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**A COLLATION AND EVALUATION OF ANALYSES
AND EXPERIMENTS ON THE RE-ENTRY
BEHAVIOR OF PuO_2 MICROSPHERES**

August 1968

Weiner Associates, Inc.
Baltimore, Maryland

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A COLLATION AND EVALUATION OF ANALYSES AND EXPERIMENTS
ON THE RE-ENTRY BEHAVIOR OF PuO_2 MICROSPHERES

By

David W. Pyatt
Peter P. Ostrowski

August 1968

Contract AT(29-2)-2518

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Weiner Associates, Inc.
Baltimore, Maryland

A COLLATION AND EVALUATION OF
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THE RE-ENTRY BEHAVIOR OF
PuO₂ MICROSPHERES

August 1968

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ABSTRACT

Weiner Associates, Inc. contracted with the USAEC to collate and compare all past analytical and experimental information generated by the AEC and its contractors on the re-entry performance of PuO_2 microspheres.

The objectives of this contract AT(29-2)-2518 were achieved by:

1. Collation of all pertinent past and present analytical and experimental work on PuO_2 microsphere re-entry.
2. Review and digestion of each pertinent analysis and experiment including, if required, discussions and/or correspondence with the technical authors to perceive as deeply as possible the philosophy of their approach and the implications and restrictions of the boundary conditions used by each in their solutions.
3. Specifically identifying those key parameters and assumptions which limit the effectiveness of the theories reviewed.
4. Derivation of a set of specific parameters common to all the analyses and experimental studies which will allow comparison of these theories and results and identification of the magnitude and extent of the present band of analytical solutions.

The conclusions reached through this analysis are:

1. For the first time, the pros and cons on all the analyses and experiments on the re-entry of PuO_2 microspheres based on a detailed review of this work, has been documented. A state-of-the-art survey was established.
2. The theories, such as the ones formulated by G.E. and Aerotherm, to predict breakup or instability for both fully and semi-molten microspheres are too approximate to be used. At best, order of magnitude values were generated. Unfortunately, the AEC-SNS requires more accurate information to determine whether breakup (and mass loss resulting from breakup) will occur for all possible atmospheric release points.
3. The most useful data for correlating breakup are the CAL shock tunnel experiments on zirconium and Fe_2O_3 particles and, to a lesser extent, the Sandia arc tunnel experiments on $^{238}\text{PuO}_2$ microspheres. However, the experimental results, for both programs, cannot be directly applied to real atmospheric release conditions.
4. Based on the theoretical studies, melting initiation can be predicted in release coordinates.

5. Once melting begins, the fluid dynamic behavior of the molten PuO_2 and its effects on the heat transfer is not known. The available thermal models do not take the flow deformation effect into account. Significant deformation will result primarily due to deceleration forces, based on Chapman's tektite studies and the theoretical analysis presented in Section II-F of this report (page II-75).
6. The only experiments where deceleration forces were included are the CAL shock tunnel experiments.
7. Based on the Sandia arc tunnel experiments, two criteria for mass loss were observed:
  - a. Non-breakup mass loss that can be attributed to vaporization, boiling of impurities or trapped helium blowout
  - b. Breakup and/or stripping

The first mechanism results in particles ranging in size from molecular diameter to approximately $1\mu\text{m}$. The second mass loss mechanism results in particles from submicron to greater than 10 microns in diameter.

TABLE OF CONTENTS

	<u>Page</u>
Abstract	iii
Table of Contents	v
List of Figures	vi
List of Tables	viii
 <u>Section</u>	
I. INTRODUCTION AND SUMMARY	I-1
A. Introduction	I-1
B. Program History	I-1
C. Program Objectives	I-3
D. Summary of Results	I-3
Section I References	I-13
II. COLLATION OF PAST ANALYTICAL AND EXPERIMENTAL ANALYSES	II-1
A. Re-entry Trajectory and Aerodynamic Heating Analysis	II-1
B. Thermal Temperature Response of Microspheres During Re-entry and its Implications on the Melting, Fracture and Mass Loss Phenomenon . .	II-15
C. Stability and/or Mass Loss of Partially and Fully Molten Particles	II-28
D. Heat Transfer and Mass Flow During the Liquid/ Solid Phase Change	II-59
E. Mass Loss by Vaporization	II-67
F. Sandia Arc Tunnel Experiments	II-71
Section II References.	II-83
III. ASSESSMENT OF FUTURE EXPERIMENTAL ANALYSES	III-1
A. CAL Levitation Experiments	III-1
References	III-1
IV. CONCLUSIONS	IV-1
Appendices A and B	

