions, and for determining solution stability. Many of these perturbation methods were developed by us under NASA support [14,21] and have been applied to problems ranging from descriptions of the flow transitions in natural convection without[21] and with phase change,[7] to analysis of combined thermal-capillary instabilities in small-scale floating zone systems,[9] and to calculations of transitions from planar to cellular and dendritic micromorphologies in two-dimensional solidification.[15-20] The combination of accurate numerical simulation and computer-implemented perturbation analysis has been crucial in each of these studies.

Finite-element calculations and asymptotic analyses of convection and segregation in idealized vertical Bridgman[1,6] and floating-zone[11-13] systems have clearly demonstrated the transitions from diffusion-controlled transport for crystal growth where only the unidirectional convection associated with growth is present to poorly mixed and finally well-mixed melts when higher levels of convection are present. These calculations have pointed out the dangers of crystal growth under conditions for low, but uncontrolled, convection, where unwanted dopant segregation across the crystal may be much more severe than on Earth. Optimization of space experiments to control convection is crucial.

One emphasis of our research has focused on the numerical simulation of directional solidification of dilute and nondilute binary alloys and of the floating-zone process for producing small single crystals. The modeling of laboratory-scale directional solidification systems has centered on the vertical Bridgman growth systems developed by A. F. Witt in the Materials Processing Center at the Massachusetts Institute of

Technology and by J. J. Favier in the Metallurgy Division of the Centre d'Etude Nucleaire, Grenoble, France. Calculations for small-scale floating zones have addressed the experiments with silicon under way at Marshall Space Flight Center. As reported in the publication cited[5] in more detail in a forthcoming manuscript,[1] calculations aimed at analysis of laboratory directional solidification experiments must include the precise conditions for heat transfer from the surrounding furnace to the melt and solid. Accurate calculations of flows and dopant segregation have been performed.[2] The finite-element calculations have successfully reproduced interface shapes, and dopant segregation in gallium-doped germanium experiments in the M.I.T. furnace[2] when both convective and radiative heat transfer to the ampoule and the finite heat-transfer rate through the ampoule are included. The same simulations are being used to model the growth of silicon-germanium alloys in the Grenoble furnace; details are given in reference 5.

Calculations of the temperature field and melt/solid and melt/gas interface shapes in small-scale floating zones have pointed out the complex interactions between heat transfer from the ambient and zone shape in determining the existence and configuration of steady-state growth configurations. For example, the length of the molten zone and its shape are coupled to the heat transfer from the surroundings through the shape of the meniscus, which determines the efficiency of the zone in accepting energy radiated and convected from the surroundings. Calculations of zone stability[9] based on the full thermal-capillary model give very different results from previous idealized results that simply treated a floating zone as a liquid drop captured between two inert solid rods. Our completed thermal model also leads the way for accurate assessment of surface-tension-driven flows based on realistic heat-transfer conditions for space experiments. Application of computer-implemented perturbation methods to these flow problems will also lead to predictions of the onset of the time-periodic flow instabilities seen both on Earth and in space.

Finally, calculations of microscale melt/solid interface morphologies in directional solidification have lead to the first complete description of the transition to deep two-dimensional cellular morphologies for dilute binary alloys without bulk convection. We have demonstrated that the initial sinusoidal instability of a planar interface described by a linear stability analysis accounting for solute diffusion in melt and solid evolves with increasing instability (either increasing growth rate or decreasing temperature gradient) until a nonlinear transition occurs as a secondary bifurcation that halves the spatial wavelength of the original cellular morphology. This secondary transition occurs so close to the onset of the cellular interfaces that morphologies with the original wavelength are probably not observable. We have computed this transition in spatial structure for a large number of solidification models[15,16] to prove that it is truly generic.

Deep cellular interfaces evolve with half the original wavelength systematically as the velocity is increased and are characterized by large cells separated by deep, thin grooves of melt.[17] The melt/solid interface at the bottom of the groove is actually re-entrant with a "droplet" of melt suspended on the bottom on the groove. Steadily growing cells do not exist beyond a limit in instability where the sides of the cells intersect and the droplet of melt would separate from the top of the cell. Calculations are currently under way[18] to detect the formation of dendritic sidearms along the cell tip as Hopf or time-periodic bifurcations from these steady-state cells and the pseudo-steady cell tips that exist when the droplet shedding has begun. These results are precursors to studies of the development of cellular morphologies in three dimensions that can be unambiguously tested in a zero-gravity environment.

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7. CRYSTAL GROWTH IN SPACE

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INTRODUCTION

Sublimation and condensation reactions are basic phenomena that are applied in a variety of important technological processes, including the purification of metals and the synthesis of materials. In the form of chemical vapor deposition (CVD), chemical vapor transport (CVT), and as sublimation or physical vapor transport (PVT), these processes are employed for the growth of single crystals of research quality and for the production of bulk and layer-type single crystalline materials for device-oriented applications. The open flow system and the generally large temperature gradients of the CVD technique result in a fluid dynamically very complex combination of gravity-driven and forced convection. This complexity is somewhat reduced for CVT and PVT processes, which are usually performed in closed systems and, thus, in the absence of externally imposed flow. Therefore, chemical and physical vapor transport reactions are ideally suited for the investigation of natural or gravity-driven convection effects on mass transport and crystal growth phenomena. In addition, the inherent thermochemical differences between CVT and PVT reactions, applied to the same material, can be used advantageously to elucidate the effects of the chemical characteristics of a system on the transport properties of the material.

In order to estimate the fluid-dynamic properties of CVT or PVT processes, to differentiate convection effects on transport and growth phenomena from possible other influences, it is imperative that the thermodynamic, kinetic, and structural properties of a system be well understood. The materials of interest, germanium selenide (GeSe) and related IV-VI compounds, have been extensively characterized in our and other laboratories in terms of their thermochemical and structural parameters. The orthorhombic structure of GeSe is anisotropic, yielding extensive growth perpendicular to the longest axis of the unit cell. Under near-equilibrium conditions, the dominant growth habit of GeSe are platelet-type crystals. Deviations from this crystallographically determined growth morphology indicate fluid dynamic and/or thermodynamic perturbations of the growth process. In addition, the structures of the IV-VI compounds are closely related to those of other technologically important materials. A detailed understanding of these relationships provides the basis for the meaningful estimation of the growth properties of other materials.

In our microgravity studies to date, GeSe has been used as a model system employing CVT and PVT reactions. The primary objectives of the space experiments were to determine absolute mass transport rates, to observe realistic effects of reduced gravity on crystal growth, and to observe possible unexpected transport and growth phenomena. The internally consistent results of eleven space experiments on the above systems justify the conclusion that basic scientific and technologically significant data have been produced through this work.

Principles of Chemical Vapor Transport

Before summarizing the microgravity results obtained to date, a brief description of the chemical vapor transport method is desirable. The basic principles and general applications of chemical vapor transport reactions have been established earlier.[1] Further extensions with respect to theoretical considerations and experimental studies have been discussed in recent years.[2,3] The essential aspects of CVT reactions are briefly summarized below.

A chemical vapor transport reaction is defined as a reaction between a condensed source material and a gaseous transport agent to form exclusively gaseous transport products. The dominant reaction for the GeSe-GeI_4 system is given by the reaction

$$GeSe(s) + GeI_4(g) = 2\ GeI_2(g) + 1/2\ Se_2(g). \quad (1)$$

The vapor species migrate from the source to the condensation region where the reverse reaction occurs with formation of single crystals. The driving forces for the vapor transport process are based on temperature gradients along the axis of the container and on the temperature dependence of the equilibrium constant of the transport reaction. Depending on the thermochemical properties of the system, net mass transport can occur from high to low or from low to high temperature. In contrast, sublimation (PVT) can only occur from high to low temperature. Diffusive mass flow is governed by Fick's law and can be represented in an oversimplified form by the equation

$$J_d \propto \Delta P_i \times \Sigma P^{-1}, \quad (2)$$

where ΔP_i is the partial pressure difference of species i between the hot and cold zone, and ΣP is the total pressure. The presence of a density gradient and of a gravitational field cause convective motion, which is bulk flow of mass superimposed on diffusive transport. In a simplified form, convective mass flux can be represented by the equation

$$J_c \propto g \times \Delta P_i \times \Sigma P, \quad (3)$$

where g is the gravitational acceleration. Equations (2) and (3) illustrate the important difference in the dependence of the mass flux J on the total pressure. This is graphically illustrated in terms of J versus ΣP in Figure 7.1. At very low pressures (region 1), when the mean-free path of the molecules is greater than the dimensions of the container, the mass flux is controlled by the rate of the heterogeneous solid-gas phase reactions at the source and/or crystal. In region 2, where mean-free-path considerations are important, mass transport is diffusion limited. Region 3 represents the convection-dominated mass transport regime. It should be noted that diffusive and convective mass transport occur simultaneously; the relative contribution of the diffusive and convective component to the total mass flux can be changed by varying the transport agent (total) pressure. Another way of changing the relative dominance of diffusive and convective flow is through the orientation of the density gradient with respect to the gravity vector. Under horizontal conditions and for $\Delta T \neq 0$, there is always diffusive-convective flow. Under vertical destabilizing conditions (hot zone at the bottom), convective flow is maximized. Under vertical stabilizing conditions (hot zone on top), convective flow is minimized and would be zero if there would be no radial ΔT (perpendicular to the density gradient). Since in most practical cases it is very difficult, if not impossible, to eliminate radial temperature gradients, there are always convective contributions on Earth, even under vertical, stabilizing conditions.

The major advantages of the chemical vapor transport technique include the use of lower temperatures. This reduces the associated reaction pressures and container problems in terms of inertness, impurities, and mechanical stability. The use of a single source material has a significant impact on the simplicity of the apparatus and of the procedures involved.

The effects of gravity-driven convection on crystal morphology of IV-VI compounds have been observed earlier in our laboratory.[4] From those and subsequent studies it was justified to conclude that in general the crystal quality in terms of surface and bulk morphology decreases with increasing convective contribution to the total mass transport. This is illustrated for the $GeSe-GeI_4$ system in Figure 7.2.[5] Under the conditions of Figure 7.2a, the mass transport is diffusion controlled; and under the conditions of Figure 7.2b, convective flow is the dominant transport mode. Because the mass transport rates for condition 7.2a and 7.2b are the same within error limits, the drastic changes in surface morphology from high-quality single-crystal platelets (Figure 7.2a) to dendritic type growth (Figure 7.2b) are solely caused by the change from dominant diffusive to dominant convective flow. At higher pressures and increased convective contributions, the deposition pattern of dendrites on the ampoule wall (Figure 7.2c) reveals the nutrient flow pattern in the ampoule.

Based on our earlier chemical vapor transport studies of GeSe (Ref. 6) and GeTe,[4] it was predicted that crystals of improved morphology relative to corresponding ground-based conditions could be obtained in a microgravity environment. This was the beginning of our earlier crystal growth experiments performed during the Skylab and Apollo-Soyuz flights.

RESULTS OF SKYLAB AND APOLLO-SOYUZ EXPERIMENTS

The primary objectives of our previous chemical vapor transport studies in microgravity environment during the Skylab and Apollo-Soyuz missions were to observe realistic effects of microgravity on crystal morphology and to measure mass transport rates in the absense of convective interferences. It was expected that the crystal morphology could be improved relative to ground-control specimens and that the mass transport rates would correspond to diffusive transport. The experimental conditions were not designed to produce optimum quality crystals in space, but to confirm the above predictions.

For this purpose, chemical vapor transport and crystal growth experiments were performed on the following materials: GeS, GeSe, GeTe, $GeS_{0.98}Se_{0.02}$, and $GeSe_{0.99}Te_{0.01}$. The transport agents employed were $GeCl_4$, GeI_4, and a $GeCl_4$-Ar mixture. For these experiments, three different temperature gradients and nine different transport agent pressures were employed yielding a total of nine microgravity experiments. The essential results of the Skylab[7] and Apollo-Soyuz[8] experiments are summarized below.

1. The chemical and structural microhomogeneity of space-grown crystals was considerably improved relative to ground-based specimens.
2. The mass transport rates observed in space were greater than expected for microgravity conditions.

First of all it is important to note that the above observations were made consistently for all nine experiments. Therefore, the results observed have a rather high degree of validity. In fact, these experiments represent the only series of microgravity experiments which are checked in space under different experimental conditions. It is the internal consistency of the results which provides the basis for their general significance.

The observation of improved crystal morphology confirmed our predictions based on previous studies carried out in our laboratory. However, the observation of greater mass transport rates in microgravity environment than predicted represents a highly unexpected and major finding which is of basic scientific and technological significance. It is important to emphasize that the discrepancy between observed and expected mass transport rates in space increases with increasing transport agent pressure. This observation strongly suggests that the cause of the flux anomalies observed in microgravity environment is related to the effects of homogeneous gas phase reactions in the above vapor transport systems. Because of the unconventional nature of the flux anomalies, additional experimental and theoretical ground-based studies on the GeSe-GeI_4 system were performed.

CONTINUED GROUND-BASED STUDIES OF THE GeSe-GeI_4 SYSTEM

In order to further elucidate the above discussed flux anomalies, the continued investigation of the GeSe-GeI_4 vapor transport system was directed toward four major tasks. These were (1) the quantitative analysis of the vapor phase of the above systems; (2) the investigation of the relation between diffusive and convective mass transport; (3) the development of comprehensive expressions for the computation of diffusive mass transport rates; and (4) the measurement of the effects of orientation of the density gradient relative to the gravity vector on mass transport rates. The results of these studies, relevant for the microgravity experiments, are briefly summarized below.

Based on available thermochemical data of the GeSe-GeI_4 system, which were in part produced in the author's laboratory, the composition of the vapor phase and the partial pressures of gas phase species were computed.[5] The results of these extensive calculations are graphically represented in terms of partial pressures as a function of total pressure in

Figures 7.3 and 7.4 for the temperature of the source (793 K) and condensation region (693 K), respectively. The results demonstrate that the GeSe-GeI_4 system is a multicomponent, multireaction system, that the vapor phase compositon is very complex,and that homogeneous gas phase reactions do occur in this system. The dominant vapor species are GeI_4, GeI_2, and Se_2. This confirms that the main transport reaction is correctly represented by Equation (1). However, an accurate computation of partial pressures and mass transport rates requires the consideration of the minor species present in the vapor phase.

The relation between diffusive and diffusive-convective mass transport was experimentally analyzed by determining the mass transport rate of the GeSe-GeI_4 system as a function of inert gas pressure.[5] For this purpose, increasing amounts of inert gas were added to the GeSe-GeI_4 system for selected and fixed pressures of transport agent GeI_4. The mass transport rates of GeSe in terms of reduced flux J' as a function of total pressure are represented in Figure 7.5. The numbers on the flux curves indicate the fixed pressure of GeI_4 used in the different areas. For the GeI_4 pressures of 0 and 0.14 atm the transport is diffusion controlled, of and for the GeI_4 pressures of 0.5 to 1.8 atm the overall transport mode is convection dominated. The linear decrease of flux with increasing argon pressure, following a straight line with a slope close to minus unity, reveals the existence of a diffusion barrier under all conditions in the diffusion and convection dominated pressure regimes. The positive deviation of the flux from the straight lines at higher argon pressures indicates the "onset" of another convection mode. Within the range of the linear portions of the flux curves, the addition of argon increases the density of the diffusion barrier at the source and growing crystals, but the thickness of the diffusion boundary layers remains unchanged for a given GeI_4 pressure. At higher argon pressures, when the flux begins to increase again, the thickness of the boundary layer decreases further with increasing argon pressure as the result of a new convective mode. From the linear portions of the flux curves and other known experimental parameters, the thickness of the boundary layers can be calculated for different GeI_4 pressures. Assuming that a diffusion boundary of thickness δ exists at the solid-gas interface at the source and growing crystal, the boundary layer thickness 2δ as a function of GeI_4 pressure calculated from the above experimental results is represented in Figure 7.6. At very low GeI_4 pressures, the boundary-layer thickness corresponds to the distance between the source and growing crystal, which is 15 cm for these experiments. With increasing transport agent pressure and increasing convective contribution to the total mass transport, the diffusion path length (boundary-layer thickness) decreases progressively. It is important to note that the boundary layer does not reach a minimum thickness within the pressure range of this investigation as was suggested in the literature.[9] These investigations represent the first experimental evidence for the existence of a boundary layer in closed vapor transport systems under all conditions.[5] The result of convection on the mass transport rate of a system can be interpreted as an effective "shortening" of the length of the diffusion path.

The dependence of the mass transport rates on the orientation of the density gradient with respect to the gravity vector was thoroughly investigated for the $GeSe$-GeI_4 system as part of the continued studies.[10] The results are graphically represented in Figure 7.7. For curves 1-3, the hot zone is on top (stabilizing condition), curves 4 and 5 represent the horizontal ampoule orientation, and for curves 6-9 the hot zone is at the bottom (destabilizing condition). Curve 1 is for the vertical, stabilizing and curve 8 for the vertical, destabilizing orientation. At very low pressures of GeI_4, the mass transport rates are independent of orientation, indicating diffusion controlled transport. The measurable onset of convective transport under present conditions is at about 0.15 atm pressure. With increasing transport agent pressure, the dependence of the mass transport rate on ampoule orientation becomes more pronounced.

In order to compare the experimental data with theoretical values computed for diffusion controlled transport, comprehensive transport equations were developed based on a rigorous treatment of the processes involved.[10] For this purpose, the simultaneous existence under given experimental conditions of chemical vapor transport, of sublimation, and of Stefan flow was considered. The resulting transport equation represents a one-dimensional model for the calculation of diffusion controlled mass fluxes in multicomponent, multireaction systems which is generally applicable to other systems.[10] For the computation of accurate partial pressures and mass transport rates, the presence of all gaseous species in the system discussed above was considered.

Theoretical mass transport rates computed for diffusive transport are given by the solid curves 1-9 and are compared with experimental data in Figure 7.8.[10] The curves 1-9 represent increasing levels of computational rigor and of completeness of the chemical system considered. Curve 9 is based on the most complete computational expression. The excellent agreement of experimental results with computed data (curve 9) over two orders of magnitude in pressure demonstrates the validity of the computational model. At pressures below about 0.1 atm (diffusion controlled range), the experimental mass fluxes for all orientations are in very close agreement with theoretical data (curve 9). Above 0.1 atm GeI_4 pressure, only the data obtained under vertical, stabilizing conditions follow the theoretical curve. In view of the close agreement between experimental and theoretical data (curve 9) over a wide pressure range, the observed deviation of the experimental data from curve 9 at higher pressures is considered to be real and not due to experimental uncertainties. The flux data represented by asterisks were obtained for this system and for the same temperature conditions in microgravity environment. The microgravity result obtained at 1.5 atm GeI_4 pressure is about 300 percent greater than the flux under vertical, stabilizing conditions and about 400 percent greater than the theoretical value (curve 9). These discrepancies are well outside any reasonable error limits. The comparison between theoretical, ground-based, and microgravity results confirms the validity of the flux anomalies observed earlier for all Skylab and Apollo-Soyuz experiments.

RESULTS OF STS-7 SHUTTLE EXPERIMENTS[11]

The STS-7 Shuttle experiments are a direct consequence of the earlier Skylab and Apollo-Soyuz experiments on the $GeSe-GeI_4$ and related systems. The primary objectives of the Shuttle experiments are the continued investigation of mass transport phenomena in microgravity environment and the quantitative determination of absolute mass transport rates observed for different conditions. In order to modify the thermochemical parameters of the transport systems, the transport agent GeI_4, employed in the earlier Skylab and Apollo-Soyuz experiments, was replaced by the inert gas xenon in the Shuttle experiments. Under these conditions, the source material GeSe sublimes through an atmosphere of xenon according to the reaction

$$GeSe(s) = GeSe(g). \quad (4)$$

This process is also called physical vapor transport in the literature. The mass transport or sublimation rate of GeSe is controlled by the thermodynamic and fluid dynamic parameters of the system. The latter are dependent on the xenon pressure and the orientation of the density gradient relative to the gravity vector for otherwise fixed conditions of temperature gradient and ampoule geometry. In order to minimize the errors associated with mass flux measurements, the mass transport rates to be observed were maximized by employing a large temperature difference between the source and condensation region of the ampoule. Under these conditions, the growth of very small GeSe crystals (majority less than 1 mm edge length) and polycrystalline deposition were observed in all ground-based experiments for the horizontal and vertical, stabilizing orientation of the transport ampoule. For the two experiments performed in microgravity environment during the STS-7 Shuttle flight, one temperature gradient and two different xenon pressures were employed.

The results to date of the above GeSe-xenon STS-7 flight experiments are summarized as follows.

1. The mass transport rates of GeSe observed in microgravity environment are in close agreement with theoretical predictions based on diffusion controlled mass flux. These results were expected and demonstrate that the mass transport of the GeSe-xenon system is diffusion limited under microgravity conditions.

2. The deposition pattern and crystal growth phenomena observed for this system in microgravity environment were completely unexpected. As indicated above, under ground-based vertical, stabilizing conditions, polycrystalline deposition, and crystal dimensions less than 1 mm were observed. On Earth all crystallites are firmly attached to the ampoule wall.

In space, individual and much larger single crystal platelets are obtained. Most importantly, the largest space grown crystal platelets (about 10 x 4 mm^2) had no direct contact with the ampoule wall. They were supported by other crystals extending from the ampoule wall.

3. Present results of crystal characterization studies reveal that the surface and bulk morphology of the space grown crystals is considerably improved relative to corresponding ground-based specimens.

The combined observations of the STS-7 flight experiments demonstrate that these experiments were rather successful and continue the series of our earlier space results. With respect to the mass transport rates, the close agreement between predicted and observed mass fluxes for the GeSe-xenon system (STS-7) supports our proposed hypothesis for the earlier flux anomalies of the GeSe-GeI_4 system (Skylab and ASTP). The STS-7 results are internally consistent with those of the Skylab and Apollo-Soyuz flights. This demonstrates the reliability of the thermodynamic data employed and the high confidence level in the results obtained. The considerably improved morphology of GeSe crystals obtained by PVT (STS-7) relative to ground-control specimens is consistent with that of GeSe crystals grown by CVT (Skylab, ASTP).

SUMMARY AND CONCLUSIONS

The combined observations of our vapor phase crystal growth experiments in microgravity environment, employing CVT and PVT techniques, can be generalized to yield the following results. Microgravity effects on mass fluxes in vapor transport systems have been systematically established. Positive effects of reduced gravity conditions on crystal size, morphology, and crystallographic and chemical microhomogeneity have been identified. The observation of predicted mass fluxes in microgravity for simple systems and of unexpected transport phenomena for complex systems demonstrates the influence of the chemical characteristics of the transport system. The considerable morphological improvement of space grown crystals has direct practical consequences for the production of materials for device oriented applications. The observation of unexpected crystal growth phenomena could imply a new concept for the synthesis of materials and for the growth of large crystals in space.

In combination with systematic and critical ground-based studies, these space experiments have contributed to our basic understanding of transport processes. The application of this knowledge to the crystal growth of other electronic materials is an important aspect of present and future flight experiments. It is justified to conclude that the new information and observations produced by these experiments are both of basic scientific importance and of technological significance.

ACKNOWLEDGMENTS

The support of this work by the National Aeronautics and Space Administration through the Microgravity Science and Applications Division, Washington, D.C., and through the Experiments Development Office, George C. Marshall Space Flight Center, Huntsville, Alabama, is gratefully acknowledged.

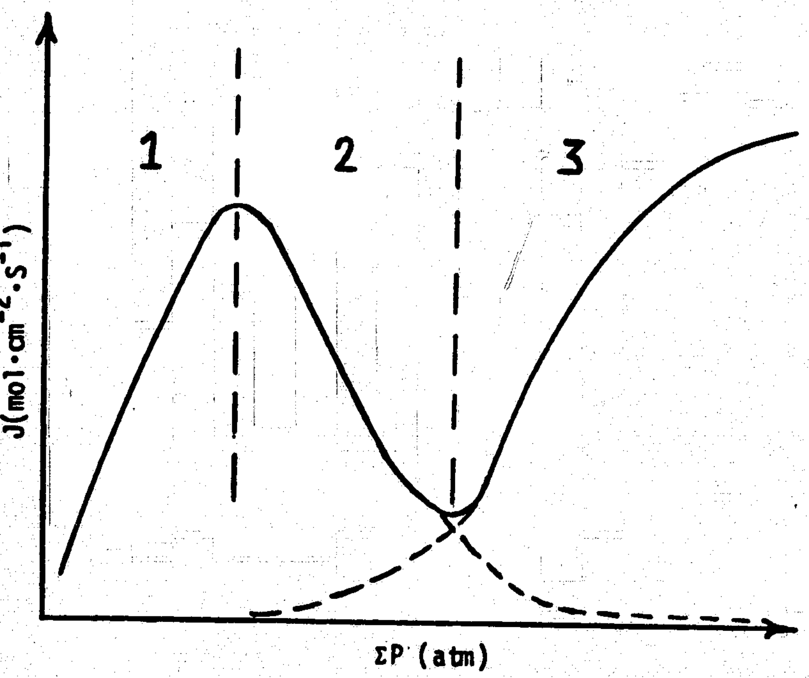

FIGURE 7.1. Dependence of the mass flux J on the total pressure.

FIGURE 7.2. Optical photomicrographs of representative GeSe crystals obtained under different growth conditions in the GeSe-GeI_4 system: (a) 0.04 atm initial GeI_4 pressure, diffusion limited range, magn. 3.75X; (b) 0.38 atm initial GeI_4 pressure, convection dominated range, mass transport rate within error limits of that of condition (a), magn. 5.25X; (c) 1.00 atm initial GeI_4 pressure + 6.70 atm Ar, magn. 3.75X. Photograph (c) was taken through the wall of the closed transport ampoule.[5]

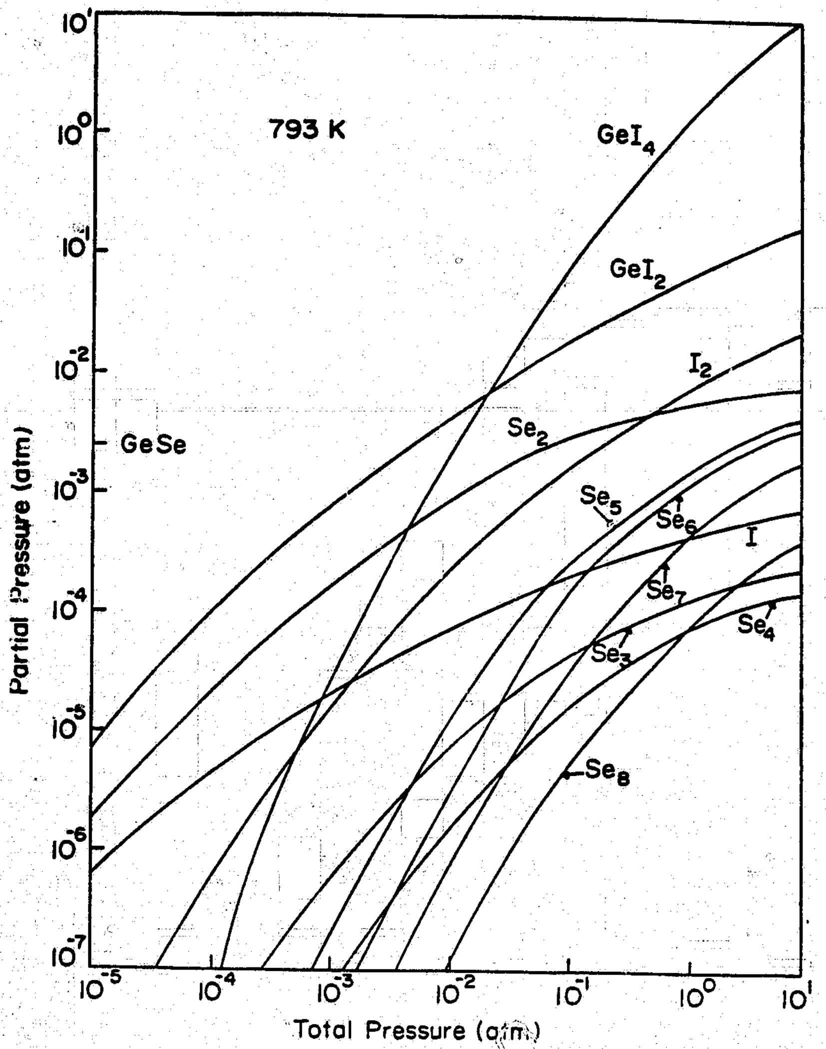

FIGURE 7.3. Equilibrium partial pressures of gaseous species in the GeSe-GeI_4 system as a function of total pressure at 793 K. The vapor pressure of GeSe is also shown on the ordinate.[5]

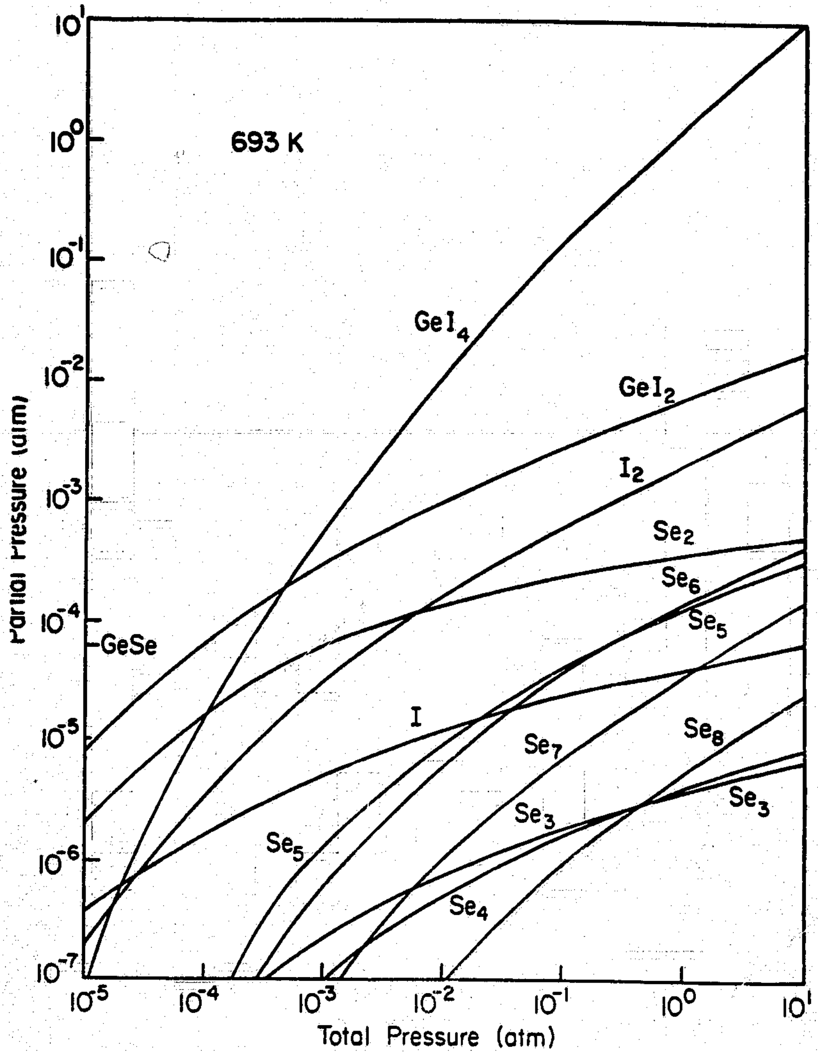

FIGURE 7.4. Equilibrium partial pressures of gaseous species in the GeSe-GeI_4 system as a function of total pressure at 693 K. The vapor pressure of GeSe is also shown on the ordinate.[5]

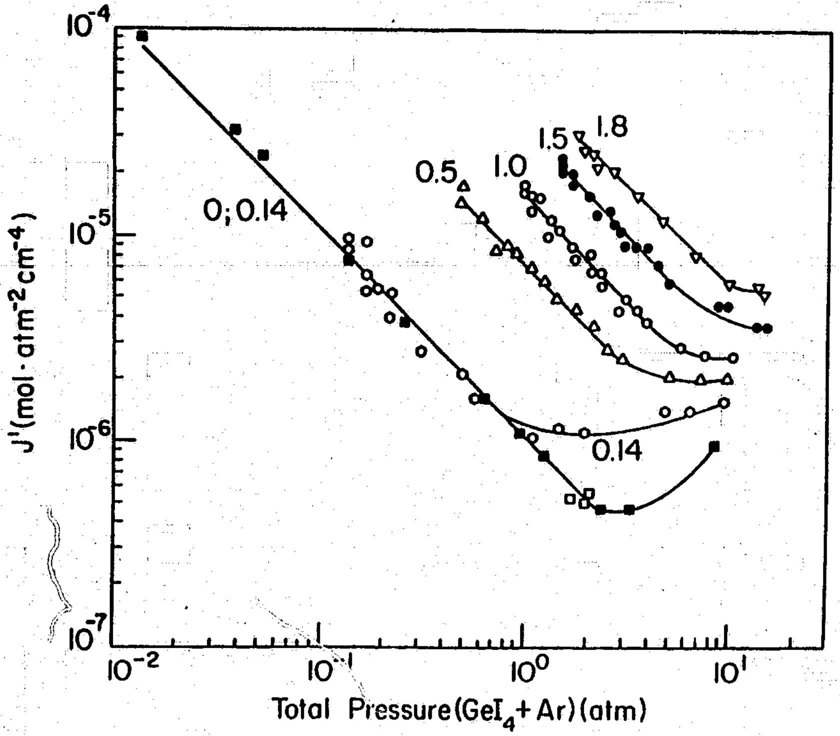

FIGURE 7.5. Variation of the reduced flux J' with total pressure (GeI_4 + Ar) in the GeSe-GeI_4-Ar system for given initial pressures of GeI_4 (■ indicate 0 atm GeI_4), (□ indicate 0.14 atm GeI_4, He added instead of Ar). The numbers on the curves represent the initial GeI_4 pressures.[5]

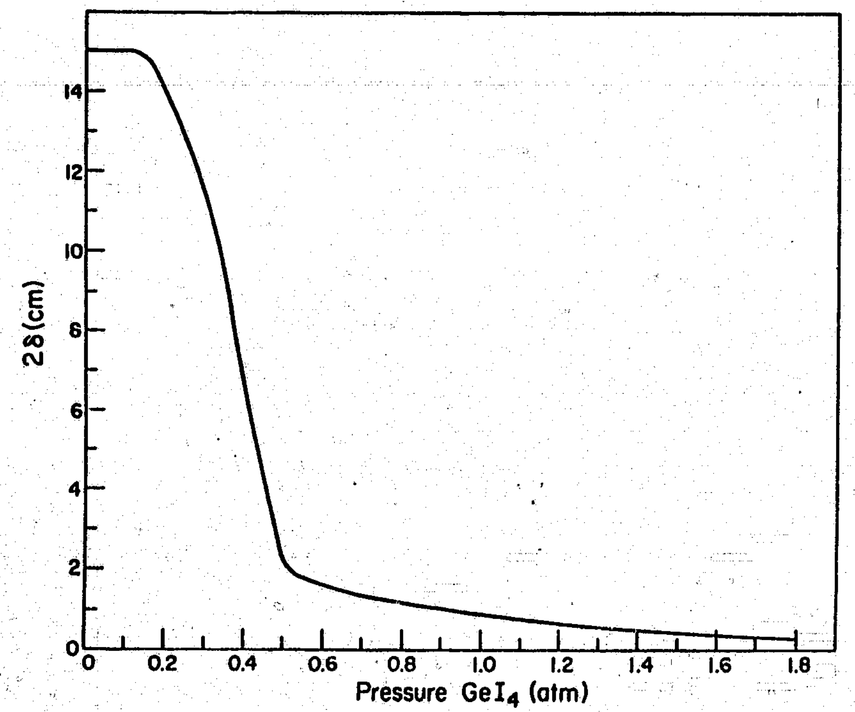

FIGURE 7.6. Variation of the boundary layer (diffusion barrier) thickness (2δ) with GeI_4 pressure in the GeSe-GeI_4 system.[5]

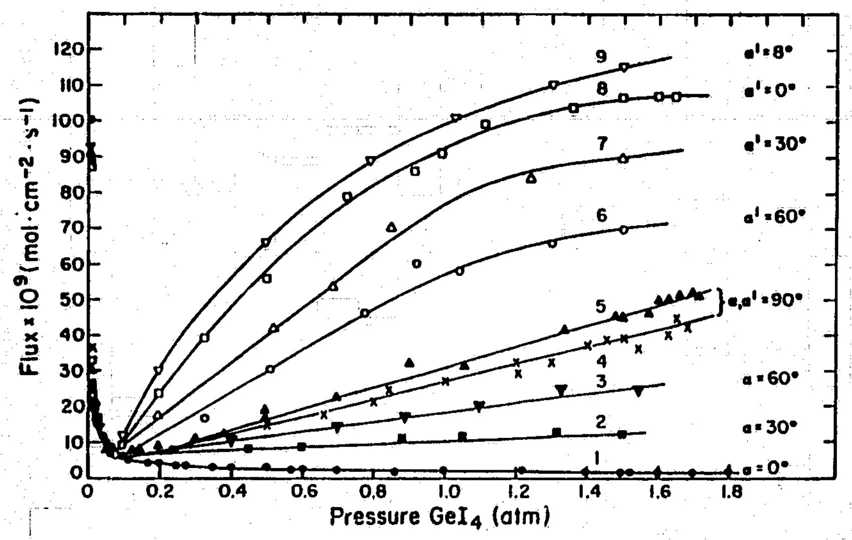

FIGURE 7.7. Mass transport rates of the GeSe-GeI_4 system as a function of ampoule inclination with respect to the gravity vector. Angle α denotes inclination with hot zone above (curves 1-3; curve 1 vertical, stabilizing) and α' denotes inclinations with hot zone below (curves 6-9; curve 8 vertical, destabilizing). Curves 4 and 5 (α = α' = 90°) horizontal orientation of ampoule with and without indentations near hot end of ampoule, respectively. Values on the abscissa are initial pressures of GeI_4.[10]

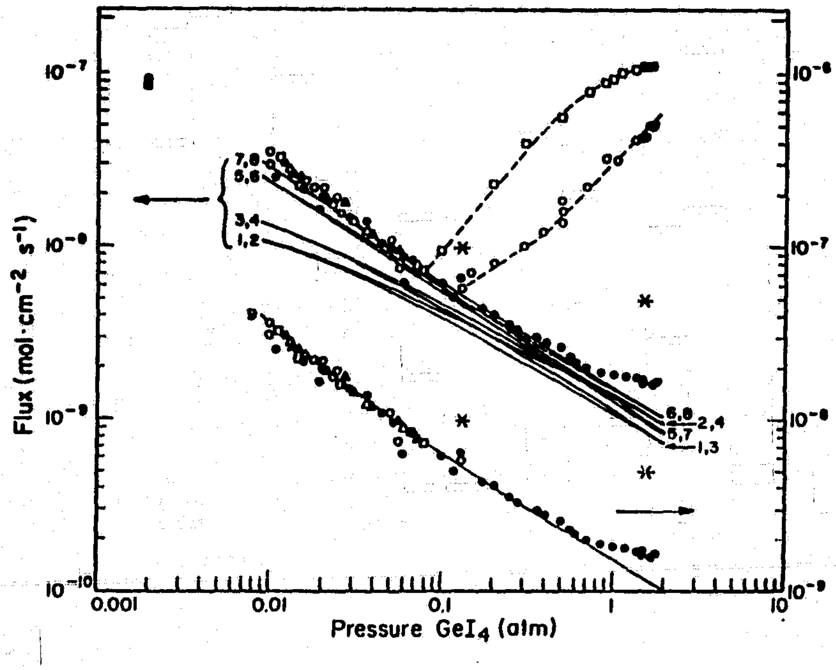

FIGURE 7.8. Comparison of experimental mass transport rates of GeSe-GeI_4 system with computed fluxes based on diffusion and Stefan flow. The solid lines indicate theoretical predictions and their calculational bases: curve 1, main transport reaction; curve 2, all transport reactions; curve 3, main transport reaction and Stefan flow; curve 4, all transport reactions and Stefan flow; curve 5, main transport reaction and sublimation; curve 6, all transport reactions and sublimation; curve 7, main transport reaction and Stefan flow and sublimation; curve 8, all transport reactions and Stefan flow and sublimation; curve 9, all transport reactions and Stefan flow and sublimation; different diffusion coefficient calculation. Experimental data: (O) vertical stabilizing orientation; (●) horizontal orientation; □ vertical destabilizing orientation; (Δ) ampoule inclinations of 30° and 60° with respect to vertical, hot zone above; (▲) ampoule inclinations of 30° and 60° with respect to vertical, hot zone below; (*) transport rates under microgravity conditions. Curves 1-8 left-hand-side ordinate and curve 9 right-hand-side ordinate. Values on the abscissa are initial pressures of GeI_4.[10]

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8. OVERVIEW OF THE PROGRAM IN GLASSES AND CERAMICS IN MICROGRAVITY SCIENCE AND APPLICATIONS

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INTRODUCTION

The purpose of the program in glasses and ceramics in microgravity is to explore containerless processing of these materials in low gravity. The usual processing of glasses in a container often leads to contamination from the container and nucleation of crystals at the container walls. Thus containerless processing offers the possibility of making purer glasses and new glass compositions that normally crystallize at the container wall.

Some potential applications of glasses require high purity. For example, fiber-optic waveguides need low optical absorption to increase the distance between amplifiers. Impurities in the glass fibers increase absorption.

New glass compositions have a variety of potential uses. A wider range of optical transmission in the infrared can reduce losses in fiber-optic cables at the

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Some potential applications of glasses require high purity. For example, fiber-optic waveguides need low optical absorption to increase the distance between amplifiers. Impurities in the glass fibers increase absorption.

New glass compositions have a variety of potential uses. A wider range of optical transmission in the infrared can reduce losses in fiber-optic cables at the absorption minimum (see Figure 8.1). Crystalline materials with this desired transmission range are known but cannot now be made into glasses because they crystallize during cooling. New laser host materials are needed. Many other high-value and low-volume needs could be satisfied by new glass compositions.

Another goal of containerless processing is to learn more about materials processing and properties so that known processes and materials can be improved. An example is the measurement of surface energies of corrosive liquids free of a substrate or container. Thermodynamic and thermophysical properties such as heat capacity, thermal conductivity, and emissivity are needed for materials development and understanding and could perhaps be measured under containerless conditions.

The glass and ceramics program is summarized in the following sections: introductions of persons conducting research in the program, ground-based studies, flight equipment, flight experiments, and applications.

PARTICIPANTS

A list of participants in the glass and ceramics program with their main areas of contribution and institutions is in Table 8.1.

GROUND-BASED STUDIES

Processing

In order to make pure, homogeneous starting materials for glass processing the glass components can be mixed as liquid organic compounds and precipitated as a gel. This method is already used to prepare starting materials for glass shells used as laser fusion targets (Downs). Properties of glasses made from gel starting materials are being compared with the same glass composition starting materials (Neilson-Weinberg). A wide variety of gel glasses are possible, for example tantalates (Downs) and germanates (Mukherjee) as well as silicates.

The homogenization of melts will be examined in flight experiments by Day.

Bubbles are formed in glass melts, and large bubbles are removed on the ground by rising to the melt surface. The gas in small bubbles can dissolve in the glass, causing the bubbles to shrink and disappear. The theory of bubble growth and shrinkage under a variety of conditions has been treated by Weinberg, Subramanian, and Cole. Bubble motion and Marangoni flows in melts are being investigated by Subramanian and Cole.

Lee and Wang have built a drop tower and make large (>0.5 mm) shells of good surface quality. Wang will give more details of this program in his presentation.

Ethridge has examined fiber drawing of glass.

Properties

Nucleation and growth of crystals in glass melts during cooling limit glass formation. Kinetics of crystallization have been studied in zirconium fluoride glasses by differential scanning calorimetry, x-ray diffraction, and light and electron microscopy by Doremus and Neilson and Weinberg.

Surface tension of zirconium fluoride melts was measured by Doremus for comparison with flight experiments, and surface segregation of components such as lead, vanadium, and molybdenum in silicate glasses studied by Uhlmann.

Equipment

A single-axis acoustic levitator for use up to 1500°C with multiple samples has been built by Whymark and Rey at Intersonics Corp. A three-axis acoustic levitator for the mid-flight deck has been built at the Jet Propulsion Laboratory (JPL) under the direction of Taylor Wang. Williams and Lofgren at Johnson Space Center are developing a single-axis acoustic levitator for examination of silicate melts.

Dunn has built several gas-jet levitators for use up to 1000°C with remote control of the sample position.

Scientific Results

To this point I have tried to list all programs impartially. However, I would like to select a few scientific results for special mention, because evaluation of science quality is emphasized as a workshop goal. These results have all been published in reviewed journals.

Theories of acoustic behavior in a cubical furnace have been developed by Barmatz and Wang at JPL. These results are of importance in the broader understanding of acoustic behavior in particular geometries, as well as being necessary for design and control of the high-temperature acoustic levitators.

Theories of bubble behavior have been explored extensively by Weinberg and Subramanian, as mentioned before. These results are of value in understanding glass melting, as well as shell formation and space processing of glass.

Studies of crystal growth in flouride glasses have been reported by Doremus and Neilson and Weinberg, and are important in predicting the stability of these glasses, their ease of formation, and their properties.

Sol-gel formation of glasses as reported by Mukherjee and Downs is valuable in developing this new technique for making glasses.

Flight Experiments

Two experiments on glasses have been flown on the Space Shuttle, and repeats of these experiments are planned on future Shuttle flights.

Day used the single-axis levitator built by Intersonics Corporation for his samples. In addition to three inert samples, there were five different glass samples, as listed in Table 8.2. These experiments were designed to test the possibility of melting glass in space. They include studies of melting and crystallization during cooling of different glass compositions (samples 1, 2, 3, and 5), of bubble behavior (samples 1 and 4), and homogenization (samples 1 and 4).

In the first flight the cooling shroud stuck open before any of the glass samples could be processed. Sample 1 was partially melted and showed bubble formation in post-flight analysis. The moving-picture camera did not operate properly, so no in-flight pictures were recorded. Other samples were not processed as planned.

The heating sequence for flight experiments by Doremus and Elleman in a three-axis acoustic levitator built at JPL is given in Table 8.3.

In the first flight the sample was observed during the first portion of the heating cycle (up to about 500°C) but was then lost from view because of acoustic instabilities. The surface of the sample showed changes, probably softening, but the viewing capability was not adequate to resolve details. The sample was recovered as coarse granules; a dark indent on one chunk indicated that the sample stuck to the wire restraining cage in the furnace. The sample showed a fine crystalline microstructure (<1 μm) compared with the much larger columnar crystals that grow on slow cooling in a container. X-ray diffraction showed the same crystalline phases in the flight and ground samples, but in different proportions.

Additional flights for both of these experiments are planned.

Previous low gravity experiments have been carried out in drop tubes, KC-135 airplane flights, and rockets, mainly to test equipment. Some earlier drop-tube experiments by Ralph Happe (deceased) showed the possibility of glass formation in oxide systems such as tantalates in which glass formation is difficult.

APPLICATIONS

Some possible high-value low-volume applications of glass to which containerless processing could lead were mentioned in the introduction. The formation of glass shells as laser fusion targets will be described by Taylor Wang; other applications for the larger (>1 mm diameter) shells are developing.

There are a variety of potential applications in optics. Pure, moat, and crystal-free fibers are required for longer fiber lengths without amplification. The zirconium fluoride glasses are especially attractive for fiber optics, because their wider infrared range of transmission should lead to a lower minimum loss (see Figure 8.1). These glasses also have the potential for use as optical components (lenses, prisms) and as environmental sensors of different factors such as pressure, temperature, and gas composition.

New laser hosts are needed for different wavelengths and bandwidths and higher power. Glasses made only in containerless processing might fulfill these needs.

SUMMARY

The program includes a vigorous set of ground-based experiments on processing and properties of glasses pertinent to flight experiments. Acoustic and gas jet levitators are being developed. Flights in the Space

Shuttle have started, but more are needed to explore the value of container processing of glasses. There are a variety of potential applications of the resulting glasses.

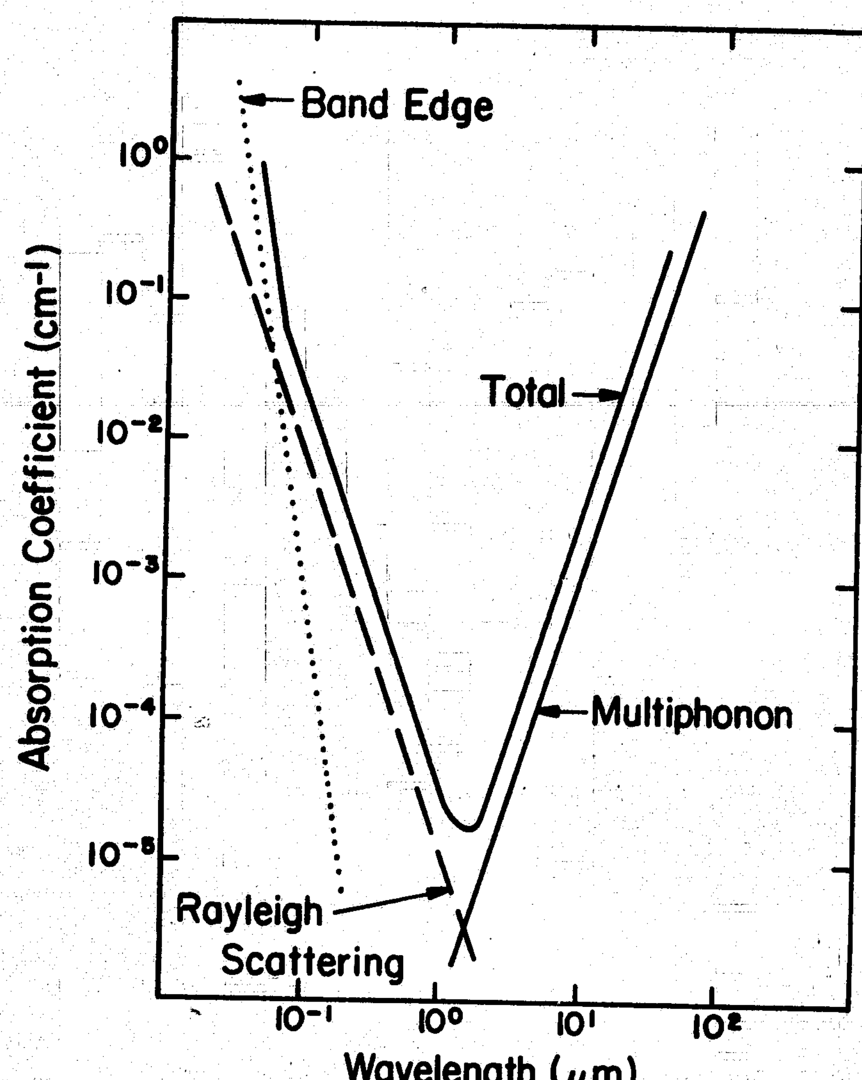

FIGURE 8.1 Schematic plot of intrinsic optical absorption coefficient versus wavelength in an insulator.

TABLE 8.1 Some Principal Investigators in the Microgravity Program in Glass and Ceramics

MARTY BARMATZ, JPL. Acoustics, levitators.
DELBERT DAY, Missouri Rolla. Processing of silicate glasses.
ROBERT DOREMUS, RPI. Processing of fluoride glasses, nucleation, and crystallization.
RAY DOWNS, KMS Fusion. Gel synthesis; glass shells.
STANLEY DUNN, Sedun. Gas-jet levitators.
DAN ELLEMAN, JPL. Measurement of melt surface tension.
ED ETHRIDGE, Marshall. Glass processing and fabrication.
MARK LEE and TAYLOR WANG, JPL. Fluid mechanics, acoustics, glass shells.
SHARMA MUKHERJEE, JPL. Gel synthesis.
GEORGE NEILSON and MIKE WEINBERG, JPL. Nucleation and crystallization, bubbles, gel synthesis.
P. C. NORDINE, Midwest Research. Measurement of gaseous boundary layer compositions with laser-induced fluorescence.
SHANKAR SUBRAMANIAN and ROBERT COLE, Clarkson. Bubbles, fluid mechanics.
DONALD UHLMANN, MIT. Nucleation and crystallization, surface segregation.
R. R. WHYMARK and C. REY, Intersonics. Acoustic levitators.
R. J. WILLIAMS and G. E. LOFGREN, Johnson Space Center. Acoustic levitators, redox control of iron silicate melts

<u>Addresses:</u>
JPL: Pasadena, CA
RPI: Troy, NY
KMS Fusion: Ann Arbor, MI
Marshall Space Flight Center: Huntsville, AL
Clarkson College: Potsdam, NY
Midwest Research Institute: Kansas City, MO
Johnson Space Flight Center: Houston, TX

TABLE 8.2 Glass Compositions and Treatments in Flight Experiments of Delbert Day

40% Ga_2O_3 . 35% CaO . 25% SiO_2

1. Hot-pressed plus SiO_2 particles. Melt at 1500°C.
2. Divitrified glass, colored drop on surface. Hold at 1500°C.

   56% Ga_2O_3 - 44% CaO
3. Divitrified glass. Hold at 1500°C.

   33% Na_2O - 67% B_2O_3
4. Glass + Bubbles. Hold at 900°C.

   45% Na_2O - 55% SiO_2
5. Divitrified glass. Hold at 1350°C.

TABLE 8.3 Sample Compositions and Heat Sequence in Flight Experiments of Robert Doremus and Dan Elleman

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Samples: Glass spheres 1 cm in diameter with a small boss and a bubble about 2 mm in diameter.

Glass Composition: 62 mol% ZrF_4, 33% BaF_2, 5% LaF_3

<u>Heating Schedule:</u>

1. Heat to 600°C. Glass should soften above 300°C, then crystallize, and finally the crystals melt above 540°C.
2. At 600°C. Measure surface tension and viscosity of melt by oscillation and rotation of sample. Observe bubbles.
3. Crystallize glass on cooling.

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9. SPHERICAL SHELL TECHNOLOGY AND SCIENCE

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ABSTRACT

Various aspects of a new technology for the rapid and economical production of spherical shells composed of metal, glass, or plastic are described. Topics presented include principles of production, materials considerations, post-formation processing, and certain areas of supporting science. Examples of products are shown.

INTRODUCTION

The production method to be described operates by action of the fluid-dynamic and capillary forces which prevail when a liquid material is extruded through an orifice to form a jet. In particular, the nozzle is annular in form, with one of two embodiments used by us being shown in Figure 9.1. The shell material issues through the outer passageway and the fill material, which may be either gas or liquid, through the interior. Hydrodynamic instability causes pinchoff of the jet with concomitant encapsulation of the fill material. Surface tension provides a spherical form to the resulting hollow or compound droplets prior to solidification and before the termination of free-fall. Solidification occurs in our work by freezing of the shell material, but it could be by chemical action. Noteworthy features of the method are that suitable nozzles are relatively simple fabricate, that the size of the nozzle governs the size of the product, that the shell production rate for a given nozzle is very high, and that a wide range of materials can be accommodated.

Gravity will later be shown to play an important role in the precision of shells ultimately producible by the present methods. Limitations on accuracy arise because falling shells are subject to aerodynamic drag forces during cooling and solidification. Thus, processing in the zero-g environment of space might be necessary in the fabrication of such products as inertial confinement fusion (ICF) targets. However, most potential applications carry requirements which can be met in a 1-g environment.

The objectives of the present work were therefore set with an eventual production in space in mind, but with Earth-based production as a more immediate need. Accordingly, we are: (1) developing the fluid-flow methods; (2) examining various materials problems as they relate to the fabrication of nozzles and to the quality of the products; (3) developing facilities necessary for optimizing the free-fall and solidification phases of production; (4) conducting analytical and numerical studies on the fluid flow; (5) identifying potential applications for the products and dealing with the requirements pertaining thereto.

The applications being considered include: ICF targets, high-strength light-weight structural materials, encapsulation of phase-change materials for heat storage, encapsulation of hazardous materials, insulation materials, neutrally buoyant catalytic agents, recyclable filter materials, fire retardant materials, explosives and solid propellants, heat transporting slurries, and shock absorbing armor plate. The requirements on size, materials of composition, and geometrical precision vary considerably with the specific uses. Further description of the first three of the potential products will be given and will serve to illustrate both the range of requirements and the attainable specifications.