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
I-1	Approximate Distribution of Debris as a Function of Weber Number for Sandia Arc Tunnel Experiments	I-11
II-1	Comparison of Various Continuum-Free Molecule Flow Drag Coefficient Bridging Techniques	II-8
II-2	Release Conditions for Melting of Stable and Spinning Spheres	II-16
II-3	Release Conditions for Melting of Stable and Spinning Spheres from Various Investigators	II-17
II-4	Partial Melting Curves for Two Sphere Sizes	II-19
II-5	Thermal Properties - PuO_2	II-21
II-6	Comparison of Time Temperature Response Analyses from Various References	II-23a
II-7	Comparison of Time Temperature Response Analyses from Various References	II-23b
II-8	Plasma Jet Apparatus for Melting Ablation	II-36
II-9	Fully Molten Particle Breakup for $W_{e\text{CRIT}} = 4$ and 6	II-38
II-10	Weight Distribution by Crater Size CAL Experiments	II-29
II-11	Application of the Reynolds Stability Theory when the Disturbance Wave Length is a Fraction of the Microsphere Circumference	II-49
II-12	αR as a Function of Release Conditions and Melt Layer Thickness for a 250μ Diameter PuO_2 Microsphere	II-50
II-13	αR as a Function of Release Conditions and Melt Layer Thickness for a 50μ Diameter Microsphere	II-50a
II-14	Dynamic Pressure as a Function of Velocity and Altitude	II-51

LIST OF FIGURES (continued)

<u>Figure</u>		<u>Page</u>
II-15	Comparison of Instability Models Used by Stanford and G.E.	II-53
II-16	Breakup Phenomena as a Function of Estimated Radius Fraction Melted	II-57
II-17	Partially Molten Particle Instability Determined by Critical Weber Number	II-58a
II-18	Coordinate System for Reference II-13 Analysis . . .	II-63
II-19	Comparison of Calculated and Measured Surface Recession of Tektite Glass in a Wind Tunnel	II-64
II-20	Comparison of Calculated and Measured Surface Recession of Teflon in Wind Tunnel	II-64
II-21	Sandia Arc Tunnel Experiments. Comparison of Lines of Constant Heat Flux with Lines of Constant Weber Number	II-72
II-22	Coordinate System for Approximate Solution to Boundary Layer Equation for a Liquid Layer in Free Molecular Flow	II-75
II-23	Effect of Deceleration Forces on Liquid Layer Shear	II-78
II-24	Effect of Weber Number and Deceleration Forces on Liquid Layer Shear	II-79

LIST OF TABLES

<u>Table</u>		<u>Page</u>
II-1	Aerodynamic Flow Regimes	II-3
II-2	Thermal Properties of $^{238}\text{PuO}_2$	II-20
II-3	Peak Temperature of PuO_2 Microspheres with an Emittance of 0.75 Compared to Those with an Emittance of 0.55	II-22
II-4	Predicted Melting Times, Aerotherm Model	II-26
II-5	Predicted Melting Times (Reference II-4)	II-29
II-6	Various Non-Dimensional Parameters to Describe Liquid Droplet Breakup	II-30
II-7	Correlation of Approximate Values of Critical Weber Number Obtained from Various Sources for Fully Molten Particles in Hypersonic Flow	II-33
II-8	Approximate Critical Weber Numbers as Determined Experimentally and Theoretically for Partially Molten Spheres	II-41
II-9	CAL Test Conditions	II-56
II-10	Comparison of Time to Remove Liquid Mass at Stagnation Point to Time for Melting	II-61
II-11	Viscosity for Several Stony Meteoric Materials in Poise	II-65 b
II-12	Expected Tumor Incidence from Inhalation, Critical Weber Number = 5.0	II-70
II-13	Sandia Arc Tunnel Experimental Conditions	II-71
II-14	Percentages of Total Mass of $^{238}\text{PuO}_2$ Ablated from Microspheres	II-82

I. INTRODUCTION AND SUMMARY

A. INTRODUCTION

One of the most promising aerospace nuclear safety philosophies developed in the past by the Atomic Energy Commission during the course of its Space Isotope Fuels Development Program, was the "Microsphere Isotope Fuel Form."

The concept ideally combined the properties of fuel form inertness to the re-entry, impact and post-impact environments with release at high altitudes, during re-entry destruction of the RTG, yielding dispersal of the microspheres during their terminal flight, to such an extent that eventual ground and airborne concentrations may not present a physical, radiological or toxic hazard to the earth's population.

Although some doubts about the re-entry performance and stability of the microspheres still exist, the potential of this fuel form demands that work be continued to exactly determine its real potential for survival during re-entry.

In the near future, the AEC will still have the responsibility of fueling isotope SNAP systems and the hard realism of short lead time programs will force the use of available well known fuel forms rather than developmental fuels.

Consequently as many of the remaining unknowns of the microspheres have to be determined as possible in order to make the optimal management decisions on application to present and near future programs and on new fuel form development programs.

B. PROGRAM HISTORY

During the years from its inception, the PuO₂ microsphere fuel form has been fabricated, tested and incorporated into several state-of-the-art RTG's and is being considered for several advanced design concepts.

As the development of this fuel form progressed, certain pessimistic analyses of its re-entry performance have indicated the possibility of surface melting and mass loss under certain high altitude release conditions.

Because of the great difficulty in experimentally simulating the exact re-entry environment (thermal, chemical and surrounding flow fields simultaneously) on the ground, the key question of whether any of the molten surface of the fuel form is lost from the microsphere in a respirable size range that could experience prompt fallout, still remains speculative at this point in time.

Because of the importance of this problem, the Atomic Energy Commission undertook to fund a series of analytical studies during 1966 through 1968 on the re-entry history of PuO_2 microsphere fuel forms with emphasis on predicting the probability and magnitude of the surface melting and mass loss.

Many studies were completed in June 1967 under the supervision of Sandia Corporation, by Aerotherm Corporation, Stanford University, The General Electric Company, The Martin-Marietta Corporation, The General Motors Corporation and the Cornell Aeronautical Laboratories.

However, after a review of these technical reports, it became clear that a firm technical picture of the total re-entry history of the microsphere cannot be gleaned from any one of these analyses.

Although the mathematical set-up of the problems may be reasonable, the complexities of the real physical action and interaction of the surface melt with the microsphere core and the surrounding flow field have not been exactly defined.

In addition, the lack of knowledge about the real physical properties of the surface melt at the temperatures and under the influence of the re-entry environment is still in doubt.

Since this data is vital in defining the coefficients of the differential equations controlling the character of any meaningful analytical solution, the validity of present analytical approaches may be seriously affected.

Because of the difficulties outlined above, it has become evident that new experimental programs to generate this information and support the analysis may be required for a meaningful solution to the problem and a final judgment as to the validity of the entire "Microsphere Fuel Form" safety philosophy.

This test program should use the real fuel and simulate the real flight conditions of velocity, deceleration and altitude as close as possible.

C. PROGRAM OBJECTIVES

Early in 1968 Weiner Associates, Inc. contracted with the USAEC to review all past analytical and experimental work which was done or in progress on the microsphere re-entry problem and compile a detailed state-of-the-art behavioral description of re-entering RTG microsphere fuel forms based on this collation, evaluation and comparison.

This was accomplished by:

1. Collations of all pertinent past and present analytical and experimental work on PuO_2 microsphere re-entry.
2. Review and digestion of each pertinent analysis and experiment including, if required, discussions and/or correspondence with the technical authors to perceive as deeply as possible the philosophy of their approach and the implications and restrictions of the boundary conditions used by each in their solutions.
3. Specifically identifying those key parameters and assumptions which limit the effectiveness of the theories reviewed.
4. Derivation of a set of specific parameters common to all the analyses and experiments studies which will allow comparison of these theories and results and identification of the magnitude and extent of the present band of analytical solutions.

D. SUMMARY OF RESULTS

As discussed in the preceding sections, a considerable amount of theoretical and experimental work on the aerothermodynamic behavior of PuO_2 microspheres has been performed. The effort has centered on (1) melting for various release conditions, (2) breakup or no breakup of the partially or fully molten particles, and (3) the character of the debris resulting from breakup if it occurs.