1. Hollow Jet Instability

When the liquid and gas velocities for the nozzle shown in Figure 9.1 are set within certain ranges the hollow jet exhibits a remarkable instability which we were apparently the first to observe and exploit. The instability spontaneously generates large amplitude axisymmetric oscillations which culminate in pinchoff of the jet within approximately five diameters, causing an encapsulation of the gas within the liquid. Figure 9.2 shows in detail the encapsulation cycle for a flow of water, and Figure 9.3 shows a more general aspect of the process. The latter photograph has been rotated for display, but the process works equally well for any orientation of the nozzle. Under typical conditions, neither gas nor liquid is lost; conversion is complete. Moreover, a frequency stability and corresponding uniformity in shell mass exceeding one part in 103 is observed.

We have now studied this instability extensively and have shown that for the self-sustained oscillation to occur the liquid velocity must exceed the speed of capillary waves present upon the cylindrical sheet emerging from the nozzle. This is given by $(2\sigma/\delta\rho)^{1/2}$, where σ is the surface tension, δ is the liquid sheet thickness, and ρ is the liquid density. Also, it is necessary that the gas velocity exceed that of the liquid by a factor of between approximately two and ten. The instability can be understood and, to some extent, predicted by a static analysis of the capillary forces. By inspection of the third frame of Figure 9.2, the intermediate configuration may be regarded as being composed of a spherical bulb which is connected to the nozzle by a cylindrical neck. The pressure required to sustain the spherical bulb is $4\sigma/Rs$, where Rs is the radius of the sphere. At the same time, the pressure required to sustain the neck against collapse is $2\sigma/Rc$, where Rc is the cylinder radius. According to this, the neck closes when the bulb becomes twice the diameter of the former. Experiments show that the shells are in fact approximately twice the nozzle diameter.

The foregoing indicated the means by which the product diameter is controlled. The wall thickness is set by either of two adjustments: by change of the nozzle annular gap dimension or by change of the gas flow rate. Increase of the latter increases the formation frequency, providing less material for each shell and rendering it thinner. It may be inferred from the simplified analysis that the formation frequency depends upon both geometrical and material properties. Frequency ranging from 50/s to 4000/s have been measured for shells between 8.0 mm and 750 μm. Multi-orifice designs could accommodate very high production rates.

2. Theoretical Studies

Numerical modeling of the hollow jet instability is currently being completed. In this, the hollow jet is replaced by an infinitely thin surface with specified mass per unit area, with specified velocity of issuance, and with appropriate surface tension. Viscosity is neglected. Finite element analysis is applied to a suitably chosen initial configuration, and the temporal development computed. It is required that the initial configuration be reproduced exactly at a later time equal to one period. Figure 9.4 presents the computed behavior for conditions approximating those of the experiment shown in Figure 9.2. This and other computations show that the essential physical mechanisms are understood and that results which compare favorably with experiment are obtained.

Also, we have completed an analytic stability of an annular jet flow, i.e., one which has either a gas or immiscible liquid flow within a surrounding liquid jet. Parallel-flow, inviscid, small-perturbation approximations apply. Although the analysis is more applicable to the liquid-liquid case mentioned later, it is interesting to note that the predicted wavelength for the liquid-gas case corresponds to a frequency in agreement with that seen in the experiments.

3. Shell Sphericity and Concentricity

The interior and exterior shell surfaces are readily understood to tend toward spherical form under the action of surface tension. There are, however, two sets of forces which may limit the degree of sphericity attainable prior to solidification: those of aerodynamic drag during free-fall and those due to non-Newtonian behavior of the liquid. These are discussed below. For many applications concentricity is as important as sphericity. Unfortunately, we do not understand the forces which promote centering, but, fortunately, we observe that solidified shells possess fair to excellent concentricity.

We have performed considerable experimental and theoretical work on the matter of centering. In one experiment, a rigid, heavy sphere coated with a layer of liquid was levitated acoustically. When a small amplitude oscillation in the vertical direction was induced by modulation, the solid immediately rose against gravity to the center of the liquid. In another experiment, a shell formed of oil of slightly different density than the interior and exterior water about it assumed concentric form when caused to oscillate in any of several natural modes. These tests demonstrated conclusively that oscillation plays a role in centering. It is to be noted that the pinchoff occurring in the shell formation process is beneficial because it induces a ringing oscillation of the shells as each filament interconnecting adjacent members breaks. Theoretically, we have performed an inviscid analysis to determine the centering force due to shell oscillation. Although valuable information on oscillation modes and frequencies has been obtained, conclusions on the centering force have proven elusive.

4. Materials Considerations

The physical properties of materials must be considered in the design and fabrication of nozzles and in selecting materials for shell fabrication. Many molten shell materials of interest are extremely corrosive, so that compatibility with nozzle materials is a foremost concern. We use stainless steel nozzles for forming shells of low melting temperature materials such as plastics and we use graphite for materials such as glass and aluminum. The fragility of graphite places a lower limit on the size of nozzles which can be fabricated, and hence on the size of the product. There is a critical need for more suitable nozzle materials.

In addition to corrosiveness, a property of molten glass of great concern is the viscosity-temperature characteristic. Although the hydrodynamic instability of the shell formation mechanisms remains effective with liquids of substantial viscosity, that property is important because it may require pressures for extrusion of the melt which exceed the strength of, say, graphite nozzles. Fortunately, moderate viscosity glasses are available. Liquid metal jet flows, on the other hand, are subject to chemical reactions such as oxidation which form a strain-resistant skin layer and result in non-Newtonian behavior. Such layers strongly interfere with the jet instability process, and for that reason attempt is always made to eliminate reactive gases in the proximity of the nozzle orifice. Also, fluxing agents have been found to be effective in the elimination of this problem.

5. Problems Which Are Specific to Shell Size

As indicated, the production of small shells requires nozzles which are "doubly small." Because fabrication becomes ever more difficult with progressive miniaturization, and because of nozzle materials constraints, an alternative nozzle design, shown in Figure 9.5, is employed for producing hollow shells below 500 μm, approximately. There, the fill-gas tube terminates inside a convergent nozzle, offering the advantage of a relaxed mechanical tolerance on the concentricity and easing the fabrication to small nozzles. Although this design has a general similarity to that described earlier, the fluid-dynamic process is fundamentally different. Here, a gas bubble adheres to the end of the gas tube until it attains a critical size and is then swept downward by the rapid flow of liquid. The cyclically injected bubbles alter the flow impedance as they pass through the nozzle, rendering the flow periodic. This gives a nodular form to the issuing column, and pinchoff occurs at the necks between bubbles. It is expected that shells in the 100-μm size range will be produced by use of this design.

Shells in the one-centimeter size range are also sought. The ground-based production of these presents an interrelated set of problems which arise from heat transfer and gravitational effects. Solidification times are high because of the large mass, and distortion of the exterior liquid surface due to aerodynamic forces becomes significant as the shells accelerate in free-fall. Worst of all, the captured gas rises within the shell, thereby producing a decentering of the interior surface. Because of these problems, successes to date in the production of metallic shells within ambient air have been limited. Recent analytical studies of heat transfer and drag have pointed the way to greatly reducing the severity of the problems. Figure 9.6 presents the history of solidification and acceleration deficit due to aerodynamic drag for an 8.0-mm shell of low-melting alloy falling in chilled helium at conditions typically attained in the drop tower facility described below. Here the shell experiences no more than 0.07 g before solidification is complete, in contrast to the case of solidification in room air, where terminal velocity is sustained before solidification is complete.

6. Facilities

Notable among several facilities developed in order to gain control over the various problems arising from chemical effects, drag, and heat transfer is the 14-m cryogenic drop tube shown schematically in Figure 9.7. The tube may be filled with suitable gases at pressures as low as 10^{-5} Torr. Three temperatures zones are incorporated. The first is designed to be set at a level near the melting/liquidus temperature of the shell material to allow the shells to become fully formed and rendered symmetrical. The second is 10 m long and is cooled to LN_2 temperature, while the third can be set to an intermediate temperature if desired.

7. Examples of Products

To date, shells have been produced of tin, lead, aluminum, a gold-lead-antimony alloy, glass, and plastic. Sizes have ranged from 250 μm to 8.0 mm, approximately, but not all sizes have been produced from each of these materials. Our earliest success was with tin, chosen because its ratios of surface tension to density and of viscosity to density do not differ greatly from those of water. It quickly became apparent that the physics of the process generally favor the production of small shells in a 1-g environment because solidification is rapid and surface tension forces are high. Figure 9.8 displays a digitally enhanced x-ray shadowgraph of a 750-μm specimen. Shells of its population are buoyant in water, implying a wall thickness of four percent of radius, approximately. Quality in all respects is high. Success in forming aluminum shells by use of graphite nozzles has followed readily. Also, nozzles of that material have been employed in the production of glass shells, with a number of successful runs having been accomplished. Figure 9.9 presents an x-ray shadowgraph of several 300-μm shells; 2.5-mm shells have also been produced in quantity.

ICF target shells must be dimensionally precise, smooth, homogeneous, and of high strength. Studies were therefore initiated on the aforementioned gold-lead-antimony alloy as a model material. This alloy has the advantage of being among those metals most readily solidified in an amorphous state. Such metals are devoid of crystal structure and show high strength and surface quality. In preliminary work success in forming non-hollow droplets as large as 1.5 mm came quickly. The forming of amorphous shells has been more difficult because cooling rates adequate to prevent crystallization must be attained without distortion of the liquid specimen. The cryogenic drop tube has proven adequate, and a photograph of a shell produced there is presented in Figure 9.10.

8. Extended Applications

It is expected that several of the foreseeable applications will require shells which are non-hollow, or which require further processing before use. Relevant work is mentioned here. One potential use of shell technology is in heat storage. Many heat engines and refrigerators use regenerator subsystems wherein heat is stored temporarily during part of the cycle. Small, solid metal spheres are often used. If shells were filled with a salt or other phase-change substance, the heat capacity of a given regenerator volume would be substantially improved. A somewhat similar application is in space garments for extravehicular activity, where body temperature control is critical. It is envisioned that metallic shells filled with a paraffin of appropriate melting range could be incorporated in the garment construction without unduely restricitng the astronaut's activity.

Current work relating to the encapsulation of heat storage media is both experimental and theoretical. For the former, the annular jet technique is again employed, but here the jet instability is much weaker because of the absence of a hollow core, so that periodic stimulation to induce uniform pinchoff is appropriate. The encapsulation of a paraffin in Cerrobend alloy (70°C melting point) is being attempted. Also, fundamental studies on the heat transfer and melting processes within a spherical shell have been completed recently, resulting in a dimensionless correlation of the heat transfer data. Figure 9.11, from that work, depicts the progression of the liquid-solid interface. Theoretically, the amplification rate of the annular liquid-liquid jet has been predicted, showing a substantial increase with respect to the familiar Rayleigh instability. Also, the effect of the narrowing of the downward-directed jet under acceleration of gravity has been studied.

Aluminum shells may prove useful in the production of a light-weight structural material. As shown in Figure 9.12, performed shells would be arranged in a close-packed hexagonal manner within a skin of similar material prior to sintering. Uniformity in diameter of each shell would assure good contact with its twelve neighbors. Shell thickness would be set in accordance with bond strength, and, as an added feature, individual shells could be pressurized for added compressional strength. The sintering technology for this has been successfully applied and samples of the core material prepared and evaluated.

An acoustic levitation instrument development by us provides the capability for applying precision coatings to shells. Many ICF designs, for example, employ a multilayer configuration, with each layer being subject to strict limits on uniformity. Here, a focusing acoustic radiator suspends the shell against gravity while the coating is applied. Figure 9.13 shows the radiator, a 30-cm aluminum dish formed as a segment of a sphere with 130 PZT crystals attached to the under surface. The crystals are driven at their resonant frequency, approximately 100 kHz, to form an intense acoustic field at the center of curvature. A small reflector placed there produces a standing wave which supports the specimen. The coating is applied by atomization, and modulation of the field oscillates the sample to render the coating uniform before solidification. Excellent results have been obtained.

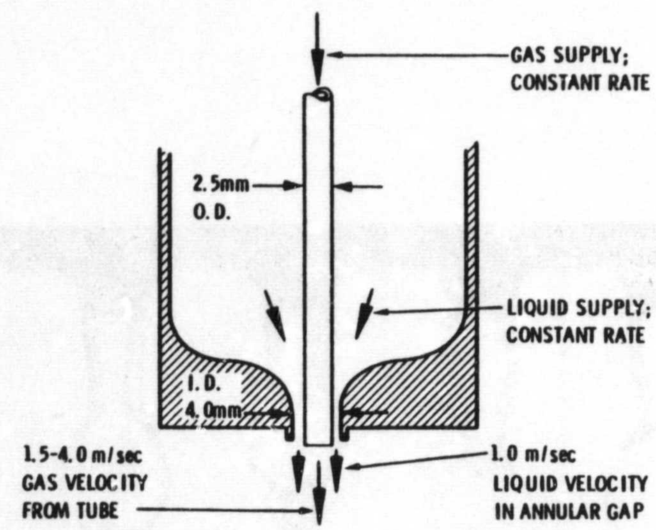

FIGURE 9.1. Nozzle schematic, with typical flow conditions indicated.

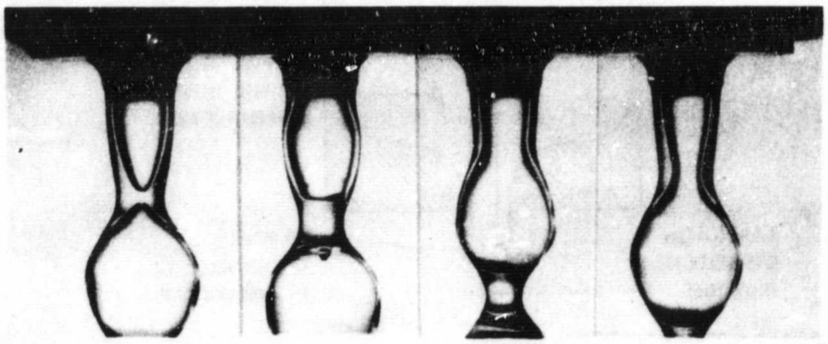

FIGURE 9.2. Shell formation cycle in water.

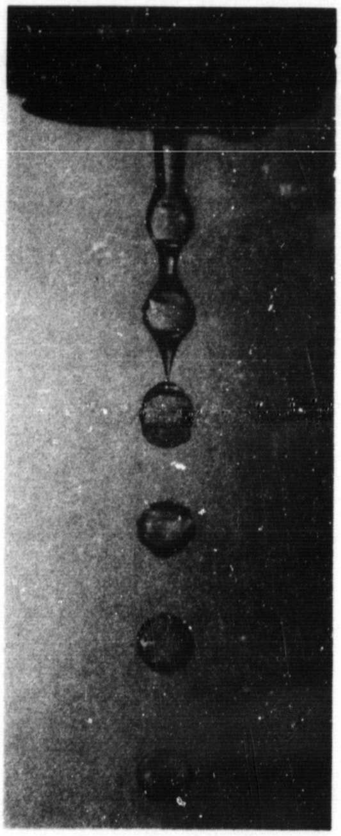

FIGURE 9.3. More distant view of shell formation.
Photograph rotated 90°.

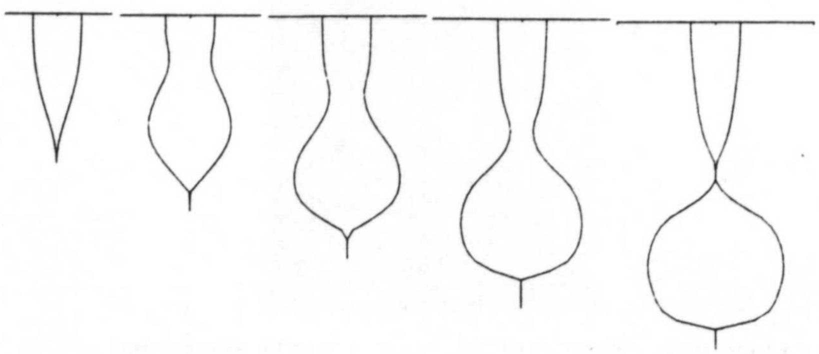

FIGURE 9.4. Computed shell formation cycle.

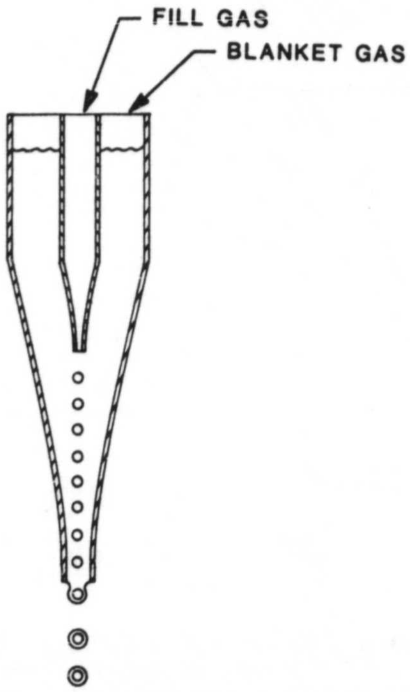

FIGURE 9.5. Nozzle configuration for production of small shells.

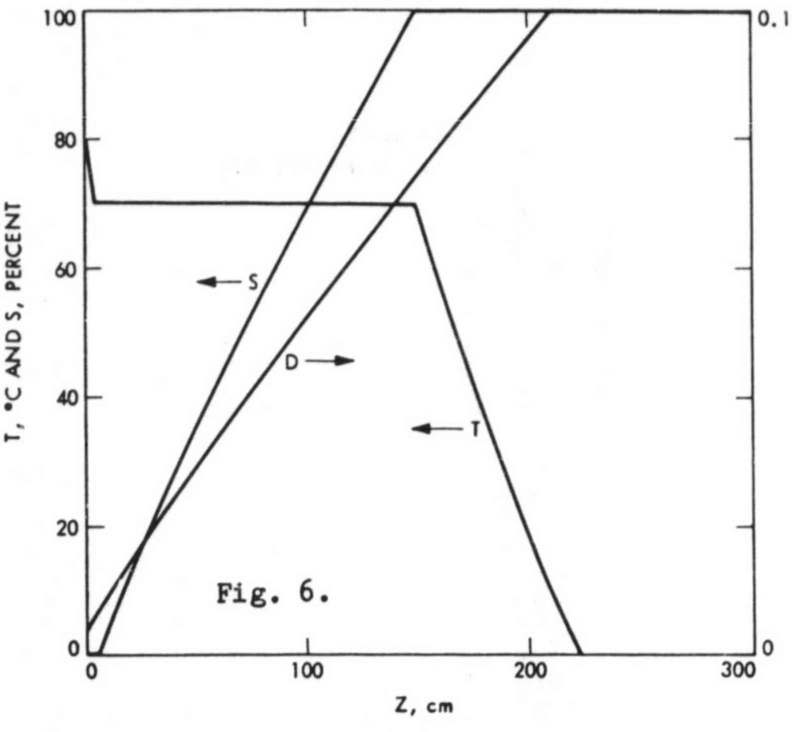

FIGURE 9.6. Calculated temperature, T, solidification percentage, S, and drag-induced acceleration deficit, D (in units of G), for an 8.0-mm shell of low-melting alloy (Cerrobend) falling through distance, Z, in He gas at -200 °C.

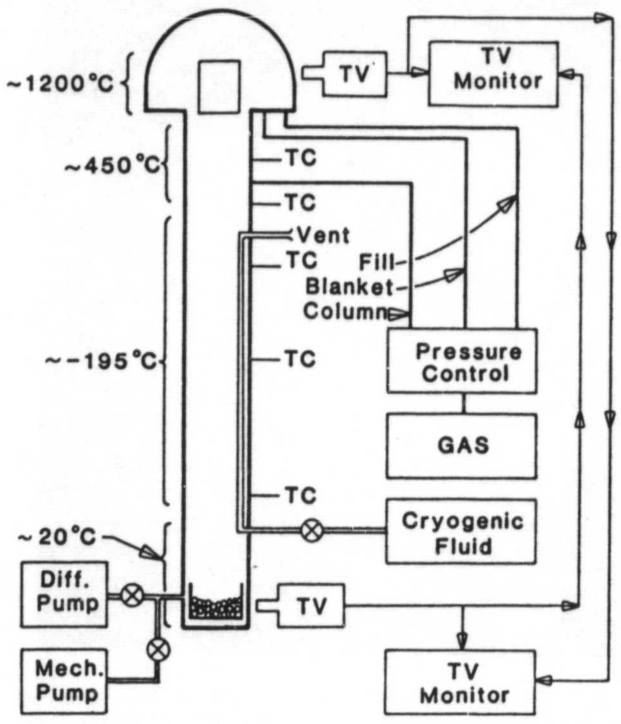

FIGURE 9.7. Schematic of 14-m cryogenic drop tower.

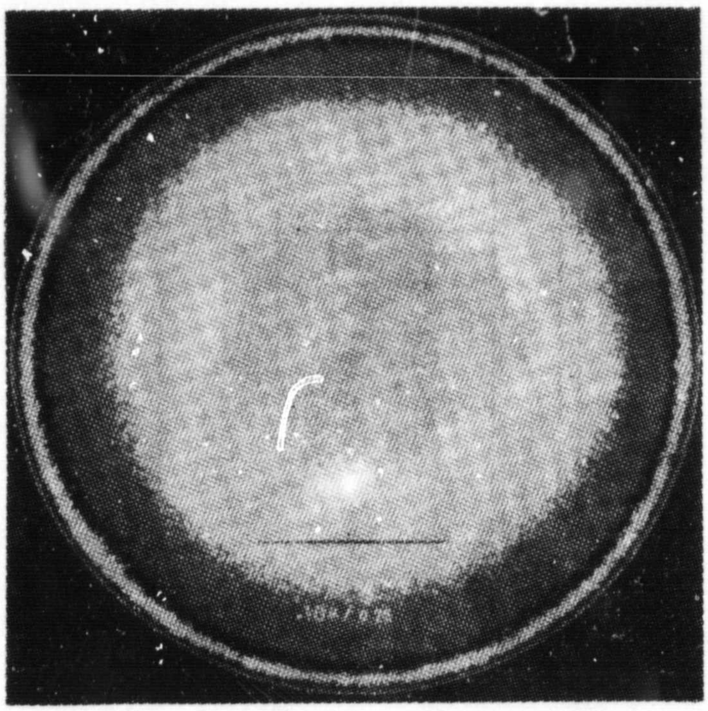

FIGURE 9.8. X-ray shadowgraph of 750-μm tin shell. From a color photograph prepared by JPL's Image Processing Laboratory.

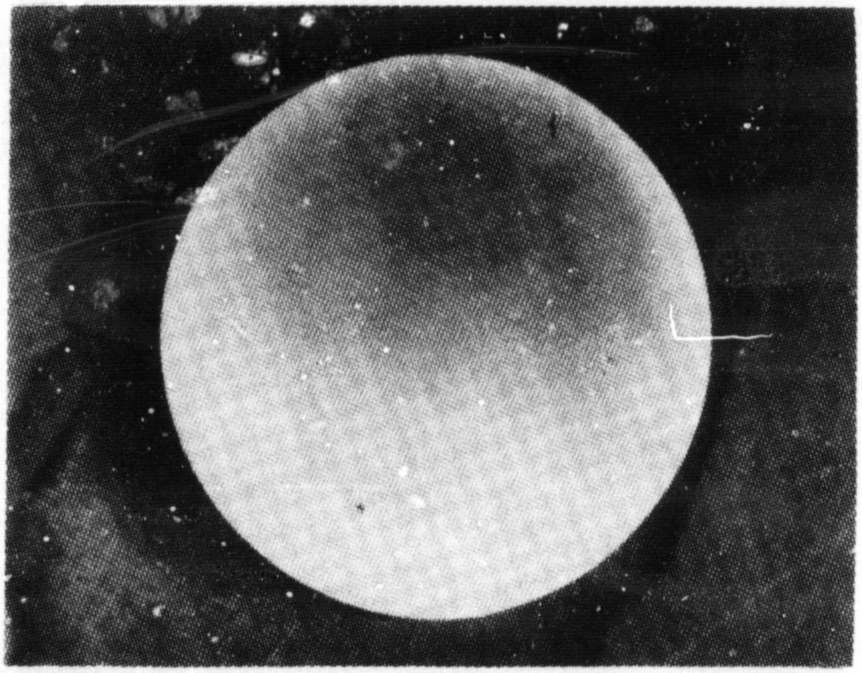

FIGURE 9.9. From an x-ray shadowgraph of a large array of high-lead content glass shells. The average diameter is 300 μm.

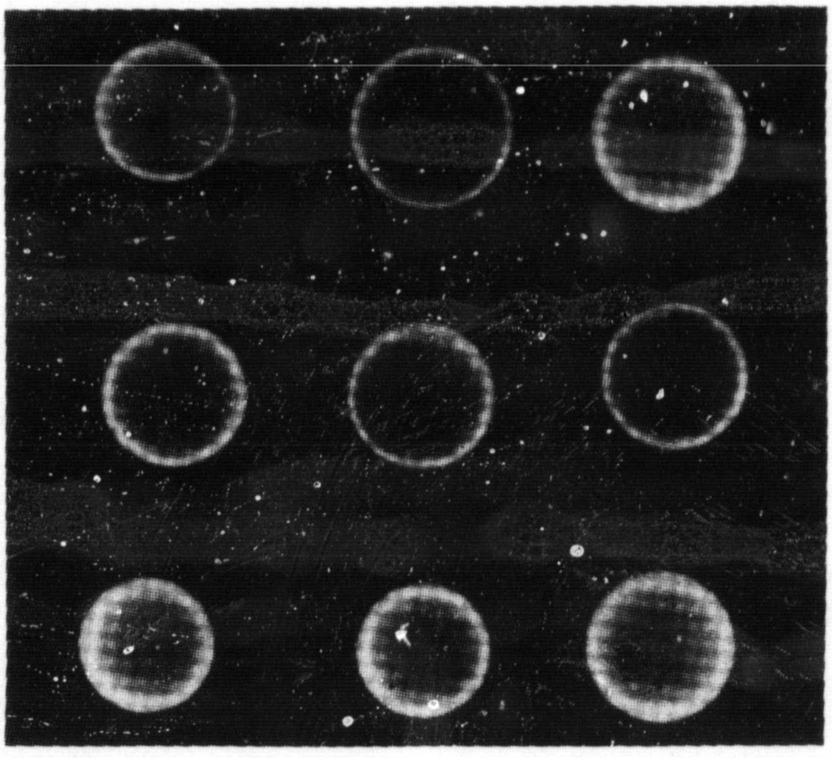

FIGURE 9.10. SEM photograph of AuPbSb amorphous shell. 1.5-mm diameter.

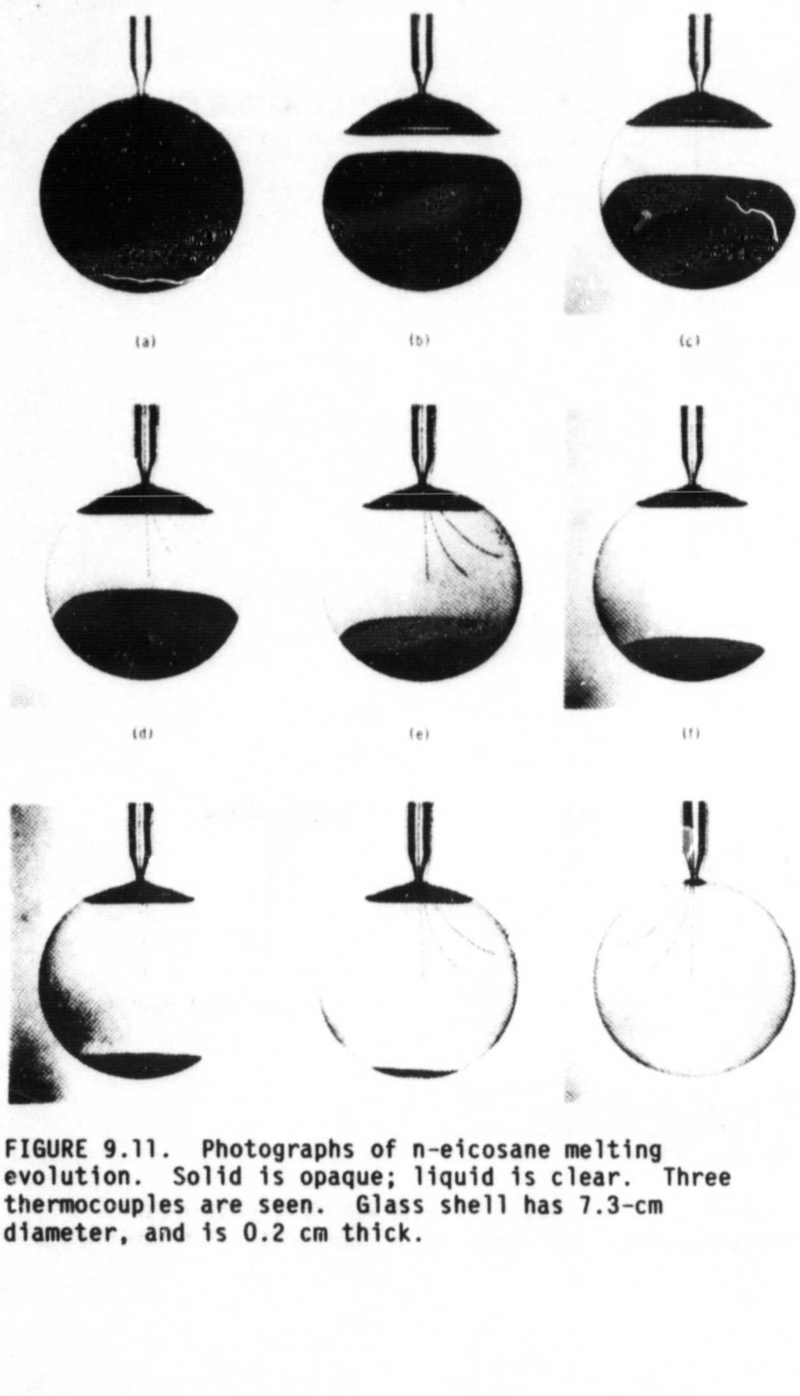

FIGURE 9.11. Photographs of n-eicosane melting evolution. Solid is opaque; liquid is clear. Three thermocouples are seen. Glass shell has 7.3-cm diameter, and is 0.2 cm thick.