The theoretical analyses have been performed by The Sandia Corporation, Aerotherm Corporation, The Martin Company and General Electric--Valley Forge. Experimental studies have been performed by Sandia, Cornell Aeronautical Laboratories (CAL) and General Motors Defense Research Laboratory (GMDRL). As a result of these studies, many conclusions have been made by the various investigators on the melting and breakup characteristics of microspheres. In some cases the conclusions of all, or most, of the investigators agree. However, in some areas, primarily the area of mass loss or particle breakup, the conclusions are quite different. The ballistic gun range experiments at GMDRL have just been completed as of this writing and a firm set of conclusions cannot yet be made on this program. However, the CAL and Sandia experiments, as well as the theoretical studies, have all been essentially completed, and the results have been documented, evaluated and final conclusions were postulated.

In attempting to compile and collate a behavioral description of PuO₂ microsphere re-entry, the salient conclusions from the work of past investigators have been reviewed and tabulated. In general the various studies agree as to the conditions for the initiation of melting. However, considerable disagreement of molten particle breakup is evident. As to the character of debris resulting from particle mass loss, the only information pertinent to PuO₂ microspheres obtained to date is from the Sandia arc tunnel experiments. Thus, a true collation of this phenomenon is somewhat limited.

The key conclusion of the various investigators and the opinion of WAI on these conclusions are summarized here, under the subject areas of melting behavior, breakup behavior and debris resulting from breakup.

1. Melting Behavior

Sandia Corporation Conclusions (Reference I-1 to I-3)

- a. Based on the latest thermal analysis technique, the SPHERE-2 code, the conditions for the initiation of melting in altitude vs. velocity coordinates have been calculated and are given in Figure II-3 in Section II-B. Also calculated are lines of constant percent melting -- 50% and 100% -- in release coordinates; these curves are shown in Figure II-4 for 50 μ m and 250 μ m diameters.

WAI: A collation of melting initiation curves are shown in Figure II-3 of this report as calculated by Sandia, Martin and Aerotherm. The agreement between the various analyses is only fair. Also, as discussed in Section II-B, the 40 node Sandia thermal model is an accurate simulation of an actual microsphere in a hyperthermal environment until melting begins. Thus, WAI feels that the Sandia melt initiation curves--for both spinning and non-spinning spheres--are reasonably accurate. However, once melting begins, the Sandia model does not account for molten PuO₂ flowback; it is assumed that the spherical shape continues; this is discussed in Section II-D. Thus, the calculated lines for 50% and 100% melting can be considerably in error.

WAI feels that engineering decisions on mass loss should not be based on the Sandia melting analysis once appreciable (>15-20%) melting takes place.

Aerotherm Conclusions (Reference I-4)

- a. A matrix of release conditions to be analyzed by Aerotherm was defined by Sandia and was analyzed for 50 and 250 μ m spheres. These conditions are given below:

<u>Altitude</u> <u>(ft.)</u>	<u>Injection Velocity</u> <u>(ft./sec.)</u>
244,000	25,200
200,000	16,500
181,000	21,000
118,000	12,000

For all cases, except a 250 μ m sphere released at 118,000 ft., material melting was predicted. No melt initiation curve was generated.

WAI: A review of the Aerotherm thermal model and basic assumptions leads WAI to conclude that the temperature vs. time and melting initiation predictions for the above release cases are fairly accurate.

- b. For the assumptions that there is no liquid circulation and no surface thermochemical reactions, most of the re-entry trajectory release points result in a prediction of complete melting.

WAI: The above conclusion is true only if the assumption, as stated, of no appreciable liquid flow results once melting begins, is true. However, analyses performed by Aerotherm and experiments performed by Matting and Chapman (Reference I-5) have shown that significant flowback will result once melting begins on the front face. The same limitation is true of the Aerotherm model as well as the Sandia SPHERE 2 Code and the Martin analysis: no deformation of the liquid material is considered. WAI feels that appreciable instead of complete melting is a valid technical conclusion.

- c. "The accuracy and physical relevance of the melting calculations can be improved by considering surface thermochemical reactions and melt layer dynamics. To include the effects of surface thermochemistry in the calculation of thermal response and melting would require an extension of existing calculative techniques. A calculation of melt layer dynamics in two or three dimensions even without coupling to an energy equation is a task of considerable magnitude, and hence a rigorous inclusion of viscous deformation in a transient melting calculation requires a substantial extension of existing computation methods."

WAI: This conclusion is reaffirmed by WAI, and this is one of the key reasons why a meaningful light gas gun experiment, such as the one planned at NOL in FY '69, is necessary.