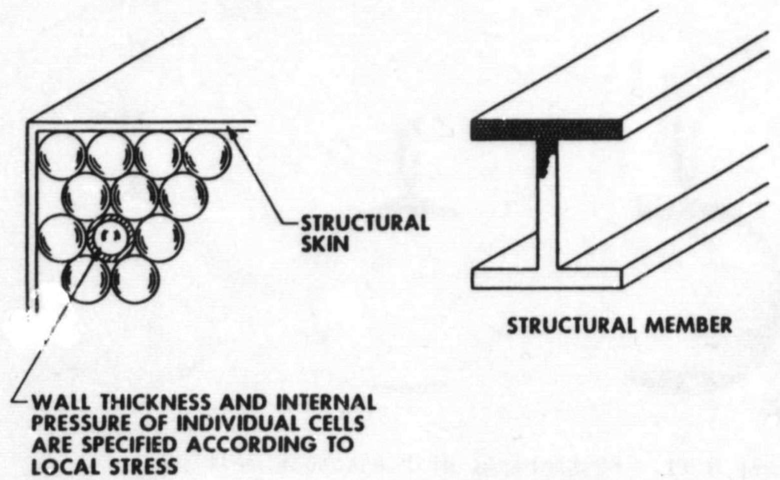

FIGURE 9.12. Schematic of a proposed light-weight structural material composed of sintered aluminum shells.

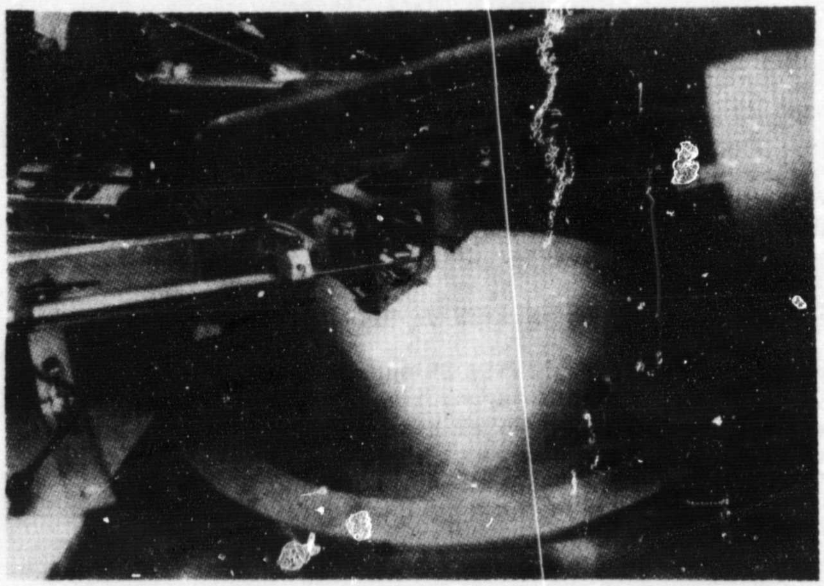

FIGURE 9.13. Focusing acoustic levitation instrument. Radiator dish is 30 cm in diameter.

10. MICROGRAVITY SCIENCES AND APPLICATIONS: AN OVERVIEW OF NASA's PROGRAM AND EXPERIMENTS IN MICROGRAVITY BIOTECHNOLOGY

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This requested overview has as its purposes (1) the review of the broad scientific objectives of microgravity biotechnology, (2) the general content of NASA's program in microgravity biotechnology, and (3) the applications and commercial potential of microgravity biotechnology.

BROAD SCIENTIFIC OBJECTIVES OF MICROGRAVITY BIOTECHNOLOGY

The Office of Microgravity Sciences and Applications (MSA) of the National Aeronautics and Space Administration (NASA) is guided, among other resources, by Discipline Working Groups (DWG's) sponsored by the Universities Space Research Association (USRA) under contract with NASA. The Microgravity Biotechnology DWG recently articulated three broad scientific objectives: (1) utilize the microgravity environment to enhance certain separatory processes for the purification of biological materials for therapeutic and diagnostic application to diseases; (2) utilize the microgravity environment to enhance crystallization of proteins and other biological materials for the design of new pharmaceutical agents and protein engineering research; (3) obtain basic information on the effects of the microgravity environment on certain biological processes in order to develop new therapeutic agents and to provide new insight into normal and abnormal physiology at the cellular and whole organism levels.

Practically all activities of the MSA Office fall into these three categories, and the discussion that follows will adhere to these subdivisions of activities.

GENERAL CONTENT OF NASA'S PROGRAM IN MICROGRAVITY BIOTECHNOLOGY

Each of the above broad objectives has a history of accomplishments on the ground and in space, and several projects have been undertaken in each category to test specific hypotheses relevant to each of these objectives.

(1) Utilize the microgravity environment to enhance certain separatory processes for the purification of biological materials for therapeutic and diagnostic application to diseases.

Free-fluid processes have dominated the research efforts in this area for the simple reason that these processes are severely (100-1000-fold) restricted by gravity with respect to processing rate and purity. It is a common misconception that the current technologies are sufficient for the task of production and purification, in economically viable quantities, of genetic engineering products, synthetic peptides, cloned genes, monoclonal antibodies, and transplantable cells. These products are normally present in heavily contaminated extracts containing as many as 5000 different unwanted substances. Free-fluid 2-phase extraction and free-fluid electrokinetic separation methods have the greatest potential for rapidly achieving high purity separations of industrial quantities of modern biologicals, especially those that are difficult to manage by high-volume column or batch chromatography or high performance liquid chromatography. It is somewhat paradoxical that analytical electrophoresis, especially two-dimensional gel electrophoresis, offers the greatest resolution of any separative technique, but no form of electrophoresis or isoelectric focusing is currently used for industrial scale purification of biological compounds. Thus, separatory processes under investigation, considered in chronological order, in space flight experiments are free-zone electrophoresis, isotachophoresis, continuous-flow electrophoresis, isoelectric focusing, recycling isoelectric focusing, and two-phase aqueous polymer extraction.

Free-Zone Electrophoresis

Early experiments on Apollo flights were originally designed to test the hypothesis that convective flow and particle sedimentation would be absent in particle electrophoresis in microgravity. This hypothesis was verified, and it was discovered that electroosmosis becomes the dominant source of fluid motion in the absence of an acceleration field. More careful experiments were performed on the Apollo-Soyuz test project to test the hypothesis that electroosmosis could be prevented by coating chamber walls with neutral polymer and that living cells could be separated under sterile conditions. Experiments with test particles were used to demonstrate that electrophoretic mobilities were as predicted on the basis of earth-based analytical electrophoresis measurements. The last free-zone electrophoresis experiments were flown on Shuttle flight STS-3 to verify results of human kidney cell separations on Apollo-Soyuz and to test the hypothesis that zone sedimentation, normally caused by density instability resulting from too high dispersate concentrations, and high cell density would not interfere with electrophoretic motion. The first objective was not achieved, owing to technical failure, but electrophoretic migration of erythrocytes in microgravity was shown to be independent of cell concentration. All of the free-zone electrophoresis experiments in microgravity used particles (cells or latex spheres) as test objects, and no experiments were done with macromolecular solutions. Although no additional free-zone electrophoresis experiments are currently planned, the advent of high-capacity density-gradient cell electrophoresis (Tulp) could renew interest in this area, but the method is otherwise a low-capacity method relative to continuous-flow electrophoretic separation (CFES).

Isotachophoresis

M. Bier and colleagues performed a modest isotachophore-isotachophoresis experiment using dyes on Skylab, and a similar experiment was performed on ASTP using erythrocytes as marker particles and photographic monitoring of band migration. The hypothesis that high-density (including cells, which normally sediment) separands could be maintained in sharp bands between separator ions was tested. By using erythrocytes in the ASTP experiment, this hypothesis was demonstrated, and a geometrical configuration that would have been severely affected by convection on the earth was used successfully in microgravity.

Continuous-Flow Electrophoresis

This activity is currently the cornerstone of the microgravity biotechnology program, mainly owing to an aggressive approach taken by McDonnell-Douglas Astronautics Corp. (MDAC). The three main constraints imposed on this method by gravity are zone sedimentation, convection, and particle sedimentation (if particles or cells are being purified). Early flight experiments (ASTP and STS-4 through STS-8) were designed to test the hypotheses that the absence of convection permitted a less rigorous cooling procedure and hence a 3.5-mm-thick chamber in place of a 0.5-mm-thick chamber and that the absence of zone sedimentation permitted higher sample concentrations--up to the solubility limit (40 percent w/v for some proteins) rather than only a fraction of one percent. Without establishing any upper limits, the experiments showed a purification rate that was 730 times that possible on Earth by the same method. Experiments with particles (latex spheres, canine pancreas cells, human kidney cells, and rat anterior pituitary cells) demonstrated the obvious: particle sedimentation is absent in microgravity and does not affect cell migration as it does in any electrophoresis apparatus on Earth. Ground-based and microgravity experiments have been performed with the latter two cell types; Hymer reports in detail on pituitary cells. In electrophoretic studies of kidney cells using the ground-based Continuous Flow Electrophoretic Separator (CFES) at McDonnell-Douglas (MDAC) in St. Louis electrophoretic fractions of two cell lines were examined, and it was found that one cell line resolved into 2 populations that produce urokinase (UK), while another line resolved into 4 or 5 peaks of fractions that were high in UK production. High-activity fractions had 2-5 times as much activity as corresponding numbers of unpurified cells. High mobility fractions produced S1, the 35 K MW component, while medium and slow cells produced the 54 K MW component. Whole unseparated cultures produced plasminogen activators with 150 K, 80 K, 54 K, 35 K, and 20 K molecular weights. The isoelectric pH of S1 is 8.0-9.5 while that of S2 (54K MW) is lower. In the ground-based CFES it was found that small cells were slow (low electrophoretic mobility) and large cells were fast. It was recommended that cell cycle phase distributions be done on fractions from an experiment to see if cell cycle is a determinant of separation. A very recent Shuttle flight included an experiment to demonstrate commercial feasibility of this process. A series of NASA-sponsored experiments with macromolecular solutions was also performed by Snyder and colleagues to confirm that high concentrations of very soluble products could be purified; a potentially commercial substance (Pneumococcus polysaccharide useful in vaccines) was one of the test objects. Experiments in which conductance discontinuities were deliberately introduced showed that the low-conductivity buffer used in the CFES renders the system very sensitive to conductance discontinuities. Other problems remaining to be solved include the management of potential contamination by microorganisms whose multiplication is favored by Kennedy Space Center vehicle recycling procedures. Continued cooperation using this device as a scientific research tool for NASA-sponsored scientists is expected through the extension of a Joint Endeavor Agreement between NASA and MDAC. Several laboratories are planning future experiments with the device.

Isoelectric Focusing

Gravity interferes with free-fluid isoelectric focusing, which is perhaps the most promising preparative free-fluid method because it is an equilibrium process. Two effects are apparent in the laboratory: sedimentation of products that precipitate rapidly at their isoelectric pH and the "slumping" of partially purified bands as their density increases beyond that of the surrounding buffer (this phenomenon occurs whether bands are stratified vertically as in a density gradient or horizontally as in a rotating tube, Hjerten, Bier). Bier and colleagues, on STS-11, Feb. 1984, tested the hypothesis that these phenomena would be absent in static free-fluid isoelectric focusing of two colored proteins by taking time-lapse photographs and monitoring current and voltage in 8 small chambers. The experiments were also designed to test different fluid-stabilizing geometric configurations and to examine possible effects of electro-osmosis. Surprisingly, several chambers exhibited a current-voltage profile that indicated the presence of disturbing flow phenomena that developed around the time of establishment of the natural pH gradient of the ampholytes. This experiment is planned for repetition while continuing ground-based research is directed at an understanding of the unexpected flow patterns. Mathematical modeling of this system has been done and continues, along with simulation experiments with a rotating tube ("Rotofor").

Recycling Isoelectric Focusing (RIEF)

This process, developed by Bier, is already capable of purifying up to 10 grams (possibly more) of a protein from a relatively simple mixture. It cycles mixed ampholytes or buffers through 12 parallel chambers, and the sample is inserted into one of the 12 corresponding circulation tubes after equilibration of the pH gradient formed by ampholytes or buffers. The resolution is currently limited to one part in 12 of the total pH range, and this resolution is compromised by "slumping" of bands as they cycle through the 12 separation chambers, which are separated by nylon mesh screens. This concept is about to be developed into a 50-chamber system, and a microgravity version is being designed for flights in 1986-87. It is expected to test the hypothesis that industrial-scale isoelectric purifications can be performed (under ANY conditions).

Two-Phase Aqueous Polymer Extraction

When two polymers are dissolved in water above a critical concentration of each, two immiscible phases form. Some substances are differentially soluble in the two phases. Thus Albertsson found that it was not only possible to separate biological macromolecules by this gentle procedure, but that live cells could also withstand exposure to these solutions. The thermodynamic theory of this process has been the subject of several papers by Albertsson and by Brooks. H. Walter and D. Fisher have performed numerous cell separations with this method and have identified free-fluid affinity properties that enhance the process. Brooks has proposed that this method would be enhanced in the absence of gravity, because the interfacial energy (a significant determinant of the partition coefficient) is always balancing the gravitational potential energy (a significant determinant of cell motion in suspension). No zero-gravity experiments have been performed yet, but planning at the laboratory level in at least three locations continues, while the national DWG continues to study the potential of microgravity in improving separations. It is expected that the hypothesis that interfacial tension will be a greater determinant of partition will be tested within the next year.

Competing Processes

Although free-fluid processes hold great promise for high-volume purifications, processes other than those just mentioned are undergoing rapid development. High-performance liquid chromatography (HPLC) can be used on an industrial scale for a limited number of macromolecular substances today, and this technology, especially its applications to proteins, is evolving very rapidly. Large-volume serial columns are in use by Pharmacia and Polysciences; for example, refractory column substances are being developed by Perkin-Elmer and Toyo Soda, and hydroxylapetite columns that can purify monoclonal antibodies directly from ascites fluid (Juarez-Salinas et al.) have been developed recently. Preparative affinity methods are in use on a small scale, but scale-up simply awaits inexpensive procedures for producing and recycling ligand materials. Once this is achieved, affinity magnetic filtration (Kronick and Gilpin) can be expected to become routine, at least for cells. Although the electrophoretic purification of cells in the presence of gravity will always be ambiguous, pragmatic means of scaling up density gradient cell purification are under development (Tulp) using the reorienting gradient procedure (Shortman).

(2) Utilize the microgravity environment to enhance crystallization of proteins and other biological materials for the design of new pharmaceutical agents and protein engineering research.

Computerized chemistry has made possible the synthesis of substances that react with specific stereochemical species whose three-dimensional structures are known. Similarly protein engineering methods (solid-phase synthesis and semisynthesis guided by computer-processed structural information) have made possible the synthesis of substances that mimic peptide hormones, for example. In both cases high-quality protein structural information forms the basis for proceeding (Ulmer). High-quality protein structural information depends on good x-ray diffraction patterns from protein crystals of adequate size and high quality.

Crystallization of Simple Substances

Experiments were performed on Skylab and ASTP to demonstrate the effect of the lack of convective flows on the size and quality of organic and inorganic crystals growing from solution. These experiments were analogous to similar experiments on the crystallization of metals and semiconductor substances from non-convecting melts on these and earlier spaceflights. The hypothesis that the absence of adverse concentration gradients and the resulting buoyancy (convection) would improve product uniformity and crystal size was successfully demonstrated.

Crystallization of a Standard and a Nonstandard Protein

The Spacelab I experiment of Littke and John was designed to test the hypothesis that crystals of proteins, like those of simpler substances, are larger and of higher quality when grown in the absence of convective flow and sedimentation in microgravity. Two proteins were crystallized. Lysozyme (MW = 14,307) had been solved by D. Phillips nearly 20 years ago with and without its substrate. Instead of the usual small needles, large blocks formed, up to 1000 times the volume of typical earth-grown crystals. Similarly, beta galactosidase (whose three-dimensonal structure is unknown owing to the lack of suitable crystals) crystallized into flat needles 27 times as large as any achieved previously.

Planned Crystallization Experiments and Service

On the basis of a few experiments, some 7 or 8 working groups have proposed new experiments in protein crystallization on the Space Shuttle for 1985. If these are successful, a microgravity crystallization service will probably evolve. The experiments are, by the standard of any kind of space experiment, very cheap. A two- or three-chamber vessel with valves separating the chambers and opening at a suitable time to permit the precipitant to diffuse into the protein solution is all that is needed. In many cases only enough temperature control to keep the samples liquid below 30°C is necessary. As Littke and John point out, the septum between chambers should have as great a cross-sectional area as possible, and some substances may benefit from crystallizing in a linearly increasing temperature gradient. It is estimated that a few hundred samples could be crystallized in a mid-deck locker and about 100 samples could be accomodated, with necessary controls, in a "Get-Away-Special" cannister. Certain peptide hormones and oxidative enzymes are among early candidates for study.

Protein Crystallization Theory

Although over 100 proteins have been crystallized for successful structure studies, very little work has been done, relative to the extensive studies on solid-state and electronic materials, to determine the mechanisms, thermodynamics, and kinetics of protein crystallization. For this reason, NASA is sponsoring theoretical studies and systematic experimentation in support of the pragmatic microgravity effort. The studies of Feigelson (Stanford) will concentrate on the crystallization of the lysozyme molecule, its mechanism, thermodynamics, and kinetics--information previously of rather little interest after high-quality crystals had been obtained.

(3) Obtain basic information on the effects of the microgravity environment on certain biological processes in order to develop new therapeutic agents and to provide new insight into normal and abnormal physiology at the cellular and whole organism levels.

Several processes under exploration in microgravity biotechnology involve living cells. These are to be purified, grown, observed, or processed in microgravity in some other way. Additionally, living organisms, especially humans, but also plant and animal test objects, will continue to be exposed to the microgravity environment. As programs of fundamental biological exploration evolve, meaningful new discoveries continue to be made. These are at the subcellular level, the intracellular level, the intercellular level and at the level of interacting cellular systems, such as blood and differentiating organs.

BIOREACTOR TECHNOLOGY

The notion of a microgravity bioreactor is over a decade old (Nyiri and Toth, Mattoni). One of the many attractions of cell growth in microgravity is the potential for controlling and maintaining a constant environment around every cell--bacterial, fungal, plant, or animal. Cells would not sediment and become concentrated, nor would aeration (and the accompanying foaming) be necessary to keep them suspended. Computer-controlled, slow replacement of nutrients and growth factors would be possible in a uniform sense. Coupled to this attraction is the notion that secreting cells might behave differently in microgravity, spending more energy multiplying and secreting and less energy developing surface forces and maintaining their shape against gravity--conjectures explored but not yet proven. Finally, if one of the above-mentioned microgravity free-fluid purification processes were to be found feasible and advantageous, then an on-board bioreactor to provide on-board raw material would be useful; it could reduce the number of visits required to replenish the purification system. Cooney has described this type of integrated approach to bioreactor technology in which production begins with "feedstock input" and concludes with purified product and in which certain valuable resources are automatically recycled. Nyiri and Toth reported that cultures of Salmonella grew to higher densities in microgravity than on the ground. The first space bioreactor has been designed for animal cell growth on microcarrier beads, with microprocessor control, no gaseous headspace, circulation and resupply of culture media, and slow mixing at very low shear. A ground-based version has been used to test reactor vessel design, on-line sensors, effects of shear, nutrient supply, and waste removal from continuous cultures of human cells attached to microcarriers beads. A small space bioreactor is being constructed for flight experiments in 1985 to verify systems operation under microgravity conditions and to measure the efficiencies of mass transport, gas transfer, oxygen consumption, and control of low shear stress on cells. The desirability and effectiveness of microgravity fermentation depends, however, on the role played by gravity in vital cellular phenomena: cell growth, cell attachment, cell differentiation, and cell secretion.

Cell Growth

It has been assumed in the past that some form of graviception must be present in dividing cells if weightlessness affects cell multiplication rate. Jane Shen-Miller proposed, for example, that cells in germinating plant seedlings might sense the gravity vector on the basis of the position of the dictyosomes of their Golgi apparatus, while Pollard, and subsequently Todd, suggested that animal cells may lack organelles that experience significant Stokes sedimentation. A view is currenty emerging that suggests that relief of cells from doing work against the gravity vector through their contractile or cytoskeletal activities may enhance cell growth rates. Early observations on the microgravity adaptation syndrome in humans raised fundamental questions about the growth of cells in human subjects in weightlessness. Montgomery, supported and advised by a host of cell biologists, conducted an experiment on Skylab in which the growth of human diploid lung fibroblasts in culture, attached to a transparent surface, was monitored by cinematography and periodic fixation. Metabolic and cytogenetic evaluations were also made. No changes in growth rate, cell morphology, karyotype, or cell migration were observed. The rate of glucose consumption was, apparently, slightly depressed. This rather uninteresting but reasonable result was not subjected to further verification. In collaboration with users of the Salyut Space Station, Planel showed that tetrahymena and paramecia grew significantly more rapidly in microgravity than on Earth, and that energy consumption was somewhat reduced. He was able to explain this apparent paradox through a model in which the energy required for these ciliates to swim is drastically reduced in microgravity so that substantially more energy is available for cell reproduction.

Cell Attachment

Cogoli and Morrison, in experiments aboard Shuttle flights STS-7 and STS-8, asked whether or not cultured human embryonic kidney cells were capable of attaching to collagen microcarrier beads ("Cytodex III," Pharmacia). Within 3 hours a greater average number of cells attached per bead in microgravity, and, once attached both the flight and ground control cells grew at the same rate during the first 25 hours. Cell-to-cell attachment was also greater among the cells free-floating in weightlessness. Practical implications include the possibility of seeding microcarrier cultures in microgravity and the knowledge that cells could reattach to microcarrier in slow mixing space bioreactors if they detach from bead surfaces, as they typically do during mitosis.

Cell Differentiation

Single-cell systems differentiate by expressing a tissue-specific function in response to a chemical stimulus. Human T lymphocytes cultured from peripheral blood respond to the lectin concanavalin A by entering and completing their cell division cycle. In experiments sponsored by the European Space Agency (ESA) Cogoli and Tschopp found that this response is absent in microgravity, and its absence might explain any immunological decrements experienced by space workers, whose humoral immunity (not dependent on T lymphocyte proliferation) is unaffected by weightlessness (Voss). Experimental results such as these have important implications for differentiation-dependent biological processes in microgravity bioprocessing in particular and in manned spaceflight in general.

Cell Secretion

A widely advertised claim that interferon production by human leukocytes increased more than 20-fold in Soviet spaceflights has stimulated an interest in cell secretion in microgravity. In one of the electrophoresis experiments described above, Hymer and Grindeland examined flight control pituitary cell suspensions that had been flown but not subjected to electrophoresis in space. Flight control suspensions had a greatly reduced level of growth hormone production relative to ground control suspensions. A carefully controlled repetition of this experiment has been planned in order to confirm this finding, as the discovery of a "secretory lesion" at the cellular level could have profound implications relative to the known metabolic perturbations (i.e., reduced bone mineral production, reduced circulating growth hormone) that occur in space workers during weightlessness.

Biorheology

A rationale for studying haemorheology in microgravity has been introduced by three different working groups, who also point out that careful preflight-plan study is required before a true necessity for a zero-g study can be established. For example, experiments of Coakley and King, designed to measure shear rates of blood in a cylindrical viscometer, do not work on Earth because RBC sedimentation produces a nonuniform suspension which distorts the shear rate measurement seriously owing to the nonlinear dependence of viscosity on concentration. Three specific gravity-dependent experimental problems have been discussed:

1. RBC sedimentation, especially in pathological or polymer-treated blood.
2. Density-driven convection due to the natural density gradient of components of the experiment.
3. The role of other interfacial energies in blood vessels.

In blood, cells form rouleaux and not random aggregates in tiny vessels. Measuring viscosity under these conditions requires that the blood cell aggregates be natural (rouleaux) and that cell-free gaps not appear due to sedimentation. A planned and funded experiment in zero-g blood rheology has been developed by Leo Dintenfass of Sydney Hospital. Measurements to be made include:

1. The maximum size of RBC aggregates that form in anticoagulated blood.
2. The effects of drugs and other agents on maximum aggregate size.
3. The effects of pathological conditions on RBC aggregate size.

Aggregate size varies 10X-100X among individuals. The RBC contribution to blood viscosity is reduced in pathological conditions. Blood viscosity is minimum in 10-20 micrometers capillaries and higher in both smaller and larger capillaries. Blood viscosity does not have the same concentration dependence as it does in polymer solutions. Random aggregates (rather than rouleaux) form after a myocardial infarction. The experiment design involves placing whole blood between two plates separated by as little as 12 micrometers. The plates are untreated except for optical polishing. In flight experiments, the chamber is flushed with saline, the temperature is measured, and freshly drawn blood from one of 6 donors, diluted to 30 percent flows at 2 ml/min. Such an experiment is being scheduled for flight in the near future. Cokelet, Goldsmith, and Meiselman proposed ground-based studies of the role of RBCs and their aggregates in establishing shear rates and velocity profiles in artificial single capillaries, bifurcations, and networks.

(4) Organization of Microgravity Science and Applications research programs and projects in microgravity biotechnology.

NASA Centers and NASA Headquarters

Research administration and funding of biotechnology projects are the responsibility of the Office of Microgravity Sciences and Applications in the Office of Space Science and Applications. Funds are disbursed to university investigators whose proposals must pass scientific and technical review or to Marshall and Johnson Space Centers for their ongoing intramural and extramural projects.

Discipline Working Groups

The Universities Space Research Association organizes and maintains 6 "Discipline Working Groups" (DWG) to serve both the MSA office and university-based investigators. These bring together scientists from academia, industry, national laboratories, and NASA centers for the purposes of program assessment, planning, and review at the request of the MSA Director. The Microgravity Biotechnology DWG meets at least twice per year, usually in connection with an activity relevant to its mission, such as at a scientific meeting, a NASA Center, or USRA headquarters.

Scientific Review

The MSA office retains a list of scientific reviewers broadly distributed through the disciplines relevant to microgravity biotechnology; this list is augmented continuously by suggestions from principal investigators and from the DWG. Their role is occasional review of the program in general and evaluation of proposals submitted by potential principal investigators.

University Bioprocessing Centers

Through a recent Congressional mandate the MSA office has funded two university-based bioprocessing research centers, whose role is to identify and explore microgravity biotechnologies, compare them with the best corresponding ground-based technologies, evaluate them with suitable biochemical and biophysical analytical methods, and provide ground-based and space-based services using their unique combinations of facilities. One center is at the University of Arizona, Tucson, and the other is at the University City Science Center, Philadelphia.

Joint Endeavor Agreements

Commercial firms can interact with NASA programs by exchanging services for launching and operation in space. An early example is the CFES operated by MDAC, cited above. Several joint endeavor agreements in MSA are pending, and some of these are in biotechnology.

THE APPLICATIONS AND COMMERCIAL POTENTIAL OF MICROGRAVITY BIOTECHNOLOGY

Current Private Sector Participation

MDAC and Johnson and Johnson division of Ortho Pharmaceuticals are participating, through a joint endeavor agreement (JEA), in the development of a microgravity-purified product. A feature of the JEA mechanism permits the corporations to use proprietary substances and procedures in NASA projects, as in this case. Similarly, a very large JEA has been established with 3M Corporation, which plans a 10-year program in organic crystallization and solid-state film production, some of which is biotechnology related and some of which is not. Their project began on the most recent STS flight.

Anticipated Private Sector Participation

Joint endeavor agreements with Instrumentation Technologies Associates of Philadelphia and other small firms are in the planning stage. In some cases, where microgravity research is justified, the JEA mechanism will assist in development of such biotechnolgoy firms. Private sector participation was set as a condition for the awarding of grants to the university-based bioprocessing centers. Companies involved in the activities of these two centers include, but are not limited to Kureha Chemical Industries, Cobe Laboratories, Schering-Plough Corp., Ionics Inc., Abbott Laboratories, and Hana Laboratories (University of Arizona). American Diagnostics, Pharmacia, Immunocon, Philadelphia Biologics Center, Polysciences, Instrumentation Technologies Associates, Smith Kline and Beckman protein crystals in space could play a key role in advancing this technology.

Protein Crystallization as a Special Case

Exceptionally widespread interest has developed in the past weeks in protein crystallization in microgravity. At least two consortia involving universities and industry have been formed, and proposals for microgravity experiments are pending. Firms that are involved include Schering-Plough, Upjohn, Johnson and Johnson, and Instrumentation Technology Associates.

Crystallographic studies of proteins and nucleic acids are of considerable interest in the pharmaceutical industry, since the structural findings from them are useful in designing drugs that bind to specific sites. Protein crystallography is also of importance to pharmaceutical, chemical, and biotechnology firms that are involved in protein engineering efforts (Ulmer). At least two specific scientific laboratories have expressed interest in nucleic acid crystallization in microgravity. Growth of suitable macromolecular crystals is a major hurdle in applications of protein crystallography, and the development of optimum methods for growing protein crystals in space could play a key role in advancing this technology.

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11. CONTINUOUS-FLOW ELECTROPHORESIS (CFE) AT UNIT AND MICROGRAVITY: APPLICATIONS TO GROWTH HORMONE (GH) RESEARCH AND DEVELOPMENT

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I. INTRODUCTION

Growth hormone (GH) is an interesting molecule. It regulates a number of key biological processes; e.g., it (a) enhances body growth; (b) increases protein synthesis (amino acid transport, ribosome number, mRNA synthesis); (c) decreases carbohydrate uptake; (d) increases mobilization of fat; and (e) enhances mitogenesis in hemopoietic, muscle, and liver tissue.

The amino acid sequence and structure of the mammalian GH molecule has been known for some time. Human GH (hGH) contains 191 amino acids and has two intramolecular S-S bonds. One S-S bond pair is located between half cystines at positions 53 and 165, the other between 182 and 189. The resulting large disulfide loop is a characteristic future of the pituitary somatomammotrophins.

There is good experimental evidence to suggest that the multiple biological actions of GH are produced by different parts of the molecule. hGH is extremely susceptible to proteolytic attack within a short segment between residue 134 and 149 on the disulfide loop. Enzymes such as plasmin, thrombin, trypsin, or bacterial protease cleave at several sites within these residues to yield a 2-chain structure linked by a S-S bridge. Growth promoting activities, as well as diabetogenic activities, are often enhanced after cleavage.[1] It has also been shown that the 2-chain molecules have sulfation factor activity; i.e., somatomedin-like properties may reside within the GH molecule.[2,3] Reduction after thrombin exposure yields an entire 1-133 N-terminal fragment which stimulates DNA synthesis in rat cartilage in vitro.

These observations challenge the dogma that GH exerts its growth promoting activities by stimulating release of somatomedins from the liver. They also raise a number of intriguing questions. For example, does posttranslational processing of GH activate the hormone? Does activation occur at the pituitary cell membrane, in the blood, or at a receptor in the target cell? The observations by Grindeland[4] that GH in plasma has a higher biological (B) to immunological (I) (B/I) activity than that in the pituitary gland appears to argue for hormone activation at the pituitary cell membrane.

The cell type in the pituitary responsible for production of GH is the somatotroph. On average, 35 percent of the cells in the gland are somatotrophs. Several years ago work from our laboratory showed that two subtypes of somatotrophs could be separated from each other by sedimentation.[5] Type I cells had a density of <1.068 g/cm^3, whereas Type II cells were of higher density, 1.070-1.085 g/cm^3, presumably owing to their full complement of dense cytoplasmic secretory granules. More recently we have discovered that the activity of the GH released from Type II cells in culture has a B/I of 5, whereas that from Type I cells is 1 (Grindeland and Hymer, 1985). Interestingly, when Type II cells are implanted into a GH deficient rat using the hollow fiber technology of Hymer et al., recipients show significant augmentation in growth and muscle mass. Elucidation of the structure of the bioactive form of the GH molecule released by Type II cells remains a long-term goal of our laboratories.

II. SEPARATION OF GH CELLS BY ELECTROPHORESIS

A. Rationale

Approximately 35-40 percent of the cells contained in the anterior pituitary gland of the adult male rat contain GH.[7] Others contain prolactin (PRL, ~20 percent); gonadotropin (FSH/LH, ~10 percent); thyrotropin (TSH, ~5 percent); adrenocorticotropin (ACTH, ~5 percent) or are fibroblasts, endothelial cells, or unidentified. Since some of these hormone producing cell classes have unique size and/or densities, cell separation techniques based on sedimentation differences have been useful in preparing enriched cell populations.[7] In general, 2-3-fold enrichments, with purities approaching 90 percent in the case of GH cells, can be achieved. For many studies, however, this purity is not good enough. We have therefore invested some considerable time in trying to determine if the different hormone cell classes bear unique surface charges, and are therefore separable by electrophoresis.

The first report indicating that GH cells might be electrophoretically separable came from our laboratory in 1983.[8] These studies showed that GH cells were enriched (on the basis of hormone content) by either density gradient electrophoresis or continuous flow electrophoresis (CFE). As seen in Figure 11.1, (a) GH containing cells are among the most mobile and (b) their mobility depends on field strength (ET). As shown in Figure 11.2, cells from the more mobile regions produce and secrete hormones in cultures; i.e., they are alive.

However, measurements of pituitary cell electrophoretic mobility by two analytic methods, viz. microscopic electrophoresis and laser tracking electrophoresis, revealed little distinction between cell classes.[8] We hypothesized that the high density of the Type II GH cell might result in sedimentation during preparative electrophoresis. If so, cell enrichments achieved by preparation gel electrophoresis might be the result of density mediated events, and not solely to differences in cell surface charge.

If the experiment were done under conditions where the (presumed) contribution of sedimentation would be minor, i.e., in microgravity, would there be any separation of GH cells at all?

B. Results of GH Cell Separation on CFE at Unit and Microgravity

We have recently completed analysis of 3 ground-based trials in addition to the cell electrophoresis experiment done on STS-8. Parameters examined and results obtained are summarized in Table 11.1.

Table 11.1 Comparison of Data Obtained in Ground-Based and Microgravity-Based CFE Pituitary Cell Separation Experiments

PARAMETER	UNIT GRAVITY	MICROGRAVITY
1. Cell profile after electrophoresis	1. Spread over ~50 tubes	1. Spread over ~50 tubes
2. Cell enrichment-immunocytochemistry	2.	2.
a. GH	a. 2-3x enrichment in mobile fractions	a. 2x enrichment in mobile fraction
b. PRL	b. 3x enrichment in low mobility fractions	b. ~2x enrichment in low mobility region
c. LH	c. 2x enrichment in most mobile area	c. 2x enrichment in most mobile area
3. Cell enrichment-intracellular hormone (RIA)	3. Parallels immunocytochemistry	3. Parallels immunocytochemistry
4. GH cell culture (RIA)	4. GH released from both high and low mobility somatotrophs	4. GH not released (?)
5. GH cell culture (Bioassay)	5. Bioactive GH perferentially released from high mobility fractions	5. Not done
6. LH, FSH, TSH cell	6. a. LH and FSH output from more mobile cells b. TSH--no evidence for separation c. PRL--output from less mobile cells	6. Not done
7. Character of GH after HPLC on gel sizing columns	7. a. M_r GH = 22 K for all regions b. more GH from high mobility regions	7. a. M_r GH = 22 K for all regions b. more GH from high mobility regions
8. Character of GH after HPLC on reverse phase columns	8. Evidence for multiple GH forms in somatotroph subpopulations	8. Evidence for multiple GH forms in somatotroph subpopulations

C. Conclusion

On the basis of the results obtained in 8 CFE ground-based trials and 1 in microgravity, we conclude that different pituitary cell classes do indeed bear different surface charge. When used in combination with established sedimentation procedures, CFE should prove quite useful for preparation of pituitary cell classes of high purity.

Why should some cell classes bear different surface charge? The report of Kaplan et al.[9] identifies secreted insulin bound to the surface of the pancreatic islet cell. If some portion of secreted pituitary hormones are also present on the plasma membrane, different mobility classes might result. Electrophoretic selection of cells with secreted surface hormone should provide a useful tool for study of the cell biology and biochemistry of hormone secretion.

III. GH SECRETORY CAPACITY OF MICROGRAVITY EXPOSED RAT PITUITARY CELLS

A. Rationale

Until the recent report by Cogoli et al.[10] there was little evidence to show that microgravity had any affect on cell function. Montgomery had shown that microgravity-exposed WI-38 human embryonic lung cells had normal rates of proliferation and motility, although glucose consumption was significantly lower in cells flown in space. Cogoli's experiment addressed the issue of microgravity and ability of human lymphocytes to divide when exposed to concanavalin A. The data in Figure 11.3 (from the Cogoli paper) document suppression of Con A-stimulated mitogenesis in microgravity exposed cells.

Cogoli concludes:

> Considering what is presently known about the behavior of cells at different g values, we can see a relatively consistent picture into which our results from Spacelab 1 fit very well. At high g, cells divide faster at the expense of reduced mobility, since energy consumption remains the same. In microgravity, lymphocytes show a dramatic reduction in proliferation rate, reduced glucose consumption,

but a strong increase of interferon secretion. WI-38 embryonic lung cells, which differ from lymphocytes in that they do not undergo differentiation steps, grow and move normally at 0 g, but they also consume less glucose. In conclusion, most of the cells investigated appear to be sensitive to gravity; the effect seems to be stronger with cells such as lymphocytes, which are transformed by mitogens from a dormant to an activated state.

The results we have obtained so far have contributed to an increase in the knowledge of the influence of gravity on basic cellular mechanisms, to clarifying certain biomedical aspects of the effect of spaceflight on the immune system, and to developing useful biotechnological processes. Although the mechanisms involved in gravitational effects on cells are still unknown and a gravity sensor has not yet been identified, we can conclude on the basis of results to date that cells are sensitive to gravity.

In addition to the CFE experiment, we also had the opportunity on the 8th flight of Shuttle to expose dispersed rat pituitary cells to microgravity. The purpose of this second experiment was to find out what effect, if any, weightlessness might have on subsequent GH secretion. Of course the results of the Cogoli experiment were unknown to us at the time (Sept. 1983). Muscle wastage in space is reasonably well documented and GH at unit gravity has important anabolic effects on muscle. This issue was addressed by Rambaut[11]:

> Among the problems associated with manned space flight is that of skeletal muscle atrophy. Degradation of skeletal muscles, especially of those used to counteract gravity on Earth, is commonly experienced in space flights. Its overt features include reduced volume of the limbs, particularly the lower extremities, and postflight reduction of muscle strength and exercise capacity.
>
> Knowledge of the exact casual factors, pathogenesis, and practical methods of prevention or amelioration of the muscle atrophy of space flight is not sufficient to permit the rational development of

dependable countermeasures that may be conveniently employed with a minimum of interference with the mission demands of space crews.

B. Results

Overall growth hormone production (i.e., intracellular and released) was 20X greater in cultured cells maintained at unit gravity versus those kept under similar conditions in space. Production of PRL, on the other hand, was similar between the two treatment groups.

That secretion of GH in astronauts may be compromised was inferred from a previous Skylab mission. Thus after 73 days in flight, serum GH levels were ~50 percent lower than preflight (1.3 ± 0.2 versus 0.65 ± 0.1 ng/ml; $P < 0.005$). (C. Leach and P. Rambant, pp. 204-216 in Biomed. Results from Skylab, 1977, Ed. Johnson and L. Dietlien). Interestingly, a depression of GH in blood of subjects maintained on strict bedrest also show a 50 percent reduction in serum GH in early phases of the experiment (R. Grindeland, unpublished).

In summary, the Cogoli data, coupled with the information on plasma GH levels in astronauts and our STS-8 experiment, implicate a microgravity-induced lesion in GH cells at the cellular level. Future studies can be expected to elucidate mechanism(s) of this interesting effect.

IV. HUMAN PITUITARY GH SECRETORY GRANULES: POSSIBLE COMMERCIAL APPLICATION OF CONTINUOUS-FLOW ELECTROPHORESIS

A. Introduction

In the Textbook of Endocrinology by Williams, one finds the following statements concerning GH in the human post mortem pituitary:

> Fortunately, the human pituitary is particularly rich in GH, and the somatotropic granule resists autolytic dissolution after death. Despite the fact that radioimmunoassays suggest a much higher content, the yield of somatotropin with present extraction methods is between 8 and 16 percent of the dry weight of human pituitaries, equivalent to 4-8 mg of hormone/gland. No significant changes in GH content with age are evident.

Since a majority of the hGH contained within the gland is in granules, it is presumably this form(s) which is extracted for eventual therapeutic use.

A number of years ago the National Pituitary Agency (NPA) was established for the purpose of providing purified hGH (extracted from human post mortem pituitary tissue) for clinical medicine. As far as we are aware, this agency continues to be the sole source for hGH. The issue of hormone availability was recently addressed in a memo from Dr. Raiti, Director of the NPA.

PITUITARIES AND HUMAN GROWTH HORMONE--CAP AND NPA--
CURRENT REVIEW

Pituitaries

1. The NPA was set up by CAP and NIAMDD in 1963. In part, this was to avoid fragmentation. It was intended that there should be a single collection program to process all pituitaries and distribute all hGH to hypopituitary patients in the U.S.A.
2. At the November 1979 meeting, the CAP reaffirmed unanimously its wish to have all its members send all glands to the National Pituitary Agency. (See The Pathologist, Feb. 1980, page 69).
3. The NPA provides all hGH of highest quality and FREE of charge to all patients.
4. The NPA needs the pituitaries not ony for hGH but also for research into FSH, LH, TSH, Prolactin, Neurophysins, Endorphins, Lipotropins, Diabetogenic factor, and so on for structure and synthesis studies as well as for clinical research.
5. The commercial companies SELL the products of any glands that they obtain. Each pituitary is worth at least $180.00 to them and perhaps $1,000.00 (when one adds in TSH, LH, FSH AND PRL each at $1,000.00 per mg).

Commercial: 1 pituitary (frozen) = 12 IU of hGH
x $15.00 per IU
= $180.00 per pituitary

NPA: Costs for hGH production and distribution are 75 cents per IU or $9.00 per pituitary. Distributed free of charge.

There are serious ETHICAL questions related to the buying of human tissue then selling the products for profit.

hGH Status

1. There is currently no waiting list of patients. We are supplying hGH to all diagnosed patients who apply through the established channels. More patients are being diagnosed and we need hGH for them.
2. Each child needs only the hGH from 30 glands for one year's treatment.

The NPA and the hypopituitary dwarfs need your help. This hGH is given free of financial exploitation, so that these children may grow.

Salvatore Raiti, Director, NPA

It is, of course, no secret that several firms in this country are producing hGH by recombinant DNA technology. It has been argued that children in the lowest 5th percentile in height may also benefit from hGH therapy. If so, it may then become necessary to increase hGH production. The results of 3 clinical trials using engineered hGH were described at the NIH-sponsored hGH symposium held in Baltimore, November 1983. In each of these trials it was reported that hormone administration resulted in patient growth. However, treatments were discontinued because ~1/3 of patients in each trial developed circulating antibodies. At the time of this symposium it was not clear if this problem was due to impurities in the hormone preparation. It is emphasized that this was the state of affairs one year ago; we are not certain if it has changed significantly since then.

In light of the information presented in sections I and II in regard to the issues of discrepancies of biological: immunological activities of GH, we have begun to investigate the quality of hGH obtained after CFE on the McDonnell Douglas device.

B. CFE of Human Pituitary Granules.

We prepare a crude mitochondrial-secretory granule fraction from human post-mortem pituitary tissue by conventional homogenization-differential centrifugation procedures. We then electrophorese the particles on the McDonnell-Douglas CFE device and collect the particles in tubes containing dilute NaOH. Since granules are not stable under alkaline conditions, hormone is released and subsequently assayed by RIA and bioassay.

Results from recently completed experiments show that:

a. Symmetrical peaks of immunoreactive GH after electrophoresis are found in some tissue samples, but "smearing" of immunoreactive GH is found in others.
b. The occurrence of symmetrical hormone peaks (intact hormone?) versus "smears" (degraded hormone?) does not correlate with time after death.
c. Hormone with higher B/I (often 20-60) activities are found in the more anodal regions.

We believe our preliminary results are important for several reasons. First: if GH molecular forms are so different from one pituitary to the next, it seems probable that hGH preparations currently administered to patients are either selected for certain forms or contain mixtures of electrophoretically distinct species. Second: if CFE can discriminate and isolate hormone with high B/I activities, then this procedure would be of benefit to the medical community. Third: these concepts seem equally applicable to bovine GH, i.e., they should have commercial applications.

V. ELECTROPHORETIC SEPARATION OF GH SECRETED BY PITUITARY CELLS IN CULTURE

It has been pointed out in Section I that some GH cells secrete hormone with high B/I activity in culture. We are beginning to apply the CFE technology to the isolation of this hormone form(s) contained in cell culture media from both rat and human (when available) pituitary cells.

VI. SUMMARY

A. Applications of CFE and Microgravity to Pituitary GH Research

Our approaches to pituitary GH research are multifaceted. They range from study of molecules to whole animals. An underlying theme, nevertheless, is that of separation. The separation of GH cells, GH-containing particles, and bioactive forms of the GH molecule by CFE all appear to offer considerable promise for understanding:

a. Intracellular packaging of hormone,
b. Secretion of hormone from the cell, and
c. Structural details of the bioactive forms of GH.

We view the role of microgravity-based CFE in these efforts as important, but not all-encompassing. For electrophoretic isolation of large quantities of bioactive hGH or bioactive bovine GH (and its ultimate commercialization) microgravity may prove essential. For isolation of GH producing cells, it may not. Nevertheless, documentation that GH cell separation occurred in microgravity was key, for it gave us the information that results from pituitary cell separation experiments are indeed based on differences in surface charge, and not on density. We now proceed with ground-based CFE experiments with greater confidence.

Finally, as discussed in Section III, data are beginning to emerge which tell us that cells in microgravity show suppressed function. The failure of microgravity-exposed GH cells to subsequently produce/release GH on Earth was entirely unexpected. In light of the pivotal position that GH plays in overall metabolism and well-being, it seems essential that follow-up studies in microgravity be conducted.

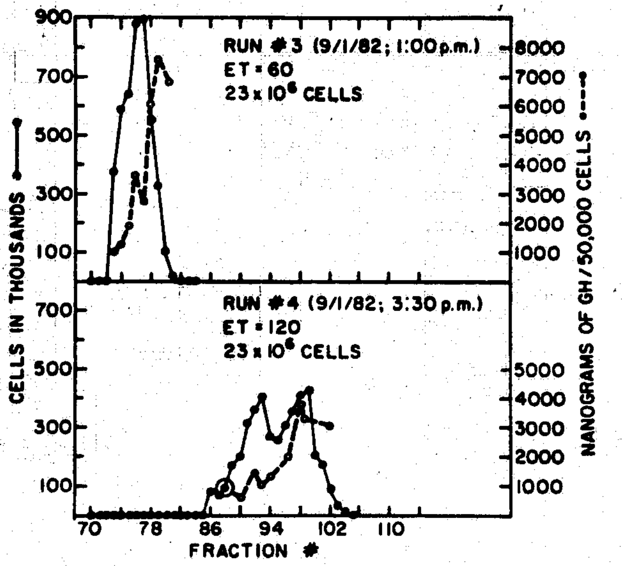

FIGURE 11.1. Electrophoretic profiles of freshly prepared rat pituitary cells and the GH content in two experiments using the McDonnell-Douglas CFES. The effects of changing ET are seen by comparing RUN 3 and RUN 4. (From Plank et al.[8])

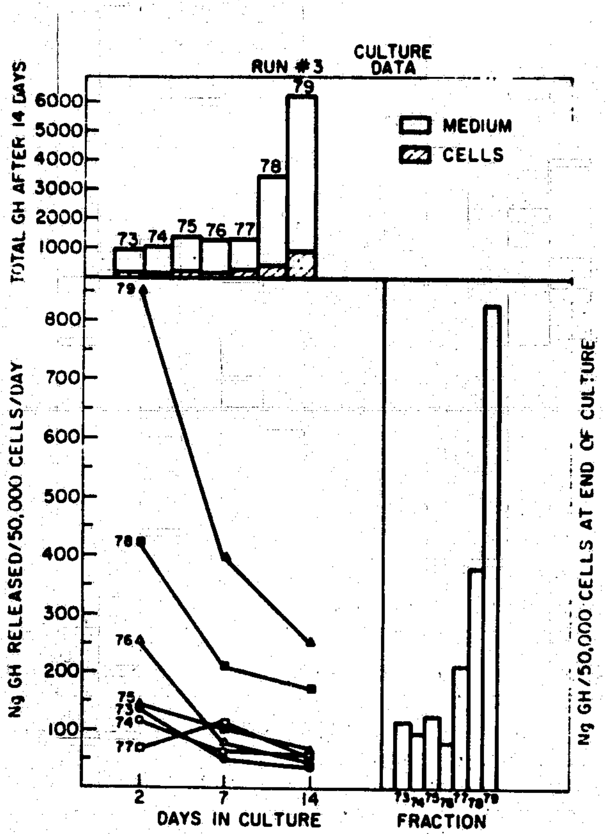

FIGURE 11.2. Production of GH by cells obtained from single electrophoretic fractions after separation using the McDonnell-Douglas CFES. Upper panel shows the total amount of GH that was found in cells and that accumulated in supernatants of the corresponding cultures of each fraction for RUN 3. Left panel is the rate of GH production per 50,000 cells averaged over 0-2, 2-7, and 7-14 days in culture after electrophoretic separation. Right panel is the total GH production in 14 days per 50,000 cells in each fraction. (From Plank et al.[8])

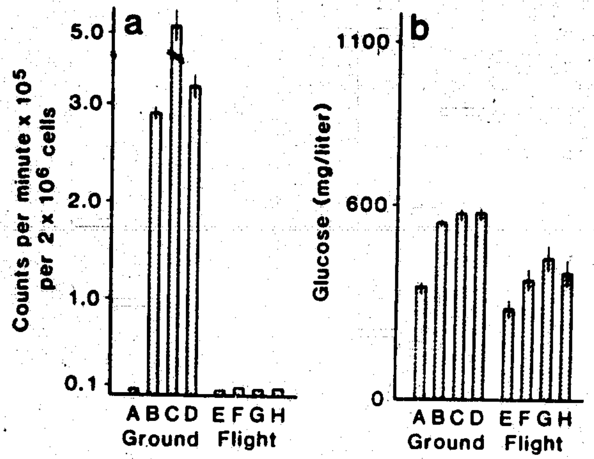

FIGURE 11.3. Lymphocyte activation induced by Con A in microgravity. Cultures of human lymphocytes were exposed to mitogenic concentrations of Con A in ground samples B, C, and D and flight samples F, G, and H, respectively. Samples A (ground) and E (flight) were unstimulated controls. (a) Activation measured after 69-h incubation at 37°C as [^{3}H]thymidine incorporation into trichloroacetic acid-precipitable material. (b) Glucose remaining in the medium measured by the glucose dehydrogenase method.[6] The initial concentration of glucose in the medium was 1100 mg/liter. The standard deviation of triplicate samples is given, except for samples A, E, F, G, and H in (a), for which it was too low to be shown here. (From Cogoli.[10])

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12. COMBUSTION STUDIES IN MICROGRAVITY

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I. BACKGROUND

During the decade of the 1950s, initial experimental studies of combustion under microgravity conditions were carried out by University of Tokyo workers who demonstrated that liquid fuel drops could burn in a spherically symmetric fashion, in accordance with simple, gravity-free combustion theory.[1,2] With the advent of NASA's requirements for knowledge of combustion processes in space environments, the 1960s saw various U.S. workers start to employ drop tower (and related) facilities to study gravitational effects on a number of combustion phenomena.[3-6]

In 1973, NASA commissioned a study concerned with examining "the physical bases and scientific merits of combustion experimentation in a space environment." The study group included: A. L. Berlad (SUNY at Stony Brook), C. Huggett (NBS), F. Kaufman (U. of Pittsburgh), G. H. Markstein (Factory Mutual Res. Corp.),

H. B. Palmer (Penn. State University), and C. H. Yang (SUNY at Stony Brook). The study was broad ranged, solicited the views of the combustion science community's many workers, and was completed with issuance of a summary report[7] in 1974. Conclusions drawn in the 1973-74 summary report are quoted below (pp. 97-99 of Ref. 7).

The review of combustion experimentation (and modeling which could benefit from the availability of a space laboratory) reveals one theme that appears to dominate all others. Extensive and systematic experimentation on the range $0 \leq g \leq 1$ is viewed as essential to the development of the understanding required in virtually all the major fundamental areas of combustion. Current theory has not been adequately guided by experiment. Current (drop tower) facilities for $0 \leq g \leq 1$ experimentation have an important role to play in future studies. But these roles are necessarily limited. Only an orbital space laboratory can provide the scales of time and space necessary to exploit substantially the scientific goals of combustion experimentation in a space environment. These conclusions apply for:

(a) Premixed Flame Propagation and Extinction Limits;
(b) Theory of Noncoherent Flame Propagation;
(c) Upper Pressure Limit Theory of Ignition and Flame Propagation;
(d) Autoignition for Large Premixed Gaseous Systems;
(e) Cool Flames in Large Premixed Gaseous Systems;
(f) Burning and Extinction of Individual Drops or Particles, Over Very Large Ranges of Pressure;
(g) Ignition and Autoignition of Clouds of Drops and/or Particles, Over Very Large Ranges of Pressure;
(h) Two Phase Combustion Phenomena Involving Large Liquid-Gas or Solid-Gas Interfaces;
(i) Radiative Ignition of Solids and Liquids;
(j) Pool Burning and Flame Propagation Over Liquids;
(k) Flame Spread and Extinction Over Solids;
(l) Smoldering and Its Transition To Flaming (or Extinction);
(m) Laminar Gas Jet Combustion;
(n) Coupling (or Decoupling) of Convectively-Induced Turbulence Involved In Various Combustion Phenomena;

(o) Transient Responses of Flames To Time-Dependent (Effective) Gravitational Fields.

This is a partial listing of the many valuable experimental and theoretical programs that can be carried out with the essential facilities provided by a Space Environment. Several of these programs show particularly outstanding promise. In this latter category must be included the areas of:

(1) Extinction Limits in Premixed and Un-premixed Gases;
(2) The Many Diverse Areas of Two Phase Combustion; Particularly the Combustion of Single Drops and Particles, Arrays of Solid Fuel Elements, and Liquid Pools.

The extent to which other noted (or uncited) combustion studies will revolutionize our understanding of the Fundamentals of Combustion depends largely on the ingenuity of experimenters and theorists who have yet to address the scientific opportunities that space-based combustion experimentation can provide.

The 1974-84 period has seen many contributions to combustion theory and experiment concerned with the centrally important problems considered in the 1973-74 study.[7] These deeper insights have served to strengthen the conclusions regarding the need for microgravity combustion experiments. The report briefly reviews the current status of major areas of combustion science where microgravity studies promise to provide otherwise unobtainable sets of needed results.