This is also one of the key reasons why no prediction can be made, based on the analyses done in the past, on catastrophic breakup.

General Electric Conclusions (References I-6 and I-7)

- a. Utilizing a simplified thermal model (no internal heat conduction in the microspheres; i.e., no temperature gradient), the conditions for the initiation of melting in altitude and release coordinates were generated (see Figure II-3 in Section II-B). The original GE study in SC-CR-67-2573 performed a detailed thermal analysis for the same matrix of release conditions which was given to Aerotherm.

WAI: The assumption that there is no appreciable ΔT throughout the sphere is not true, in light of the Sandia and Aerotherm studies. However, the curve for the initiation of melting agrees moderately well with the other three investigators as discussed in Section II-B of this report.

- b. "A new hypersonic melting theory has been developed in which the melting of a microsphere particle, at its stagnation point, is characterized by a first order phase transition. Numerical solutions to the liquid layer differential equations have been obtained at the stagnation point of a melting, decelerating body for both high Reynolds number and free molecular gas phase flow conditions. The surface temperature of the melt, the surface velocity of the melt, the melt thickness and the melting rate have all been correlated for stagnation point conditions in terms of algebraic equations."

WAI: The hypersonic melting theory developed by GE is a first step forward in analyzing the heat transfer and melting behavior once the stagnation point exceeds the melt temperature and molten PuO_2 runback begins. However, due to the complexity of the problem, the melting "theory" is not directly coupled to the heat transfer in the solid particle. The solution also requires the assumption of the temperature ratio at the gas-liquid interface which is not known. In short, the application of this theory to the melting of PuO_2 microspheres is quite limited.

Martin Conclusions (Reference I-8)

- a. Based on the so-called "non-equilibrium" model, which accounts for internal heat capacity but not for internal heat conduction, the melt initiation curve was generated that appears in Figure II-3 of this report. Thus, melting will occur for release conditions below this line.

WAI: Although the assumption that internal conduction effects are negligible is not valid, the melt initiation curve agrees moderately well with the Sandia non-spinning melt envelope. However, the Martin analysis assumes that the aerodynamic heating is surface-averaged around the sphere (similar to a spinning sphere), and the melting envelope should be closer to the Sandia melt envelope for a spinning sphere (See Figure II-3). In general, the Martin analysis is conservative; i.e., predicts melting at release conditions where melting may not occur based on the Sandia and GE analyses.

2. Breakup Behavior of Microspheres

General Electric Conclusions (Reference I-6)

- a. "An analytical method has been formulated to predict the stability of thin liquid layers on spherical particles. Breakup criteria of fully molten spheres have also been obtained. Body forces and external pressure gradients have been accounted for and surface shear stress has been neglected. It has been determined that whether a particle will break up or not is a function only of its release condition. Furthermore, it does not depend on liquid layer viscosity. However, if a particle is unstable, viscosity will determine how quickly the disturbances grow.

"Based on the results of these analyses, it is concluded that the SNAP-19 fuel particles can become molten relatively quickly and that particularly the larger particles, when released at the lower altitudes and higher velocities, common to possible SNAP-19 trajectories, will lose mass by means of particle breakup."

WAI: Since GE has concluded that the breakup conditions are not a function of viscosity, the parameter which governs breakup or instability is the Weber number. The critical Weber number for breakup, based on the GE analysis is 2.0 for fully molten drops and 4.18 for partially molten drops with a thin molten layer at the stagnation point. These values are in fair agreement with the CAL experiments which were run at hypersonic flow conditions for both fully and partially molten particles. However, many of the basic assumptions in the GE model; i.e., neglecting aerodynamic shear stresses and only solving for first order eigenvalues make the analysis very approximate. At best, order-of-magnitude values have been obtained for the critical Weber number for both the fully and partially molten case.

Aerotherm Conclusions (Reference I-4)

- a. "There is a significant difference between liquid layer stability when the external flow is supersonic or free-molecular rather than subsonic. This difference results in a more unstable interface in the high velocity or free-molecule case. This distinction is important in view of the fact that the bulk of existing data on liquid drop breakup is for low subsonic flows."

WAI: While it is true that many more experiments on droplet breakup have been performed for subsonic flow conditions, the experiments at CAL, as performed by Beal and Haas, have shown that the critical Weber number for molten droplets in hypersonic flow is 3-6, which is well within the reported range (1-10) for subsonic flow.