II. THE MICROGRAVITY COMBUSTION SCIENCE PROGRAM

With the completion of the 1973-74 study,[7] NASA has continued to work closely with the combustion science community -to help identify worthy researchers and proposals, as well as the general directions for NASA research efforts. The current program of research has evolved slowly and reflects an emphasis on attacking centrally important, fundamental issues in combustion science. Although the advancement of fundamental combustion science is the principal objective of the program, it is evident that the many applied needs of the combustion field continually require and exploit the inputs provided by fundamental combustion studies.

Fields of application include energy conversion and propulsion devices of many kinds, combustion safety characterization of flammable systems and structures under normal gravitational conditions.

Current research efforts include the following:

(1) F. A. Williams, Princeton University
Droplet Burning Experiment

(2) R. A. Altenkirch, University of Kentucky
Solid Surface Combustion

(3) A. L. Berlad, University of California, San Diego
Particle Cloud Combustion Experiment

(4) R. B. Edelman, Science Applications, Inc.
Gas Jet Diffusion Flames

(5) R. A. Srehlow, University of Illinois
Effect of Gravity On A Lean Limit Flame

(6) P. D. Ronney, NRC Fellow at NASA-LRC
Effect of Gravity On Near-Limit Behavior In Laminar Premixed Gas Flames

(7) P. Pagni and C. Fernandez-Pell, University of California, Berkeley
Smoldering Combustion

(8) W. Sirignano, Carnegie-Mellon University
Gravity Effects On Liquid Fuel Pool Fires: Ignition, Flame Spread and Pool Burning

(9) K. Sacksteder, NASA-LRC
Spacecraft Fire Safety

Investigations[1-3] are currently in a hardware development stage, preparatory to experimentation aboard the Space Shuttle.

A NASA/USRA Combustion Discipline Working Group (DWG) meets regularly to examine ongoing research efforts, to identify opportunities for needed new investigations, and to interact with both the NASA leadership and the combustion science community. The current membership of the Combustion DWG includes:

W. Bartok	Exxon Research and Engineering Co.
A. L Berlad (Chairman)	UCSD
T. Hanratty	University of Illinois
J. Hoffman	NASA-JSC
R. Levine	NBS
P. Meyers	University of Wisconsin

H. Palmer	Penn. State University
S. S. Penner	UCSD
B . Peters	General Motors Research Labs
K. Sacksteder (Vice Chairman)	NASA-LRC
R. Strehlow	University of Illinois
F. A. Williams	Princeton University

The NASA-sponsored microgravity combustion science program emphasizes continuing review of ongoing research, strong ground-based preparation of experiments for spaceflight, and active participation of the Principal Investigator in all aspects of of experiment.

III. WHY MICROGRAVITY COMBUSTION EXPERIMENTS?

Reduced gravity combustion experiments may prove useful owing to the facts that under microgravity conditions, natural convective processes are suppressed, gravitational settling in two-phase combustibles is suppressed, and spatial homogeneity of quiescent, nonuniform density combustibles may be achieved. Where turbulence-inducing mixing processes are employed (at $g = 1$) to achieve spatial homogeneity of nonuniform density combustible mixtures, microgravity conditions permit the maintenance of spatial uniformity while turbulence decays. Some important combustion experiments now carried out at normal gravitational conditions may be comprised in essential ways. In some cases, these compromising features can be eliminated by the use of microgravity experimentation. In other cases, $g = 1$ combustion experiments involve natural convection-coupled effects which are not adequately understood. In such cases, microgravity experimentation may be employed to help resolve these effects. In some other cases, needed combustion experiments are simply <u>not achievable</u> at $g = 1$, but <u>are</u> feasible at reduced gravitational conditions. In all instances, however, the use of microgravity combustion experimentation represents a powerful tool for the solution of centrally important issues where $g = 1$ experimental/theoretical attempts have been (and promise to be) inadequate. For more than a decade, NASA-sponsored studies[4-21] have followed the pioneering work of Kumagai and Isoda in systematically exploring the effects of gravity on combustion. In the following several cases, we examine the special significance and value of microgravity experimentation for each characteristic case. In general, it is found that the information to be derived has not been obtainable by other means.

Premixed gaseous flame propagation and extinctions

Previous studies at normal gravitational conditions have shown[7] that upward and downward premixed gaseous flame propagation are substantially different and that natural convection dominates upward flame propagation mechanisms in the neighborhood of the flammability limits. It is recognized that flammability limit theory is not fully developed and that premixed flame propagation and extinction theory can be placed on a more firm foundation through experimental studies which suppress natural convective effects. The resolution of these flame theoretical issues is important to an understanding of two-phase (pre-mixed) combustible systems as well as to single phase premixed systems.

Premixed particulate cloud flame propagation and extinction

It has been found that premixed, uniform, quiescent clouds of combustible particulates (in a gaseous oxidizer) are difficult or impossible to achieve in earth-based laboratories. This is particularly true for particulates of substantial size.[22] Development and testing of flame propagation and extinction theory for such systems requires first that reliable experiments be achieved for quiescent, premixed, homogeneous systems. The classic two-phase flammability studies developed at the U.S. Bureau of Mines[23] and elsewhere have provided important bases for comparing the relative flammabilities of various combustibles. Unfortunately, it has not been found possible to establish the quiescent, uniform clouds of particulates which are required to adequately characterize the preignition states of these two-phase combustible systems. These difficulties (at $g = 1$) derive directly from particle settling effects and/or from mixing-induced turbulence effects. It has been argued[22] that these $g = 1$ difficulties can be alleviated through microgravity experimentation. Once two-phase premixed flame propagation and extinction data are obtained (at reduced gravity) theoretical resolution of the difference between premixed single-phase and premixed two-phase flame propagation and extinction theory becomes more promising. Currently planned studies at reduced gravitational conditions include those for lycopodium-air, cellulose-air, and coal-air particulate clouds.

Smoldering

One of the most interesting combustion phenomena is smoldering. This phenomenon appears to sustain a number of characteristic processes, at normal gravitational conditions: steady and unsteady smoldering, extinction, and the transition to flaming combustion. There is the complex interaction of two-phase pyrolysis and oxidative kinetics.[24] Radiative, molecular, forced, and free-convective transport processes interact in poorly understood ways. All characteristic rates, chemical and transport, are slow compared with those which typify flaming combustion. Both experimentally and theoretically, it has been difficult to describe the roles of each of the interacting processes. No comprehensive theory is available which prescribes both the quasi-steady smoldering process and the critical conditions for transition to extinction or flaming. Can smoldering exist in the absence of natural and/or forced convection{ There is a need to address this and related central issues associated with this important combustion phenomenon. Reduced gravity experimentation is expected to help address this need.

Combustion of individual drops and particles

The classic work of Kumagai and Isoda[1] has served to stimulate subsequent studies concerned with the combustion of individual drops and particles. In the absence of a gravitational field, spherically symmetric burning has been dramatically demonstrated.[1] But questions remain which require further experimentation at reduced gravitational conditions. One may note the significant areas of the general unsteadiness of single drop combustion, nonsphericities in ignition and combustion, the interactions of nonspherically symmetric drop vaporization-combustion and drop motion, flame extinction, and explosive extinction of a burning single drop.

Radiative ignition of condensed phase fuels

Current theory and experiment show that (at $g = 1$) very large radiative flux rates imposed on very large planar-surfaced liquids or solids lead to short ignition delays which do not depend substantially on natural convective processes.[25,26] However, for low radiative flux rates and for small fuel sample sizes, natural convective processes can be important. In the limiting case where radiative flux rates are inadequate to achieve ignition, it is found that free convective losses to a cold atmosphere are centrally important.[25] Accordingly, there is a complex interplay of radiative, convective, and molecular transport (with vaporization, pyrolysis, and oxidation kinetics) which prescribes limiting radiative-induced ignitability, at normal gravitational conditions. Understanding of these complex, limiting ignitability conditions could be greatly enhanced through research which would permit the suppression and/or control of natural convective effects. Reduced gravity combustion experimentation promises to provide otherwise unobtainable insight to this important class of problems.

Large surface combustion and extinction

Two-phase combustion processes involving a gaseous oxidizer and large condensed phase combustible materials have been studied extensively under normal gravitational conditions. Complex flame propagation and extinction phenomena are observed. Gravitational collapse of partially burned solid fuel arrays sometimes tends to compromise the investigator-selected conditions for the unburned medium. Gaseous natural convective processes are generally so prominent as to obscure the roles of chemical kinetics and of other transport processes. Natural convective processes frequently determine the location of effective control surfaces at which preselected gaseous oxidizer conditions are best known. In the propagative burning of liquid pools (and of melting solids) surface-tension-induced convection processes[27] are thought to play an important role. To be able to modify, suppress, or control one or more of these effects through microgravity combustion research seems attractive. More tractable theory can then be applied to liquid pool burning phenomena which promise to be less complex.

Boundary-layer combustion

Boundary-layer combustion may be supported by flow of an oxidizing gas over a flat plate which serves to supply (by fuel vaporization or by fuel injection) gaseous fuel for the combustion process. At normal gravitational conditions operating on flow over a flat horizontal plate, current analysis shows[10] that the cross-stream buoyancy-induced body force acts to effectively produce a streamwise pressure gradient in the fluid adjacent to the plate surface. For an upwards-facing plate, a favorable pressure gradient is derived which tends to accelerate the flow, relative to the corresponding gravity-free case. Flow deceleration is prescribed for a downwards-facing plate. These gravitational effects induce changes in boundary-layer structure and flame standoff distances. It is expected that boundary-layer combustion under microgravity conditions may serve to verify these current theoretical concepts, as well as to resolve observed inconsistencies between experiments conducted at g = 1 and theory which neglects gravitational effects.

Nonflowing premixed combustible media

A broad class of important combustion phenomena are observed, at normal gravitational conditions, in nonflowing, premixed systems. This class includes gaseous and two-phase autoignition and explosion processes, kinetic oscillations associated with the low temperature oxidation of carbon monoxide, and thermo-kinetic oscillations associated with the oxidation of hydrocarbons.[11,16] For all of these phenomena, apparatus walls (and associated boundary conditions) play an important role in heat and mass transport. Heat and mass transport between wall and the gaseous medium, as well as the heterogeneous chemical kinetic processes at the walls have been found[11,16,28] to strongly influence both qualitative and quantitative characteristics of these phenomena. Moreover, there exists a corresponding set of autoignition (and other?) phenomena for premixed clouds of particulates. These two-phase premixed combustion processes cannot be studied in a similar way at g = 1 owing to the gravitational settling of particulates and/or the mixing-induced turbulence required to achieve reasonably uniform clouds of particulates. These problems are particularly severe for the case of large particles (e.g., of the order of 50 μm, or larger), mixed particle sizes, and mixed particular materials. Combustion experimentation at microgravity conditions may be used to deal with these two-phase problems.[22] For selected members of this class of problems, a number of single and two-phase combustion phenomena are discussed in the following paragraphs. The role of microgravity research in resolving essential theoretical and experimental issues is emphasized.

IV. COMBUSTION PHENOMENA IN NONFLOWING PREMIXED SYSTEMS

The systems to be discussed here are generally considered to involve the least complex combustion theory and some of the cleanest and easiest to define experimental conditions. An understanding of these phenomena is fundamental to an understanding of more complex combustible systems involving forced convective flows. Although g = 1 experimental conditions may be of primary interest, current theoretical representations are generally fashioned for g = 0 conditions, for which experiments have not yet been performed. This difficulty is compounded by the difficulty or inappropriateness of actually performing suitable g = 1 experiments.

Autoignition phenomena in premixed gaseous media

The constituative equations for thermokinetic processes in premixed gaseous media generally include[16] an energy equation, a set of chemical kinetic rate equations, mass or species conservation equations, and an equation of state. For a gravity-free field, these are of the form,

$$\rho c_v \frac{\partial T}{\partial t} = \nabla \cdot (\lambda \nabla T + I) + \sum_{j=1}^{r} R_j \Delta_j, \qquad (1)$$

$$\xi_{j1} + \xi_{j2} + \xi_{j3} + \xrightarrow{k_j} \xi_{j4} + \xi_{j5} + \xi_{j6} + \Delta_j \qquad (2)$$

$$\text{for } j = 1, 2, \ldots, r$$

$$\frac{\partial c_i}{\partial t} = \nabla\cdot(D_{i,m}\nabla c_i) + \sum_{j=1}^{r} \dot{c}_{ij}''' \quad (3)$$

for $i = 1, 2, \ldots, n$

$$pv = \sum_i c_i RT \quad (4)$$

where c_i is the volumetric mole concentration of the ith chemical species and where i = 1,2, ...,n; c_{ij}''' is the volumetric molar rate of production of the i^{th} chemical species by the jth kinetic process, and where j = 1,2, ...,r; c_v is the specific heat at constant volume; D is a diffusion coefficient; I is a photochemically insignificant (thermal) radiative flux density; k_j is the rate constant for the jth kinetic process; R is the universal gas constant; R_j is a molar reaction rate corresponding to the species and energy release Δ_j; T is the absolute temperature; Δ_j is the molar heat of reaction for the jth irreversible kinetic process; λ is a local thermal conductivity; ξ_{jq} are the concentrations associated with the q = 1, 2, . . .,6 species that participate in the jth kinetic process. Equations (1)-(4) ignore any transfer of heat and mass that may derive from natural convective effects, yet the autoignition phenomenon itself is recognized to be dependent on experimental apparatus size as well as heat and mass transfer between the bulk of the gaseous system and walls.[16] Equations (1)-(4) are multidimensional in nature and are typically applied to experiments performed in spherical or long cylindrical apparatus. Although it has been shown[29,30] that natural convective processes generally occur during autoignitions experiments, the constituative equations (1)-(4) have not been generalized and applied to autoignition observations at g = 1. Moreover, the roles that walls may play for both heat and species conservation appear to be obscured by natural convective processes. This is particularly true where heterogeneous chemical kinetics (wall chemistry) may play a significant role in autoignition.[31,32] In all of these previous studies, application of Equations (1)-(4) in their multidimensional form has not been made. In fact, reliable specification of the specific reactivity to be assigned to the various radial boundary conditions has not been achieved. This is due, in part, to the usual truncation of Equations (1)-(4) into a set of one-dimensionalized equations. But even if Equations (1)-(4) were used in their most general form (as is now possible), they are appropriate only in a gravity-free field.

The variety of heterogeneous chemical kinetic effects (on autoignition) that awaits convection-free investigation is enormous. The specific reactivities of chemically reactive walls are not deducible, where radial transport properties are unknown or ill-defined (natural convective effects). The very nature of this important combustion process is lost in the ad hoc representations currently employed to deal with the specific reactivity of a wall effect. Typically, kinetically significant radial boundary conditions are not imposed (by current workers) on Equations (1)-(4). Rather, heterogeneous wall chemistry is couched in homogeneous chemical kinetic schematics of the classical form.[30-32]

$$A \xrightarrow{k_{A,W}} \text{Wall} \tag{5}$$

$$B \xrightarrow{k_{B,W}} \text{Wall} \tag{6}$$

here A and B are taken to be chemically important species. It can be shown that one-dimensionalization of Equations (1)-(4) with the use of truncated analytical forms such as Equations (5) and (6) still requires detailed knowledge of radial transport properties. For noncatalytic walls acting as perfect species sinks, and in the absence of free convection, $k_{A,W}$ can be approximated:

$$k_{A,W} \doteq \frac{12D}{d^2} \tag{7}$$

Clearly, natural convective effects at $g = 1$ make Equation (7) inappropriate. Significant heterogeneous kinetic processes also serve to make Equation (7) inappropriate.

The current experimental and theoretical situation with regard to autoignition phenomena may be summarized as follows: Extensive experimental data obtained at $g = 1$ demonstrate the important roles that natural convective transport of heat and mass can play in the determination of the autoignition phenomenon. The specific reactivity of walls is not adequately described as a result of ill-defined free-convective effects and the limiting features of truncated representations of multi-dimensional boundary-value problem. More sophisticated computational approaches, coupled with reduced gravity experimentation promises to provide <u>both</u> better understanding of autoignition <u>and</u> deduction of the specific reactivities of kinetically active walls. The complexity of experimental and analytical analysis is best illustrated for a simple, idealized general case of plane-parallel plates of infinite extent, containing a stationary state process with chain branching and breaking as phase reactions (of only one significant species) subject to heterogeneous wall effects. Equation (3) can be written in the quasi-steady form

$$\frac{d}{dx} D_{i,m} \frac{dc_i}{dx} + (f_i - g_i)c_i + c_{0,i} = 0 \qquad (8)$$

subject to the boundary conditions

$$\frac{dc_i}{dx} = 0 \quad \text{at } x = 0 \text{ and} \qquad (9)$$

$$c_i = c_{i,w} \quad \text{at } x = \pm d/2$$

where f_i is a chain-branching coefficient, g_i is a chain-breaking coefficient, and $c_{0,i}$ is the rate of production (of species c_i) due to straight chain processes. In general, the diffusion equation is nonlinear and the quasi-steady value of $c_{i,w}$ unknown. Equations (8) and (9) describe the quasi-steady state of the system at subcritical conditions. Utilization of phase plane and other methods to identify the autoignition condition has been described elsewhere.[16] Reduced gravity experimentation coupled with detailed space-resolved diagnostics may serve to identify $c_i(x)$, and derivatively, $c_{i,w}$ and the specific heterogeneous reactivity of the wall surface. In practice, a long cylindrical tube may serve the reduced gravity experimenter well. At g = 1, the experiment is subject to free convective effects.

Autoignition phenomena in premixed clouds of particulates

For purely gaseous media, earthbound experimental procedures to create uniform quiescent mixtures of fuel and oxidizer are trivially routine. For clouds of particulates, establishment of earthbound uniform clouds of quiescent fuel particulates (in a gaseous oxidizer) has not been possible. This is particularly the case for large (practical) size particulates. For particulates whose gravitational settling velocities are significant, vigorous mixing techniques[22] are required to achieve uniform particulate concentrations. Under highly stirred conditions, transport properties of the medium are ill-defined. The decay of mixing-induced turbulence is slow[22] and (at g = 1) particulate settling processes proceed as mixing-induced turbulence decays slowly. Extensively documented studies of autoignition in two-phase systems (at g = 1) have served to provide important qualitative insights into the relative combustibility of a broad variety of particulates.[33,34] Unfortunately, the g = 1 data on two-phase autoignition[33,34] do not lend themselves to analysis. In general, experimental conditions are poorly defined. Uniformity of particulates is questionable, particle-gas velocity defects are operative, and the transport properties are transient and unknown. In view of the fundamental and practical importance of two-phase combustible systems, the study of the autoignition of uniform, quiescent clouds of particulates appears necessary. Current theory for two-phase autoignition[21] has not been experimentally tested. Normal gravitational conditions present great difficulties, and microgravity experimental investigations have not yet been performed.

Kinetic oscillations in carbon monoxide-oxygen combustion

There exist extensive experimental studies of the low-temperature oscillatory oxidation of carbon monoxide.[28,29] These studies were all performed at normal gravitational conditions. Although the combustion process is slow, natural convective transport of heat and mass is an ever present concern. Current theory[11] emphasizes the importance of chemically reactive wall mechanisms. Equations such as (5) are used as essential descriptors for wall chemistry involving [H], [O], [OH], [HO_2], and [H_2O_2] species. The section above on autoignition phenomena in premixed gaseous media indicates the inadequacies of equations of this form when applied to earthbound observations of combustion phenomena in closed vessels involving chemically reactive walls. The characteristic period of this oscillation phenomenon is so long (of the order of several seconds) that $g = 0$ experimentation in long cylindrical vessels would permit spectroscopic space and time resolution of chemical species concentrations in a two-dimensional reactor. Accordingly, convection-free combustion dynamics may thus be observed and analyzed. Truncation of radial boundary condition information would then not be necessary. Specific surface reactivities may then be deduced. No such microgravity experiments have yet been performed.

<u>Thermokinetic oscillations in hydrocarbon-oxygen combustion</u>

The pheomenological features which prescribe oscillations in the low temperature combustions of hydrocarbons are thought to be very different from those associated with carbon monoxide oxidation. In the neighborhood of 600 K, a small number of hydrocarbon oxidation cycles (typically less than ten) are observed in closed vessels, with temperature rises per pulse of some 5 K to 200 K.[35] Oscillatory hydrocarbon oxidation is thought to be thermokinetic in nature with the oscillatory process dependent on the heat transfer rates between the reactive medium and the reactor walls, and the interaction of these heat losses with "negative temperature regime" chemistry.[32] Natural convection dominates these heat transfer rates, in the absence of other, experimenter-imposed effects. Inasmuch as natural convection for this transient effect is not well understood, attempts to employ <u>controlled</u> heat transfer rates through vigorous stirring have been employed.[35] Without a knowledge of these unsteady heat losses, analysis of this class of phenomena is not possible.[36] Yet, at normal gravitational conditions, the currently employed heat transfer correlation of the form[35]

$$\dot{q} = \ell(S/V)(\bar{T} - T_w) \qquad (10)$$

must be considered unreliable. In Equation (10) ℓ is an estimated effective heat transfer coefficient associated with the unsteady process. S/V is the surface to volume ratio, and $(\bar{T} - T_w)$ is an estimated spatially averaged characteristic temperature rise.

If this experiment were done under reduced gravity conditions, without stirring and without natural convection, there would be no necessity for recourse to questionable heat transfer correlations. The complete set of constituative equations (1)-(4), may then be applied. No such experiments have been performed under reduced gravity conditions.

V. CONCLUDING REMARKS

This brief review has noted a number of central issues in combustion science which await resolution through microgravity combustion research. This list of crucial problem areas which can profit importantly is not exhaustive. The field of two-phase combustion promises invaluable results through microgravity research. Microgravity combustion studies promise to eliminate problems which derive from gravitational settling and ill-defined mixing processes, thereby permitting well-characterized experiments and tractable analytic problems. Where $g = 0$ theory and experiment can be brought to reliable correspondence, they can both be used as essential foundations for understanding more complex systems which derive for $g > 0$.

It is hoped that the scientific community will vigorously exploit the experimental and analytical opportunities afforded it through microgravity combustion research.

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13. FLAME SPREADING

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The spread of a flame in the gas over the surface of a solid combustible involves, in an essential way, the transfer of heat from the flame to the solid fuel immediately ahead of it.[1] This forward heat transfer brings the unburnt fuel to a condition in which it is capable of igniting, thus allowing the spreading to be sustained at a rate that is, in general, proportional to the forward transfer rate. Previous investigations aimed at elucidating the physics of the spread process have, by and large, been aimed at determining how this heat transfer occurs[2] and how the environment affects the heat transfer process.[3,4]

For flames that spread in the Earth's gravitational environment, the flame induces a buoyant flow. In upward propagating flames, the spread process is rapid and acceleratory.[5] The induced flow is in the direction of spread, and the hot combustion products form a plume that bathes the unburnt fuel, heating it by conduction and, if the optical depth of the flame is large enough, by radiation as well. In downward flame spread, buoyancy induces a flow of ambient fluid that opposes the spreading flame. The result is that the forward heat transfer occurs either by heating conduction through the gas or solid (or both).[3,4] Radiation heat transfer is usually unimportant because the flame, due to the buoyant flow, lies close to the fuel surface resulting in a small view factor. And the convective motion cools the unburnt fuel rather than heating it.

Horizontally spreading flames possess some of the same characteristics as both downward and upward spreading flames do. Topside flames must spread into an opposing flow of oxidizer that is induced by the flame plume as in downward spread, but unlike downward spread, the flame plume extends above the pyrolyzing surface of the fuel to provide a favorable view factor for forward radiation heat transfer.[6,7] Underside flames induce a flow in the direction of spread. They tend to propagate unsteadily and race ahead of the topside flame.[8,9]

Photographs of flames spreading over thin cellulose acetate sheets at nearly zero gravity show that these low-gravity flames are similar in appearance to downward spreading flames at normal gravity.[10,11] They do not exhibit a plume that precedes the leading edge of the flame as in upward spread, and the flames on both sides of the fuel bed look the same, as they do in downward spread, in contrast to horizontal spread. It is likely then that the physics of flame spreading in low gravity is more akin to that of downward spread in normal gravity rather than either upward or horizontal spread. Further support of this speculation is given by the fact that low-gravity spread rates are just less than downward spread rates at normal gravity which in turn are lower than either horizontal or upward spread rates, upward ones being the highest.[5,10]

Gas-phase conduction is the dominant mode of forward heat transfer in downward flame spread for fuels that are heated across their entire thickness (thermally thin) downstream of the leading edge of the flame and also have a large ratio of the gas-phase conduction length (gas thermal diffusivity divided by opposing flow velocity) to the solid-phase forward conduction length (solid thermal diffusivity divided by flame spread rate). In this case, an increase in the strength of the buoyant flow can only cause the spread rate to fall. The reason for this is that the residence time of the fuel vapors near the leading edge of the flame is reduced relative to the time that the exothermic reactions require for completion so that the flame temperature drops, reducing the forward heat transfer and hence the spread rate. This phenomenon can be expressed in terms of the Damköhler number, a ratio of the residence time to the reaction time. For large Damköhler numbers, the spread rate is unaffected by changes in the Damköhler number because sufficient reaction time is provided for the production of the highest temperature attainable. For small Damköhler numbers, the spread rate decreases as the Damköhler number decreases.[3]

The dominant heat transfer mode depends on the Damköhler number for fuels that are thick enough so that they may or may not be heated across their entire thickness.[4] At large Damköhler number, the heated layer thickness in the fuel is small compared to the fuel's thickness (thermally thick), and the ratio of the gas-phase to solid-phase conduction length exceeds unity; heat transfer occurs then by gas-phase conduction. Increasing the strength of the opposing flow does not change the flame temperature or the heat conducted upstream in the gas because the conduction length scale in the gas is the same parallel and normal to the fuel surface, but it decreases the heated layer depth in the solid. The reduced in-depth heating results in more of the forward heat transfer being available for vaporization with a concomitant increase in spread rate.[4] At small Damköhler number, as the flame nears extinction, nearly the entire fuel thickness is heated, and forward heat transfer is by solid-phase conduction.[4,12] Increasing the buoyant flow here decreases the flame temperature causing a reduction in the forward heat transfer and hence a reduction in the spread rate.

As is evident from the above discussion, the direction and strength of gas-phase flow are of signal importance in establishing the spread rate. For downward spread, the character of the flow field is understood, and that understanding of the physics of the process as described above is demonstrated by the success with which dimensionless downward flame spread rates, $\bar{V}_F$ where

$$\bar{V}_F = \frac{\rho_s C_s V_F \tau (T_v - T_\infty)}{\sqrt{2}\ k\ (T_f - T_v)} \quad (1)$$

are correlated against a Damköhler number[3,4,13], $\bar{B}$ where

$$\bar{B} = (B\ k\ m_{ox,\infty}/M_{ox} C_p v_c^2)(\bar{T}_f^2/\bar{T}_a)^3 \exp[-(\bar{T}_a/\bar{T}_f)] \quad (2)$$

The dimensionless spread rate $\bar{V}_F$ is a ratio of the actual heat transferred forward of the flame to the maximum that could be transferred if $\bar{B}$ were large and the flame were at its maximum temperature.[3] In the above, ρ_s is the solid-phase density, C_s is the solid-phase specific heat, $\bar{V}_F$ is the spread rate, k is the gas-phase thermal conductivity, T_v is the vaporization temperature, T_∞ is the ambient temperature, T_f is the flame temperature for adiabatic, stoichiometric combustion, τ is the heated layer depth in the solid, $\bar{B}$ is the pre-exponential factor for an assumed second order reaction, $m_{ox,\infty}$ is the ambient oxygen mass fraction, M_{ox} is the molecular weight of oxygen, C_p is the gas-phase specific heat at constant pressure, v_c is the characteristic, gas-phase velocity, and $\bar{T}_f$ and $\bar{T}_a$ are the flame and activation temperature, respectively, measured in the units of $\Delta H_c m_{ox,\infty}/iC_p$, where ΔH_c is the heat of combustion of gas-phase fuel, and i is the mass of oxygen needed to oxidize a unit mass of fuel. For thermally thin fuels, τ is the fuel bed half-thickness, while for thick fuels, τ depends on v_c but is independent of the fuel bed thickness.[4]

The expression for $\bar{V}_F$ above derives from a solution to the constitutive equations that govern flame spread into an opposing flow of oxidizer of uniform speed v_c when $\bar{B} \to \infty$ (Ref. 14) such that $\bar{V}_F$ is unity. The parameter $\bar{B}$, with which $\bar{V}_F$ correlates for finite $\bar{B}$, obtains, in part, from dimensional analysis.[3] Examples of the manner in which $\bar{V}_F$ correlates with $\bar{B}$ are shown in Figure 13.1 for downward spread over thin paper samples for which τ is the fuel bed half-thickness,* and thick samples of

*Excessive heat loss from sample edges causes some of the paper-sample data not to correlate with $\bar{B}$ alone.

polymethylmethacrylate (PMMA) where τ has been set equal to $[\sqrt{2}k_s(T_v - T_\infty)]/[\rho C_p v_c(T_f - T_v)]$ times a correlation factor to account for surface regression[12] with k_s the solid-phase thermal conductivity, and ρ the gas-phase density.[14] The characteristic, gas-phase velocity for downward spread that was used in Figure 13.1 is v_b, the buoyant velocity $(\nu g \Delta H_c m_{ox,\infty}/T_\infty i C_p)^{1/3}$, where ν is the kinematic viscosity, and g is the acceleration of gravity. This form for v_c reflects the fact that the important flow velocity is that near the leading edge of the flame that is induced by the diffusion flame above the pyrolyzing surface.[15,16]

Results such as those presented in Figure 13.1, which derive in part from dimensional analysis, represent the most powerful tools that we have for estimating flame spread rates because there are no comprehensive theories that allow the induced flow and flame spread rate for downward spread to be predicted, except perhaps the attempts by Fernandez-Pello and Williams[15] and Vedha-Nayagam and Altenkirch.[17] In most theoretical descriptions of flame spreading, the flame spread rate is determined for a prescribed flow (see, e.g., Refs. 14, 18, 19).