- b. "Two approaches to liquid-gas interface stability were investigated:
- a. the stability of an equilibrium liquid drop, ignoring surface shear, and
  - b. the stability of a flat, viscous liquid layer subjected to free-molecular flow.

Item (a) represents a combination of Taylor and inviscid Helmholtz instabilities; Item (b), a more realistic flow condition, the Craik-Benjamin instability. Viewed in terms of the particle sizes and reentry conditions of interest in the present study, the first approach indicated that equilibrium liquid drops would probably be stable. The second approach indicated that the liquid layer formed on the particle is highly unstable for most of the reentry conditions investigated."

WAI: The analysis for item (a) results in a critical Weber number of approximately 10, which is somewhat higher than the CAL experimental data. For item (b), using the density, size and viscosity for PuO₂ microspheres, the critical Weber number is approximately 0.1-0.2. The critical value of W_e was found to be a function of a non-dimensional parameter M, where

$$M = \frac{\rho \sigma r}{\mu^2}$$

ρ = material density

r = radius

μ = coefficient of viscosity

σ = surface tension

The value of critical Weber number obtained by Reynolds, as discussed in depth in Section II-C and Appendix B, assumes that $\alpha R > 100$. For most flight conditions, this assumption is not valid, and solutions for $\alpha R < 100$ must be performed. In Appendix B, the value of W_{eCRIT} was obtained for $\alpha R < 1$ and can be expressed as Equation B-6. The critical value goes to zero as δ/R approaches zero (where δ = liquid layer thickness and R = radius) and, for finite δ/R , is a function of the angle from the stagnation point.

A review of the theoretical model has shown that many approximations were made that limit the accuracy of the solution for items (a) and (b). At the very best, order-of-magnitude values can be expected. The Aerotherm study has shown that shear instability could be important in causing microsphere breakup. The CAL experiments have shown that breakup occurs at higher flow Weber numbers for partially molten spheres in hypersonic flow for values

of viscosity close to PuO_2 . This is contradictory to the Aerotherm study, and, in general, more confidence should be placed in the experimental data.

- c. In general, the particles can be expected to remain molten for ample time for liquid layer instabilities to cause destruction.

WAI: For breakup due to item (b), a distortion vs. time calculation had not been performed. Thus, although instability was predicted, the above statement should not have been positively made for all instability conditions.

- d. "The significant differences between stability mechanisms in subsonic and supersonic or free-molecular flow make quantitative extrapolation of breakup data hazardous and, likely, less reliable than the analytical criteria developed in the present study."

WAI: As a result of a review of the theoretical criteria by Aerotherm and GE, WAI feels that only order-of-magnitude values were generated. Also, the CAL experiments for fully and partially molten spheres were well-run experiments and were performed at hypersonic flow conditions; thus the CAL data should be more accurate than the theoretical studies.

Cornell Aeronautical Laboratories Conclusions (Reference I-9)

- a. The critical Weber number for fully molten spheres in hypersonic transition flow is near, but perhaps slightly lower than, the value of six customarily accepted for continuum flow.

WAI: Although CAL may be a bit presumptuous in quoting six as the "customarily accepted value," the series of experiments with zirconium and Fe_2O_3 droplets are carefully run, and a value near six for the critical Weber number was obtained.

- b. The Fe_2O_3 spheres that were heated to the partially molten state exhibited less tendency for breakup than did those which were fully molten. Weber numbers before breakup occurred were found to be as high as 10 for partially molten spheres.

WAI: Although a correlation of Weber number to breakup with percent melting cannot be accurately made, the tests do indicate a general trend of increased stability of partially molten spheres over the fully molten situation.

Sandia Arc Tunnel Experiment Conclusions (Reference I-1)

- a. The arc tunnel facility does not reproduce all of the parameters present during actual re-entry, but the literature suggests

(Reference I-1) that the primary parameter missing--the deceleration force--has a destabilizing influence on the liquid layer in the stagnation region.

WAI: An analysis that is discussed in Section II-F of this report has shown that the deceleration forces significantly reduce the effective aerodynamic shear stress at the liquid-solid interface thereby stabilizing, rather than destabilizing, the majority of the melt. Thus, there is a very likely possibility that the lack of deceleration forces make the Sandia tests too severe; i.e., mass loss is observed in the experiments but may not result for real flight conditions.