There is some limiting gravitational acceleration below which correlations like those of Figure 13.1 are invalid because of the use of v_b for v_c. Indeed, as $g \to 0$, both $\bar{V}_F$, for thick fuels, and $\bar{B}$ in Figure 13.1 approach infinity. Dimensional analysis of the equations and associated boundary conditions that are applicable to flame spreading[3,14,15] show us that this behavior is understandable.

Gas-phase motion for a coordinate system attached to the flame is described by the unsteady continuity, energy, and species equations along with unsteady momentum balances parallel (x direction) and normal (y direction) to the fuel bed surface. These equations, when placed in dimensionless form by measuring the variables in the units of arbitrary reference quantities[20,21] contain three parameters of importance. One is ν (or α) divided by $t_c v_c^2$, where α is the thermal diffusivity, and t_c is the reference or characteristic experimental time. The parameter appears in the unsteady terms, and its value is invariable small such that steady state equations apply in the gas. The second parameter is a Damköhler number, $\bar{B}$, without the cubic or exponential terms, which can be developed by physical reasoning[3] that multiplies the Arrhenius reaction term in the energy and species equations. The value of the remaining parameter, which appears in the momentum equations and is $\nu g_i \Delta H_c m_{ox,\infty}/T_\infty i C_p v_c^3$, determines whether or not buoyancy influences the spread process. Here g_i is the component of gravity in the direction in which the momentum balance is made. For downward spread, $g_x = g$ and $g_y = 0$ so that the total acceleration vector points in the direction of spread.

The above analysis requires that the distances over which heat can be transferred near the leading edge of the flame are the same in the x and y directions. Thus, forward conduction in the gas is assumed to be important with respect to determining the spread rate. This assumption is in keeping with the observation about the similarity of downward spreading flames and flames spreading at nearly zero gravity.

If buoyancy is to affect flame spreading, as it does for downward spread under the Earth's normal gravitation acceleration, g_e, the buoyancy term in the dimensionless momentum equation should be of order unity, which requires that $\nu g_i \Delta H_c m_{ox,\infty}/T_\infty i C_p v_c^3$ be of order unity so that the characteristic velocity is approximately the buoyant velocity $v_b = (\nu g \Delta H_c m_{ox,\infty}/T_\infty i C_p)^{1/3}$. As the acceleration of gravity is reduced, buoyancy will eventually not be the dominant force producing fluid motion, v_c can no longer be identified as v_b, and the correlating parameters of Figure 13.1 that contain v_b are no longer valid. Under these circumstances, the gas-phase equations alone are insufficient for determining v_c, and the physics of the coupling between the solid and gas phases must be investigated. Boundary conditions at the solid-gas interface are provided by energy and species balances along with the no slip condition that the streamwise gas speed and the spread rate are the same. In addition, the velocity normal to the fuel bed surface in the pyrolysis region is determined by the pyrolysis rate law for the solid. Because this normal velocity exists over a limited region of the fuel bed surface, the flow surrounding it is similar to a jet flow wherein fluid from the surrounding atmosphere may be entrained.[22] Consequently, the normal velocity at the fuel bed surface determines the characteristic velocity for the gas.

With v_c estimated from surface pyrolysis as v_s, a large value of the parameter $(v_b/v_s)^3$, which is the cube of the ratio of the induced, streamwise velocity to the transverse velocity, implies that buoyancy is the dominant force producing the flow, and the characteristic velocity should be identified as v_b. For small values of the parameter, buoyancy is unimportant, and its effects can be neglected in any attempt to analyze flame spreading under these conditions.

In the table below, typical values for $(v_b/v_s)^3$ are given for flame spread over paper samples in oxygen presuming a zeroth-order, surface pyrolysis rate law with a pre-exponential factor of 7.4 x 10^8 kg/m^2·s and an activation energy of 126 kJ/mol for calculating v_s.[3,23,24] Other properties were taken from Altenkirch et al.[3]

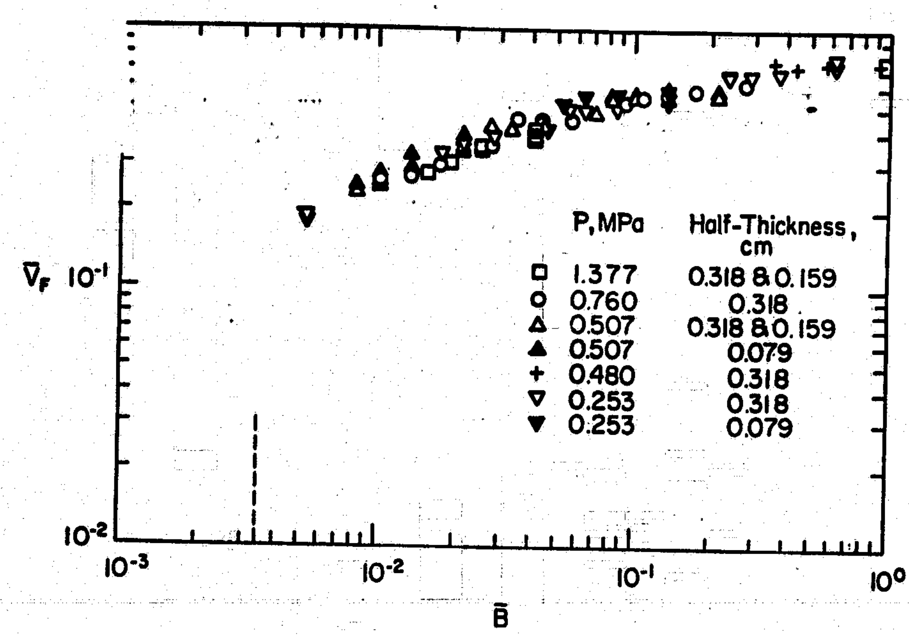
Type of environment	g/g_e	P,atm	$(v_b/v_s)^3$
Earth	1	1	1400
Earth	1	0.1	14
Space[25]	10^{-5}-10^{-4}	1	0.014-0.14
Space[25]	10^{-5}-10^{-4}	10	1.4-14

The above table shows, as we expect, that buoyancy is the main driving force generating the gas-phase flow for flame spreading under the influence of the Earth's gravity but that it can be neglected under gravitational conditions that might be encountered in the low-gravity environment of space travel although environmental conditions may be set aboard spacecraft where buoyancy is influential.

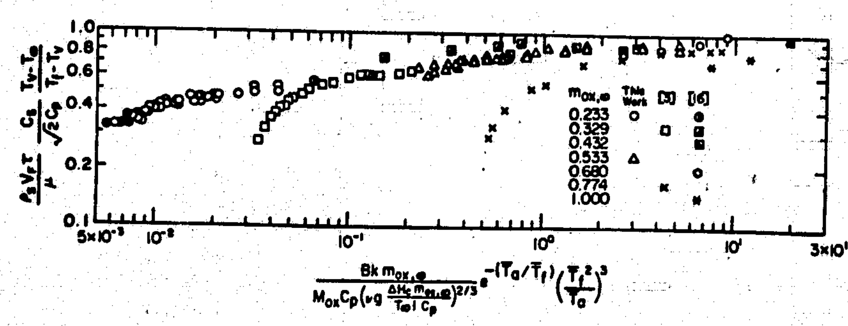
As mentioned before, $\nu/t_c v_c^2$ is invariably small so that the gas phase may be treated as though it were in steady state. For example, for conditions of the above table the smallest value of v_c would be about 3 cm/s when it is determined by surface pyrolysis, requiring that t_c >> 0.06 s, say at least 0.5 s, to establish the steady state. Whether or not the flame spreads at a steady rate though depends on the magnitude of $\nu/t_c V_F v_c$. This parameter can be obtained by placing the solid-phase energy equation in dimensionless form assuming that the length over which heat can be conducted forward through the gas applies to the solid,[24] which is necessarily the case for thermally thin fuels. Because V_F can be an order of magnitude or so less than v_c, we can expect that under certain conditions the experimental time needed to achieve a steady spread rate could be about 10 times that needed to establish a steady state in the gas. Thus it is not surprising that for some experimental conditions employed in reduced gravity, drop tower experiments[10] of unsteady spreading were observed in the 5 s of available experimental time.

Understanding of the phenomena involved in flame spreading at reduced gravity is important with respects to the fire safety aspects of space travel. To this end then, effort must be expended to understand the mechanism of flame spreading in the absence of any buoyancy induced or externally imposed gas-phase flow. Theoretical and experimental work must proceed in parallel, and the mid-deck of the Space Shuttle has been identified by NASA as an appropriate location for flame spread experimentation. Ground-based experimental facilities do not provide sufficient time (drop towers) or sufficiently low and/or controllable accelerations (airplanes flying Keplerian trajectories) to allow careful experimentation to be conducted.

A mid-deck, flame-spread experiment would consist of measuring the flame-spread rate over a solid fuel sample from motion pictures of the spread process within a sealed chamber. The film record would allow flame shape, which gives an indication of the character of the gas-phase flow, and flame color, useful in gaining insight into flame chemistry, to be determined. Gas- and solid-phase temperatures, measured with fine-wire thermocouples, would provide information about flame temperature and heat flux through analysis of the heat conduction processes in the solid, from the gas to the solid, obviously, as discussed above, an important quantity in establishing the spread rate.



Dimensionless Spread Rates for Downward Spread over PMMA Samples [4].



Dimensionless Spread Rates for Downward Spread over Paper Samples [3].

FIGURE 13.1. (top) Dimensionless spread rates for downward spread over PMMA samples.[4] (bottom) Dimensionless spread rates for downward spread over paper samples.[3]

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14. FLUID DYNAMICS IN MICROGRAVITY

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I. INTRODUCTION

One primary objective of NASA's Microgravity Fluid Dynamics Program is to develop a scientific understanding of the role of gravity in both fundamental fluid physics and fluid and transport processes related to topics in materials science, combustion, and biotechnology. By turning to the environment of space, we can begin to explore new phenomena previously unseen because of the suppressing effects of gravity. Once free of these constraints, the insights we gain will begin a new chapter in our understanding of natural processes. This is the promise of microgravity research.

In our terrestrial environment, thermophysical processes are influenced by buoyancy forces which arise owing to the presence of a gravitational field. In many physical processes, the magnitude of this buoyant force is sufficiently great as to overwhelm other forces which arise owing to pressure, inertia, surface tension, and viscous shear. As the gravitational field weakens, however, these other forces begin to grow in importance and start to play complex and often times competing roles. The study of systems in which these other forces contribute significantly to the transport process has been pursued with only limited interest because of the difficulty of conducting research.

Gravity manifests itself in many ways in fluid systems. First, the magnitude of the gravitational acceleration uniquely determines the liquid-liquid or liquid-gas interface shape. As the local acceleration field is reduced, interfacial surface tension forces, as well as forces which arise owing to contact angle, container geometry, and relative density, begin to define the shape and motion of the interface. These effects are not just limited to static situations but include the dynamics of interfaces as well. Second, gravity determines the extent of fluid stratification due to density gradients which might exist within the system. Finally, the mechanisms of heat transfer are altered when gravity is removed. Under conditions of microgravity, natural convection is virtually eliminated, and conduction and radiation become the premier modes of heat transfer.

In essence, the Fluid Dynamics Program has two equally important objectives. The first objective is to study fundamental transport phenomena under conditions of microgravity taking full advantage of the unique conditions to obtain measurements which would be impossible in a terrestrial environment. Examples of these include the measurement of transport coefficient near critical conditions. The second objective is to conduct applied microgravity fluid dynamics experiments in support of researchers working in such areas as electronic materials, ceramics, metallurgy, combustion and biotechnology. The proper planning and interpretation of the above experiments need a highly sophisticated understanding of transport phenomena which does not always form part of the repertoire of researchers in applied disciplines. Interaction between investigators in applied fields with investigators in the fluids and transport sciences will play a key role in ensuring the success of these related efforts.

The aim of the NASA effort then is to support a structured and systematic program to begin the detailed investigation of microgravity fluid dynamics. NASA is uniquely positioned to succeed in this regard, being able to make available unique facilities, such as drop towers, low-g research aircraft, and the Space Shuttle, as well as a considerable amount of available expertise in this area. Working in support of academic, industrial, and government investigators, the NASA program channels fundamental and applied research proposals into a process designed to provide a comprehensive examination of a broad range of fundamental and application-oriented fluid processes which would benefit from experimentation in a microgravity environment such as available in drop towers and aircraft, and aboard the Shuttle. Space experimentation would allow an orderly examination of processes in which the contributions of buoyancy forces are insignificant. The virtual elimination of buoyancy would then allow a systematic study of other forces/ transport phenomena masked or suppressed in a terrestrial environment. This should lead to further refinement of the existing theroretical models. The applications of this research include fundamental fluid physics issues as well as key applications in the fields of materials science, combustion, and biotechnology.

II. HISTORICAL BACKGROUND

An intensive microgravity fluid dynamics program was initiated by NASA in the late 1950s because of the need to manage liquid propellants in space vehicles. Therefore, some of the earliest experimental results on microgravity fluid behavior were obtained by NASA engineers and scientists as they addressed complex technology problems in a new and, at the time, unexplored environment. Practical problems, such as establishing liquid-vapor interface configurations to determine the location of propellant, were the driving considerations.[1]

The effort was expanded to include theoretical modeling and ground-based drop tower tests aimed at verifying these predictions. Many early technology issues, such as sloshing dynamics, propellant recovery and transport, and fluid reorientation,[2] were addressed in this manner. The success of programs like Apollo and Centaur was facilitated by the engineering solutions provided by this early low-gravity work. An in-depth understanding of the processes under examination, however, was clearly lacking. The pursuit of expedient solutions often raised a myriad of unanswered questions which were indicative of the truly complex nature of the phenomena being uncovered. Extensive use was made of ground-based and airborne microgravity facilities in an attempt to unravel the physical processes at work. The insights gained lead to the current sophisticated view of microgravity fluid dynamics and to the refinement of a set of objectives for continuing research. Serious limitations were encountered, however, in the short test duration of drop towers and low but variable acceleration levels provided by airborne testbeds which limited their usefulness as research tools. With the advent of the Shuttle a new capability for microgravity research has supplied investigators renewed interest in addressing the scientific objectives.

One other awareness was gained during the early history of microgravity research aimed at solving space technology problems. The microgravity environment provided a virtually unlimited opportunity to investigate known terrestrial processes. The known processes could be viewed from the microgravity perspective through space research and yet bring about new insights into terrestrial phenomena.

III. APPROACH

Much of the early scientific research efforts in the area of microgravity research were based on several overview studies sponsored by NASA. The first study, "Fluid Physics, Heat Transfer and Thermodynamics Experiments in Space," prepared in 1975 by the Southwest Research Institute (SRI),[3] identified several potential micro-g fluid dynamics experiments. Most experiments identified were characterized as having undergone extensive testing in both normal gravity laboratories or in the few seconds afforded by drop tower facilities. The 1975 overview identified five broad categories which it felt could be used to organize further investigation. These categories included Critical Point Thermophysical Phenomena, Fluid Surface Dynamics and Capillarity, Convection Processes, Nonheated Multiphase Mixtures, and Multiphase Mixtures, and Multiphase Heat Transfer. The SRI study identified research topics consisting of a broad spectrum of investigations ranging from fundamental investigations to experiments applied in nature.

A second overview on Materials Science was sponsored by NASA and published in 1978.[4] This overview entitled, "Materials Science Experiments in Space," was concerned primarily with defining the classes of materials research which could benefit from being conducted in space. In addition, the overview contained numerous requests, in each of the classes identified, for more research aimed at establishing the role which fluid dynamics played in the processes being studied. In the conclusions of this report it was stated that "since fluid flow is an important part of the phenomena of interest in materials science experiments in space, it is of prime importance that cooperative efforts between scientists representing these two disciplines be initiated and sustained. This cooperative effort is especially needed in the space programs." The requirement for an intensive fluid dynamics effort was substantiated even more strongly with the advent of the "Materials Processing in Space" report[5] by the Committee on Scientific and Technological Aspects of Materials Processing in Space (STAMPS) of the National Research Council. The concluding section of the report stated that an extensive research program was required in the following areas of microgravity fluid dynamics:

- Fundamentals of convection and of coupled convective and diffusive transport;
- Convection during phase changes and chemical reactions and the interactions of convective tranport with transformation processes, especially those responsible for microstructures of solid materials;
- Dependence of density, viscosity, thermal diffusivity, and mass diffusivity of melts and solutions on composition and temperature, particularly as influenced by buoyancy-driven convection;
- Equilibrium properties and dynamic phenomena at gas-melt and melt-melt interfaces, beginning with surface tensions and interfacial tensions as functions of temperature, composition, absorption of soluble trace contaminants, or accumulation of meniscus-seeking insoluble contaminants;
- Phenomena associated with the intersections of fronts and menisci with solid walls (for example edge effects in solidication and combustion) and contact angles, wetting melt spreading, and junctions where gas, liquid, and solid meet in

three-phase contact lines;

- Tests of theoretical models of fluid flow systems that experienced complicated combinations and distributions of forces or have complex compositions; and
- Parameters related to instabilities associated with critical phenomena.

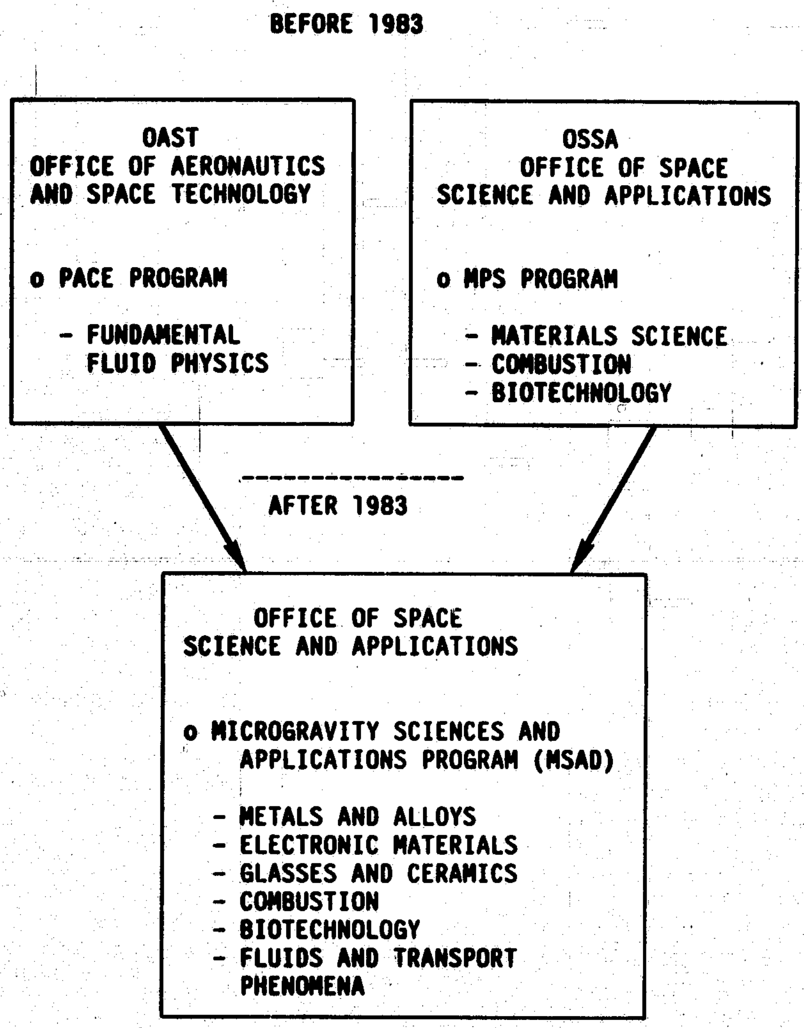
The STAMPS report also acknowledged that "space experimentation will have little value unless its planning is founded on substantial earth-based terrestrial programs." The two overviews and the STAMPS report provided the impetus for the fluid dynamics research efforts supported during the initial phases of the Materials Processing In-Space (MPS) Program and the Physics and Chemstry Experiments (PACE) Program. Until 1983, these two programs were supported by separate offices at NASA Headquarters. There was, however, communications between these two offices in an effort to avoid duplication of research activities.

The fundamental fluid physics experiments were originated out of the PACE program starting in the mid-1970s. The charter of the PACE program was broad in terms of its scientific scope, but it excluded applications research. In PACE, the individual investigations, usually from members of the academic community, were advocated by a particular NASA Field Center and, if successful, managed by that Center. The Principal Investigators selected were noted scientists in their respective fields. Ground-based definition research, which was performed utilizing NASA microgravity facilities, was the backbone of the research effort. While the space-based research could alone provide the long duration microgravity conditions required to fully explore most phenomena, the proper planning and preparation of the space-based research could only be achieved with an intensive ground-based support program. In addition to the development of techniques for space experimentation, contributions to the scientific goals of the program were often achieved in ground-based work. Complete theoretical models often accompanied the ground-based definition phase of most experiments, and in many cases, order of magnitude analyses were performed and the pertinent dimensionless parameters obtained.

In the PACE program, a wide variety of fundamental fluid physics processes were selected based primarily upon the recommendations contained within the overview studies. All experiments in the PACE Program were peer reviewed by teams of independent investigators. During 1983, a comprehensive review of the PACE program was made by an independent science board at the request of the PACE Program Manager and under the direction and supervision of the Universities Space Research Association (USRA). The purpose of the independent review was to first determine whether PACE was concentrating on the most important experiments for flight research and then to scientifically evaluate the individual experiments. Each experiment in the PACE program was peer reviewed by examiners external to the PACE process and those results presented to the science board responsible for the final selections. Ten fluid physics experiments were identified through this process.

The other portion of the fluid physics program was supported under MPS. These experiments were focused on investigating fluid processes specifically associated with material science. The STAMPS committee report had defined the urgent need for additional fluid physics research to establish a strong foundation for material studies. The MPS program responded by issuing Application Notices (ANs) which requested research proposals in this area. Proposals were sent directly to NASA Headquarters and were selected based on successfully passing external peer reviews. Successful proposals were subsequently assigned to NASA Field Centers for technical management.

In late 1983, microgravity research efforts were consolidated by integrating the PACE and MPS activities under the newly formed Microgravity Sciences and Applications Division (MSAD). The objective of this new organization, which maintained PACE as an entity within it, was to centralize program management and to bring all the program participants into closer contact.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OFFICE OF AERONAUTICS AND SPACE TECHNOLOGY
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- FUNDAMENTAL FLUID PHYSICS
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OFFICE OF SPACE SCIENCE AND APPLICATIONS
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- MATERIALS SCIENCE
- COMBUSTION
- BIOTECHNOLOGY
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OFFICE OF SPACE SCIENCE AND APPLICATIONS
o MICROGRAVITY SCIENCES AND APPLICATIONS PROGRAM (MSAD)
- METALS AND ALLOYS
- ELECTRONIC MATERIALS
- GLASSES AND CERAMICS
- COMBUSTION
- BIOTECHNOLOGY
- FLUIDS AND TRANSPORT PHENOMENA

IV. CURRENT SCIENTIFIC INVESTIGATIONS

Listed below are the Fundamental Fluid Physics Experiments currently supported by PACE within the Microgravity Sciences and Applications Division and, therefore, represent pure research investigations.

PACE Program - Fundamental Fluid Physics

Experiment	Affiliation	Investigators
1. Surface Tension Driven Convection	Case Western Reserve University	S. Ostrach Y. Kamotani
2. Free Surface Phenomena	Univ. California, Berkeley	P. Concus
	Stanford University	R. Finn
	Stanford University	B. Hesselink
	Cal Tech	D. Coles
3. Light Scattering Tests of Fundamental Theories of Transport Properties in the Critical Region	National Bureau of Standards	M. Moldover
	University of Maryland	R. Gammon
4. Precise Viscosity Measurements Very Close to Critical Points	University of Maryland	R. Gammon
	National Bureau of Standards	M. Moldover
5. Specific Heat of Helium through the Lambda Point	Stanford University	W. Fairbank J. Lipa
6. Critical Transport Phenomena in Fluid Helium	Duke University	H. Meyer
	Ames Research Center	R. Behringer

PACE Program - Fundamental Fluid Physics - Continued

Experiment	Affiliation	Investigators
7. Electrohydro-dynamics	Princeton University	D. Saville
8. Surface Tension Induced Instabilities; Benard Prob.	Lewis Research Center University of Texas	A. Chai E. Koschmeider
9. Mass Transport Phenomena	University of Toledo	K. DeWitt
10. Test of New Thermodynamic Model of Impurity Extraction by Droplets	National Bureau of Standards	G. Morrison J. Kindaid

In order to illuminate the nature of these experiments, I would like to expand upon two representative efforts. The PACE experiment on "Surface Tension Driven Convection," for example, investigates flows induced by surface tension gradients under microgravity conditions.[6-8] The Principal Investigator, Simon Ostrach of Case Western Reserve University, has identified these flows as extremely important since they dominate many of the phenomena associated with microgravity fluid dynamics and because of their importance in the fields of materials science and combustion. The proposed flight experiments shall involve an examination of the extent and nature of transient and steady-state thermocapillary flows, a determination of the effect of heating mode and magnitude on the flows, and the establishing of the temperature and velocity distributions along the free surface and in the liquid bulk. Ultimately, other important effects shall be considered in space experimentation such as variable boundary conditions/contact angles, end-wall materials, and geometrical configurations. Additionally, the investigation will examine the possibility that surface tension driven flows are inherently unstable owing to coupling between the heat transfer mechanisms and the resulting flow fields.

Paul Concus is the Principal Investigator on the PACE experiment involving "Free Surface Phenomena."[9,10] In microgravity, the free surface of a liquid can behave in striking, unexpected ways. For example, a free surface that is well behaved under terrestrial conditions can rise to arbitrarily large heights or even fail to exist when gravity is absent. The primary objective of the planned flight experiment is to provide an accurate quantitative description of the configuration of capillary surfaces in microgravity. The configurations of primary interest are those near the mathematically predicted critical points which govern the transition from existence to nonexistence in microgravity. In particular, the physical nature of the recently found type of discontinuous behavior represented by the transition to nonexistence in cylinders of trapezoidal cross section will be examined. For the case of trapezoidal cross section, a distinctly different type of discontinuous behavior can occur as a critical configuration is traversed. The importance of space to this experiment is the long duration of low gravity available which is free of external disturbances. The configurations under study will be those whose existence is extremely sensitive to boundary conditions and small perturbations around these conditions.

The "Surface Tension Driven Convection Experiment" by Dr. Ostrach and the "Free Surface Phenomena" experiment by Dr. Concus are very close to initiating flight hardware development. A third fluid physics experiment, "Specific Heat of Helium through the Lambda Point," Principal Investigator, William Fairbank of Stanford University, has undergone a thorough science review and is currently in the conceptual design phase. The "Light Scattering Experiment at the Critical Point" by Robert Gammon of the University of Maryland and M. Moldover of the National Bureau of Standards will also undergo a conceptual design review in 1985.

As stated earlier, MSAD supports experiments in both fundamental and applied sciences. The following list encompasses representative experiments of MSAD which fall outside of the PACE program in fundamental physics and are, therefore, more applied in nature.

Microgravity Sciences and Applications Division
Applied Fluid Physics

Experiment	Affiliation	Investigators
1. Convection During Unidirectional Solidification	National Bureau of Standards	S. Coriell R. Schaeffer G. McFadden
2. Modelling Directional Solidification	Clarkson	W. Wilcox T. Paptheodorou
3. Crystal Melt	MIT	H. Greenspan
4. Transient Heat Transfer	National Bureau of Standards	P. Giarrantano V. Arp
5. Thermodiffusocapillary Migration	University of Colorado	R. Sani
6. Stability of Thermocapillary Flows	Northwestern University Stanford University	S. Davis G. Homsy
7. Studies of Wetting and Multilayer Absorption	National Bureau of Standards	M. Moldover J. Schmidt J. Cahn R. Kayser
8. Solutal Convection and Its Effects on Crystal Growth and Segregation	MIT	R. Brown A. Witt
9. Float Zone Energy Stability Calculations	Arizona State University	P. Neitzel
10. Fluid Flow in Crystal Growth	MIT	R. Brown
11. Transport Processes Research	Princeton University	D. Saville

Microgravity Sciences and Applications Division
Applied Fluid Physics - Continued

Experiment	Affiliation	Investigators
12. Mathematical Models of Continuous Flow Electrophoresis	Princeton University	D. Saville
13. Transport Processes in Solution Crystal Growth	Lewis Research Center	A. Chai
14. Morphological Stability and Fluid Dynamics of Vapor Crystal Growth	University of Utah	F. Rosenberger
15. Surface Tension Studies of Liquid Metal Systems	Ohio State University	D. Stroud W. Shih
16. Dynamic Oscillations of Rotating Drops	Jet Propulsion Laboratory	T. Wang
17. Geophysical Fluid Flow	University of Colorado Marshall Space Flight Center	J. Hart W. Fowlis

This list is a representative sample. It was derived by the author based upon the MPS Bibliography provided by USRA. As can be seen, most of the research topics are related primarily to materials science. The experiments on Rotating Drops by T. Wang, and Geophysical Fluid Flow by Drs. Hart and Fowlis, flew aboard Spacelab 3 in April, 1985.

V. FLUIDS AND TRANSPORT DISCIPLINE WORKING GROUP

The Microgravity Sciences and Applications Division has organized its program into six major subdivisions in an attempt to delineate the independent disciplines involved in microgravity research. Metals and Alloys, Electronic Materials, Glasses and Ceramics, Combustion, and Biotechnology represent five of these disciplines. A sixth independent discipline, Fluids and Transport Phenomena, has also been established with a dual objective of conducting research of intrinsic interest to fluid dynamicists and also to conduct research investigations critical to the other five disciplines, recognizing that fluid dynamics plays an important role in each of them. In all of the aforementioned disciplines, a Discipline Working Group has been established to provide program coordination and promote interdisciplinary communication. The primary goals of the Fluids and Transport Phenomena Discipline Working Group are to:

a. Define the key thrust areas within the fluids and transport discipline
b. Establish the important scientific questions to be answered by space experimentation
c. Define and promote the implementation of a sound ground-based science program that will lead to important microgravity space experiments
d. Evolve the long-range plans for flight research.

To achieve these scientific goals, participation of the research scientists from the academic, government, and industrial community has been sought. The present membership of the Fluids and Transport Working Group is as follows:

Julian Szekely/MIT/Chairman
Thomas Labus/LeRC/Vice-Chairman
Sam Coriell/National Bureau of Standards
William Fowlis/MSFC
Thomas Hanratty/University of Illinois
John Hart/University of Colorado
William Langlois/IBM
John Lipa/Stanford
Franz Rosenberger/University of Utah
Dudley Saville/Princeton

Individual members of the committee have been assigned the responsibility of interacting with the other discipline working groups within the NASA Program. The Fluids and Transport Discipline Working Group met twice in 1984 to complete the initial "draft" of their charter and to develop a detailed list of thrusts to be pursued. The charter and its specific objectives must be approved by the Director of the Materials Sciences and Applications Division at NASA Headquarters, Richard E. Halpern, prior to formal acceptance. The following are the current discipline thrusts included within the working groups charter:

a. Cooperative Phase Transitions - Thermodynamic and transport properties, and the complex nonlinear phenomena which occur when fluids undergo phase transitions in the critical regions.

b. Superfluid Behavior - Specific heat of helium through the Lambda point. Test of cooperative transitions in renormalization group theory.

c. Capillary Phenomena - Equilibrium shapes of fluid masses particularly those whose existence-nonexistence is extremely sensitive to boundary conditions, capillary-dominated surface waves, droplet formation and coalescence, and thin films.

d. Surface Tension Driven Flow - Surface tension gradient induced flow due to variation in temperature/concentration. Includes flow with gradients directed along normal to the liquid-gas interface. Stability limits, onset of oscillatory motion, and free surface shapes to be determined.

e. Stability of Growth Interfaces - Interface morphology under well characterized growth conditions. Relative importance of the transport of heat and solute. Criterion for interface instability, dendritic growth.

f. Double Diffusive Flow - Thermosolutal convection and processes in which fluids are substantially influenced by the molecular diffusion of two or more components in a mixture, as well as by thermal and momentum diffusivity. Includes effects of oscillatory acceleration field.

g. Multiphase Flow - Dynamics and transport of heat and mass for liquid-liquid, liquid-gas, and liquid-solid phase including multicomponent systems. Adiabatic two-phase processes directed toward bubble dynamics (formation, coalescence) and flow hydrodynamics. Nonadiabatic processes on nucleate boiling, forced convection, evaporation, and condensation.

h. Electro-kinetic Transport - Charged fluid interfaces with their diffuse layer of space charge includes motion due to imposed electric fields (electrophoresis, electrocsmosis) or mechanical force (electroviscous effects). Increases concentrated on behavior of concentrated suspensions, "non-homogeneous" buffers and the behavior of fluid-fluid interfaces.

i. Magneto/Electrohydrodynamics - Effects of thermoelectric and/or thermomagnetic body forces. Fundamental interest centers on flow regimes and stability systems under the influence of magnetic fields will be examined too.

VI. CONCLUSIONS

The current structure of the Microgravity Science Program is the result of the recommendations and accumulated knowledge gained from several years of supporting research in the field. Contributions from key members of the science community have been infused through several oversight committees, including the 1978 STAMPS committee.

The content of the program is also the result of historical guidance. Our early efforts to explore low-gravity phenomena to address then urgent technological needs has grown to encompass a large pure and applied research program aimed at expanding our understanding of natural processes. In the area of fluid dynamics, much of the program content is a result of questions that were raised by early microgravity investigations. But an equally important part of the program is represented by the research topics which have been identified through interaction with other disciplines such as the material and biological sciences. The inherent value of such communication is reflected in the organization of the news Materials Sciences and Applications Division at NASA Headquarters. By encouraging the interaction between discipline groups, the strength of the overall program is greatly enhanced. Microgravity research efforts supported by NASA have already led to some important successes as indicated by the attached Appendix of published papers. This initial work has started the development of a sound foundation for conducting future fundamental and applied fluid dynamics research.

The establishment of MSAD must be viewed as a critically important milestone to ensuring the success of future microgravity research. Of equal significance is the creation within MSAD of the Fluids and Transport Discipline and its respective working group with a charter encouraging extensive investigations into the pure and applied sciences. This structure acknowledges the importance of fluid physics as a distinct entity while also demonstrating an awareness that fluid processes play a crucial role in other disciplines as well. Members of the working group will be faced with the difficult task in the upcoming year of prioritizing the research initiatives into a workable program plan. Once this task is completed, the future success of microgravity fluids research will be assured.

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APPENDIX

Abbreviated list of papers that have been published in microgravity fluid dynamics:

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15. LAMBDA TRANSITION FLIGHT PROGRAM

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INTRODUCTION

Following the development of the renormalization group (RG) theory of cooperative phase transitions pioneered by Kenneth Wilson[1] in the 1970s, it became increasingly clear that hard-core, definitive experimental tests were lacking. Many experiments indicated approximate confirmation of the predictions of the theory, but few cases existed where a strong confrontation could be made. Indeed, some tests which would be expected to be the most definitive, gave significant discrepancies. For example, the exponent characterizing the divergence of the heat capacity near the lambda-transition of liquid helium (at 2 K) under pressure was about three times the predicted value--well outside the apparent range of experimental uncertainty. In 1975 we began considering new ways to perform much more definitive tests of the theoretical predictions than have been made previously, and concluded that a study of the nature of the lambda transition under reduced gravity conditions offered the greatest potential for advancing our experimental knowledge in this area.

Central to the study of cooperative transitions is the idea of asymptotic behavior of various thermodynamic properties in the limit as the temperature interval from a transition is reduced to zero. Most current theoretical predictions are made in this limit. Thus, it is necessary to explore the properties of the system under study with extremely high temperature resolution. For example, the asymptotic representation of the properties of helium does not become sufficiently accurate for theory testing until one is within a millidegree or so of the lambda point, whereas conventional experimental limitations set in at the microdegree level. On the other hand, in space, the lambda transition is expected to show no intrinsic rounding to the level of 10^{-10} deg or so. The experiment described here is designed to explore the submicrodegree region of this transition using new thermometry technology which pushes the resolution of temperature close to the fundamental limits set by statistical fluctuations. This will open up a whole new region, never before explored, for testing of the theories of cooperative transitions. The tests that will be performed will be more than an order of magnitude more stringent than any performed so far and will shed new light on a major area of condensed matter physics.

One of the properties of most fundamental significance near a cooperative transition is the heat capacity of the system and its temperature dependence. This parameter is predicted to exhibit a cusp-like singularity at the lambda point, with a well-defined curvature. We plan to measure this function over the widest possible temperature range consistent with reasonable accuracy in the results, and with closest possible approach to the transition. From these data we will determine the curvature by standard curve-fitting techniques for comparison with theory. Since the maximum possible resolution is desired, we will be operating at the limit of the performance of the temperature sensors, and a large amount of the time available for the experiment will be devoted to noise reduction by integration. The measurement of other quantities of interest, such as the variation of the order parameter with temperature, will be left for possible future flights.

The experimental package consists of a sample of liquid helium attached to a pair of high resolution thermometers (HRTs) and suspended within a specially developed thermal control system. This assembly will be installed in the Jet Propulsion Laboratory Superfluid Helium Facility, to be flight tested on a Spacelab-II early next year, and will operate at a temperature of about 2 K. The experiment will be controlled by a microprocessor system which will interface with the Shuttle to allow bidirectional data transfer. The experiment will take advantage of low acceleration periods on the Shuttle to take the highest resolution data minimizing the transition rounding. We expect to fly the experiment in the mid-1988 time frame.

BACKGROUND

The most advanced tests of the RG predictions for cooperative transitions are currently from experiments performed near the lambda transition of helium. as is well known, this transition has a number of experimental advantages not found elsewhere; since the system is superfluid below the transition, thermal relaxation times are short, and even above the transition temperature, the thermal conductivity diverges. Also, since it is a fluid system, the problems with strains and crystal defects encountered with solids are avoided. The transition temperature itself is conveniently accessible and in a region where high resolution thermometry is well developed, and where the advantages of superconductivity can be readily applied. In contrast to the behavior near a critical point, the divergence of the compressibility is very weak, minimizing gravity rounding from this effect; and no special care is needed in setting the sample density. These advantages have facilitated the collection of a wide body of accurate data extending to $t \sim 10^{-6}$, where t is the reduced temperature interval from the transition, on which tests of the RG predictions have been made. To date, the data sets most useful for accurate exponent determination have been obtained by Ahlers and co-workers: these include isobaric thermal expansion coefficient data,[2] heat capacity measurements at the vapor pressure,[3] and superfluid density measurements.[4] Also of major importance are the heat capacity measurements of Gasparini and Moldover[5] as a function of ^{3}He concentration, x. Many other experiments have been reported, but generally these have either been analyzed in a restricted way, or are of lower accuracy. These experiments allow the determination of two exponents, α and ζ, characterizing the leading singularities of the heat capacity and the superfluid density, respectively:

$$C_p = \frac{A}{\alpha} t^{-\alpha}(1 + Dt^{\Delta} + \ldots) + B \qquad (1)$$

$$\frac{\rho_s}{\rho} = A_\rho t^{\zeta}(1 + D_\rho t^{\Delta} + \ldots) \qquad (2)$$

where ρ and ρ_s are the total and superfluid densities, respectively. In addition, information on the leading coefficients and other parameters in these expressions is obtained.

The RG calculations give predictions[6] for the exponents α, ζ, and Δ, the coefficient ratios A/A', D/D', and D'/D, and the difference B - B', where the primed quantities refer to Equation (1) below the transition. Also, the universality hypothesis predicts that the above quantities will be independent of "irrelevant" variables, in this case the pressure and ^{3}He concentration, and scaling predicts that $\alpha = \alpha'$. In Table 15.1 we list the theoretical predictions for the above quantities, along with the experimental results.

The most precise determination of a leading exponent reported to date is from the superfluid density data. This is because Equation (2) does not contain a parameter equivalent to B in Equation (1), easing the curve fitting task, and also because ρ_s has been determined from very precise second sound velocity measurements. The agreement with theory shown in Table 15.1 is very good, and recently Ahlers[7] has reported that even better agreement can be obtained if Equation (3) is modified by replacing A with $A_0 + A_1(\rho)t$. In this case, with $\Delta = 0.5$, he obtains $\zeta = 0.6716 \pm 0.0004$, which is within the range of uncertainty of the theoretical estimate. At face value this appears to be very strong support for the RG predictions, but on closer examination there are some remaining difficulties. These stem from the use of second sound to determine ρ_s. The basic relationship for the conversion is

$$\frac{\rho_s}{\rho_n} = \frac{U_2^2 C_p}{S^2 T} + \cdots \quad (3)$$

where U_2 is the second sound velocity, $\rho_n = \rho - \rho_s$, and S is the entropy. This equation is based on two-fluid model of He II, and is assumed reliable to the order of 0.1 percent. Independent verification[8] by alternative measurements of ρ_s shows agreement to a level of about 1 percent over a restricted temperature range, but no measurements yet exist which fully support this assumption; nor have dispersion measurements been made near T_λ. More important, even if Equation (3) can be shown to be correct to the required accuracy, the determination of ζ from the velocity of second sound inevitably requires a knowledge of the heat capacity singularity via the quantity C_p. The velocity data of Greywall and Ahlers was converted to ρ_s using a logarithmic form for C_p, with $\alpha' = 0$. as is now well known, α' is slightly negative and may well be as low as -0.026. Since C_p is a multiplicative factor in Equation (3) the exponent ζ is dependent on α', no matter how accurately the exponent of U_2 is determined. We have reanalyzed these data using the best fit function to our recently obtained data[9] with $\alpha' = -0.013$, and find a shift of ζ of 0.005, more than ten times the statistical uncertainty quoted by Ahlers. Another serious problem occurs when we use the Josephson scaling relation to test the internal consistency of the data by obtaining α' from ζ: the values obtained are incompatible with the input values used in Equation (3). For example, if the input α' is -0.013, then the output value is -0.030, in clear disagreement with experiment. These difficulties with the analysis of the second sound data imply that the determination of ζ must be subordinated to the prior determination of α' to a much greater extent than has been acknowledged previously.

Precise determinations of the heat capacity exponent have been made by three different methods: direct heat capacity measurements at the vapour pressure, isobaric thermal expansion coefficient measurements as a function of pressure along the lambda line, and heat capacity measurements as a function of ^{3}He concentration. The results obtained are summarized in Table 15.2. A quick look at the table reveals an interesting effect: all measurements at the vapor pressure give exponent values close to -0.017, while those at higher pressures or in the mixtures give values close to -0.025. Some aspects of this effect have been discussed by Gasparini and Gaeta.[10] They demonstrated that while the exponent differences are not large compared with the standard errors, the vapor pressure heat capacity data[3,5] are clearly incompatible with the optimum values of the universal parameters from the thermal expansion data.[2] When the constraints $\alpha = \alpha' = -0.026$, $A/A' = 1.11$ and $D/D' = 1.11$ were applied to the heat capacity data, marked systematic deviations were observed, indicating a possible breakdown of universality along the lambda line. The possibility of large uncontrolled systematic errors in the data seems to be discriminated against by the consistency of the many experimental results listed in Table 15.2. If, for some as yet undetermined reason, there is a weak breakdown of universality at the lambda transition, further difficulty is encountered with the superfluid density results. Since the exponent ζ was obtained from data taken over a range of pressures, consistency requires that the conversion from U_2 to ρ_s be performed using the expansion coefficient result for α', exacerbating the problems described earlier in this section.

It is unfortunate that the various high resolution measurements show small but persistent departures from the predictions, weakening the support for universality and the RG results in general. It would appear to be of very high priority to attempt a resolution of these discrepancies between theory and experiment, and it is clear that only the lambda transition has significant potential for improving the experimental results, because of the unique properties of this system. By performing new measurements at higher resolution, possibly approaching $t \sim 10^{-11}$ in space, it will be possible to simultaneously probe much deeper into the asymptotic region and to extend the dynamic range of the measurements. Improved thermometry will also allow more accurate measurements over the whole range, further increasing our knowledge of the asymptotic form of the singularity.

TRANSITION ROUNDING

In a gravity field, the temperature, T_m, at which the maximum specific heat occurs, is given by

$$T_\lambda - T_m = \rho g h \, (dP/dT)_\lambda^{-1}$$

where h is the height of the sample, ρ its density, g the gravitational acceleration, and $(dp/dt)\lambda$ the slope of the λ-line. At this temperature, the relative deviation of the specific heat from the gravity-free value is given by

$$\frac{\Delta C}{C} = (-\ln t + \text{const})^{-1}$$

assuming a logarithmic divergence in C.

At $t \sim 10^{-7}$, $\Delta C/C \sim 5$ percent, and this may be applied as a correction to the data. This correction can be expected to be accurate to 20 percent or better, so that the uncertainty in the corrected data is less than one percent.

Near T_λ the coherence length, r, diverges as

$$r \sim r_0 \, |t|^{-2/3},$$

where $r_0 = 2 \times 10^{-8}$ cm. For a sample whose height is much less than its other dimensions, a fraction of the volume $\Delta V/V = 2r/h$ is within one coherence length of the container walls. If the relative distortion of the specific heat in this surface layer is f, then the relative error in the measurement of the total specific heat is

$$\frac{\Delta C}{C} \sim \frac{2fr}{h},$$

Thus, if f = 10 percent then for $h > 20r_0 \, |t|^{-2/3}$ the relative distortion will be less than one percent. Note that if the other dimensions of the sample are made equal to h, this condition becomes

$$h > 60r_0 \, |t|^{-2/3}.$$

This leads to an optimum sample height, h_o, for a given acceleration g given by

$$h_o = 20r_o|t|^{-2/3} = (tT_\lambda/\rho g)\{dP/dT\}_\lambda$$

with a corresponding

$$t_o = [20r_o\rho g\ (T_\lambda\{dP/dT\})^{-1}]^{3/5}$$

On Earth, $h_o = 0.046$ cm and $t_o = 2.6 \times 10^{-8}$.

Table 15.3 shows the corresponding values for other accelerations, g/g_o where g_o is the value on Earth, but assuming that <u>all</u> sample dimensions are equal to h. This latter condition is necessary for an experiment in space since the direction of the residual acceleration is not fixed. Onboard the Shuttle, conditions do not appear to be significantly better than $10^{-3}g_o$ for any extended period, although possibly $10^{-4}g_o$ could be reached for a few minutes. At $10^{-4}g_o$, the optimum sample height is 3.5 cm and the resolution t_o ~2×10^{-10} for one percent accuracy. To perform this experiment, a thermometer with a resolution and null stability of the order of 4×10^{-12} K is called for, which for many systems is close to the statistical noise limit.

HARDWARE DEVELOPMENT

At the commencement of our hardware development program the highest resolution thermometry available was with germanium sensors and room temperature resistance bridges. These systems have a resolution of about 10^{-7} deg for an input power of 10^{-8} W. To go significantly beyond this, to the 10^{-10} deg range or better, a new device was needed. We designed and tested a paramagnetic salt thermometer with a d.c. superconducting readout system which has now reached the sensitivity limit set by thermal fluctuations in small systems. The principle of the device and some of its characteristics are described briefly below.

The operating principle of the new thermometer is the dc measurement of the temperature dependent magnetization of a paramagnetic salt in a constant external field. The device consists of a single crystal of paramagnetic material tightly coupled to a superconducting pick-up coil connected to a SQUID magnetometer, and located within a long niobium tube which simultaneously maintains a constant applied field and shields effectively against external fluctuations. A cross section of the thermometer is shown in Figure 15.1. The 100-mg salt pill was crystallized from aqueous solution onto a bundle of enameled copper wires to allow thermal contact to an experiment. The thermal relaxation time of the thermometer was less than 2 sec at 2 K.

After crystallization, the salt was ground into cylindrical form and a ten-turn coil of 0.005-cm-diameter niobium wire was wound directly on its surface. This coil had a self-inductance of about one microhenry and formed the lower half of a pair of astatic windings. The leads from the windings were tightly twisted and passed through a small superconducting tube to a SQUID magnetometer located in the helium bath. The astatic coils were centrally located inside a niobium tube with a length-to-diameter ratio of 18, which provided a shielding factor against external noise in excess of 10^7. Fields of up to 500 gauss were trapped in the niobium tube using a solenoid located in the helium bath.

We installed three thermometer assemblies in a four-stage thermal control system for measurements over the range 1.5 to 5 K. One thermometer was operated without a salt pill to measure background drift and to test for the presence of excess noise in the system. Two different salt materials, $Mn(NH_4)_2(SO_4)_3{:}6H_2O$, MAS, and $Cu(NH_4)_2Br_4{:}2H_2O$, CAB, were used in the remaining assemblies.

We calibrated the high resolution thermometers over a wide range of temperature against a germanium thermometer measured with a four-terminal ac resistance bridge. The sensitivity of the MAS thermometer was proportional to $1/T^2$ as expected, while the CAB thermometer exhibited a sensitivity maximum near 1.8 K due to the magnetic phase transition in zero field. The peak sensitivity of this device was found to be $9.5\Phi_0/\mu K$, where Φ_0

is the quantum of flux. The corresponding rms noise level was about 8.4 x 10^{-4} giving a maximum temperature resolution of 8.8 x 10^{-11} deg, in a 1-Hz bandwidth. For comparison the rms thermal fluctuation noise $\delta T_{rms} = (kt^2/C_s)^{1/2}$, where k is Boltzmann's constant and C_s is the heat capacity of salt, is approximately 10^{-10} deg. The uncertainties in this estimate and in the effect of the thermal response time on the noise are at present large enough to consider the discrepancy between these numbers unimportant. At 4 K the sensitivity of the CAB salt is $0.1\Phi_0/\mu K$ and the noise spectrum is indistinguishable from the empty device. On the other hand, the MAS salt has a sensitivity of about $0.3\Phi_0/\mu K$, and significant noise is visible. Unfortunately, the heat capacity of MAS at 4 K is not well known, giving a large uncertainty in the fluctuation noise estimate. At 1.8 K, the noise and the sensitivity of the CAB salt increase substantially and the noise is comparable with that expected from thermal fluctuations as noted above. For the MAS salt at 1.8 K, the resolution was 8.2 x 10^{-10} deg.

It is clear from the above observations that we have constructed a device with the potential of resolving the lambda point to the fullest extent available on the Shuttle. The basic design of the device is very rugged, with no moving parts, giving us confidence that it will perform well in the Shuttle environment. We have estimated the effect of the radiation flux on the device and conclude that while it may be detectable, it should not significantly interfere with the measurements.

In addition to the thermometry, we constructed a thermal control system to house the experiment which had a temperature stability more that two orders of magnitude better than previous devices. This was necessary to control the heat inputs from the suroundings of the sample of helium during high resolution heat capacity measurements. For the flight experiment we need to modify this system to provide a caging mechanism for the experiment. This system will be used during the ground-hold and launch phases to protect the experiment. To maintain operating conditions near the lambda point during flight, the cryogenic instrument assembly will be housed in the JPL Superfluid Helium Facility which will be flight-tested on Spacelab II.

PROGRAM PLANS

Since the flight instrument differs significantly from our laboratory test apparatus, we plan to build both a prototype and flight unit. This approach should give us sufficient early feedback of the design suitability to minimize the cost and risks associated with the flight unit. The prototype testing is expected to be completed in the next 18-20 months with the flight occurring in mid-1988. A number of follow-on flights are being considered that take advantage of our technology developments to study other properties of interest. Possible candidates include a normal helium thermal conductivity experiment, a study of the temperature dependence of the correlation length in the superfluid phase near the lambda point, and search for the analog of Josephson effects in liquid helium.

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Table 15.1. Comparison of Parameter Values at the Lambda Point Predicted by the RG Method with Those Obtained Experimentally (Quantities in Parentheses Represent Constraints)

Parameter	$\alpha(=\alpha')$	A/A'	x	D/D'	B-B'	ζ	D'/D_ρ
RG predictions	-.008 ±.003	1.03	0.521 ±.006	1.17	0	.669 ±.002	.15 ±.18
ρ_s data	—	—	.5 ± .1	—	—	.6749 ±.0007	-.17
Isobaric expansion data	-.026 ±.004	1.112 ±.022	(=.5)	1.29 ±.25	(=0)	—	±.04
^3He - ^4He mixtures	-.022	1.088	(=.5)	—	(=0)	—	—

Table 15.2. Experimentally Observed Values of Heat Capacity Exponent α at the λ-point

Group	Parameter	α
BF & K	C_{Sat}	0.0 ± .05
Ahlers	C_{Sat}	$0.00 \leq \alpha \leq -.026$
Ahlers	C_{Sat}	-.0163 ± .0017
MAP	expansion coefficient	-.026 ± .004
G & M	C_{Sat}	-.0198 ± .0037
G & M	Mixtures	-.025
T & W	C_{Sat}	-.017
T & W	Mixtures	-.024

Table 15.3. Optimum Sample Size, a_o, and Corresponding Temperature Resolution, t_o as a Function of Residual Acceleration, g/g_o

g/g_o	t_o	a_o (cm)
1	5.0×10^{-8}	.088
10^{-1}	1.3×10^{-8}	.22
10^{-2}	3.2×10^{-9}	.56
10^{-3}	7.9×10^{-10}	1.40
10^{-4}	2.0×10^{-10}	3.5
10^{-5}	5.0×10^{-11}	8.8
10^{-6}	1.3×10^{-11}	22.2
10^{-9}	2.0×10^{-13}	351.

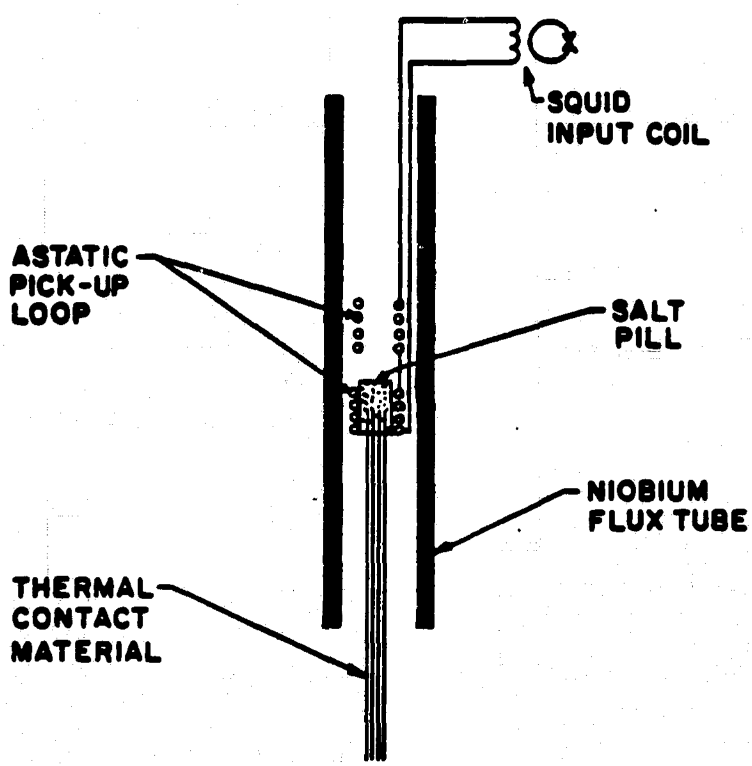

FIGURE 15.1. Paramagnetic salt thermometer.