- b. If one accepts the Weber number as a criteria for mass loss instabilities, the data from the arc tunnel experiments suggest a critical Weber number as low as one. However, based on Sandia's experience in the program, the best engineering judgment that can be made on the results is that mass loss should be expected at the conditions of re-entry that are possible for the microspheres.

WAI: Due to the likelihood that, at certain test conditions at low Weber numbers, mass loss by mechanisms other than stripping or breakup results, it is likely that a critical Weber number is somewhere between 2.2 and 4.6 based on these experiments. Thus, for some atmospheric release conditions there is a possibility of mass loss due to mechanisms other than catastrophic breakup. This is illustrated in Figure I-1.

- c. "Since there is no known analytical technique for calculating the rate of mass loss, estimates of mass loss for the various release regions are based solely on the arc tunnel results. The mechanism for the observed mass loss is not known, but the fact remains that debris is generated. The results of these debris observations essentially are extrapolated directly to re-entry, and it must be recognized that the results may be revised as more information and a better understanding of the mechanisms of mass loss are gained."

WAI: The extrapolation of the Sandia results to actual flight re-entries is not possible based on the limitations of the arc tunnel to simultaneously simulate the heating rates, enthalpy, shear stress and dynamic pressure (Weber number). Further, the deceleration forces in real flight may cause breakup only at the more severe release conditions.

3. Debris Resulting from Breakup of Microspheres

Sandia Arc Tunnel Experiments

- a. "Table IV of Reference I-1 and shown as Table II-14 of this report shows the final size distribution of the ablated debris, based on

FIGURE I-1

APPROXIMATE DISTRIBUTION OF DEBRIS
AS A FUNCTION OF WEBER NUMBER
FOR SANDIA ARC TUNNEL EXPERIMENTS

the Tracerlab analysis and modified by the sampler efficiency analysis. The arc tunnel test conditions are defined in Table II-13 on Page II-71. Although the data presented here are based on one test for each re-entry condition, it is believed that the size distribution of the debris is reasonably representative, especially for Condition A and K (See Table II-13). The Condition M data are questionable because the molten runback material was considered as large ablated particles. The final fate and debris size of this runback material in an actual re-entry cannot be determined in the arc tunnel facility. However, it is apparent that the debris size distribution is dependent on the heat input and melt rate of the microsphere."

WAI: It is felt that much of the debris for Conditions A and K can be due to vaporization (although the possibility of "fines" being removed by the flow stream is also possible). The particles larger than $0.01\text{--}0.05\mu$ may be attributed to impurities or voids in the microsphere which could cause high pressure gases to blow through the liquid layer on the microsphere or due to the condensing of vaporized PuO_2 on inert nuclei (dust) present in the air. Only for Condition M, the most severe test point, was stripping or breakup suspected (based on the fact that molten material ran back on the sting) and this condition had shown shear stresses on the order of twice the real flight values for comparable heating rates. The debris as a function of Weber Number is shown in Figure I-1.

The total gaseous and fine particulate mass loss for Condition A is shown by experiment to be approximately 1% of the original particle mass (Reference I-1) and on the order of 5 to 10% for Condition K. While a finite hazard does exist for this relatively small amount of debris, the hazard is much less than that resulting from catastrophic breakup which could occur at higher Weber Numbers. Additionally, the Weber Number for catastrophic stripping should be higher in real flight than the approximate value (4.6) where runback was observed on the sting in the Sandia experiments.

However, even if the critical Weber Number is as high as 10, flight conditions associated with the re-entry of some SNAP-RTG systems which re-enter at cislunar speeds could cause the fuel to exceed this value and result in catastrophic breakup.

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II. COLLATION OF PAST ANALYTICAL AND EXPERIMENTAL ANALYSES

A. RE-ENTRY TRAJECTORY AND AERODYNAMIC HEATING ANALYSIS

1. Introduction

In order to accurately assess the thermal response of a microsphere undergoing atmospheric re-entry, it is necessary to determine the aerodynamic drag and heat transfer coefficients. The reason for this is that for a given body and given release condition, the drag coefficient determines the trajectory (velocity vs. time), which in turn determines the aerodynamic heating rate, and the heating rate acts as a boundary condition for a transient heat conduction analysis as discussed in Section II-B. It is apparent that inaccuracies in determining the trajectory will result in inaccuracies in the aerodynamic heating rate even when the heat transfer coefficient is known with a high degree of confidence.

In this circumstance, many of the "regime effects" (where regime refers to the relative rarification of the air in the flow environment) become significant in calculating the drag and heat transfer coefficients, and allowances must be made for free molecular, transition and continuum flow, as well as Mach and Reynolds number effects. It is easily seen that any collation and comparison of the techniques used to determine the thermal response of re-entering microspheres must begin with a comparison of the drag and heat transfer coefficients.

Because of the extremely low ballistic coefficient of microspheres, their re-entry trajectory is characterized by very rapid decelerations which occur in a matter of seconds, or less. Accordingly, the re-entry occurs at very nearly constant altitude which means that the free stream density does not change. The combination of small particle radius and low atmospheric density often results in a free molecule flow field surrounding the particle. For a very low altitude release, continuum aerodynamics are applicable. In most release cases the transition between the free molecular and continuum flow regimes must also be considered.

During the re-entry of a body into the earth's atmosphere, the primary interest centers on the momentum and energy exchange between the body and the surrounding air, for these determine the physical state of the particle. Regardless of which flow regime is considered, the kinetic energy of the body is converted into thermal or heat energy in the following manner:

- a. Momentum is transferred from the body to the air through the action of aerodynamic drag.
- b. In free molecule flow, collisions between the body and air molecules transfer heat back to the body.

- c. In continuum flow, heat is transferred conductively and convectively through the boundary layer from the hot gas region behind the shock to the body surface.
- d. In transition flow, the heat transfer mechanisms are not as well defined.

Since these processes are not 100% efficient, the net result is that a portion of the body's kinetic energy is transmitted permanently to the air and the rest is returned to the body in the form of thermal energy, thereby increasing its temperature. When the temperature reaches a level sufficient to reverse the direction of the heat transfer, the thermal energy is transmitted back to the surrounding air by radiation and convection, and the body will begin to cool. This is contingent on the ability of the body to survive extreme temperatures without breaking up. In this event, virtually all of the body's kinetic energy will be transferred to the atmosphere.

It is precisely this aspect of re-entry which is of major concern. When release conditions (altitude and velocity) are severe enough, the level of aerodynamic heating can be high enough such that either partial or complete melting of the particle will occur. Under these conditions it becomes important to determine whether significant mass loss, due to vaporization, ablation, or hydrodynamic instability, might occur. Obviously, these questions can only be answered after the more fundamental one; i.e., which release conditions will result in melting of the microsphere?

Since the heat transfer to the microsphere is coupled directly to its dynamics, the first step in calculating the thermal history is to calculate the trajectory and this must be done numerically, preferably on a computer, as the differential equations which govern the motion of the microsphere are non-linear. An added complication is that the drag coefficient is a function of a Mach number in free molecule flow, while for continuum flow, it is a function of both Mach number and Reynolds number; hence provisions must be made to allow for changes in drag coefficient as a function of the flow regimes for the various release conditions.

As was mentioned earlier, the aerodynamic flow regime may be either free molecular, transitional or continuum depending on the size of the microsphere and its release conditions. This requires that "bridging" techniques be available for use in the transition regime. This is not extremely critical for the drag coefficient which varies by a factor of approximately 2 from continuum to free molecular flow, but greater care is necessary with the heat transfer coefficient which may vary by as much as two orders of magnitude between flow regimes.

2. Deceleration Profiles as a Function of Release Conditions

a. Flow Regimes

Because of the relevance of certain physical phenomena which occur, the aerodynamics of bodies in flight are usually classified into flow regimes which are characterized by (1) the speed of the body relative to the local speed of sound of the gas through which it is flying, and (2) the size of the body relative to distance between the gas molecules. These are termed the Mach number and Knudsen number respectively. Although the various flow regimes overlap and the boundaries between them are not distinct, an approximate classification is given below in Table II-1.

TABLE II-1
AERODYNAMIC FLOW REGIMES

Mach No.	Regime	Knudsen No.	Regime
$M \geq 5$	Hypersonic	$Kn \geq 10$	Free Molecule
$1 < M < 5$	Supersonic	$.01 < Kn < 10$	Transitional
$M \approx 1$	Transonic	$Kn < .01$	Continuum
$M < 1$	Subsonic		

Because of the relatively high melting temperature of PuO_2 (approximately 4500°R), hypersonic speeds are required to produce significant aerodynamic heating rates for microspheres in the 50 to $250\mu\text{m}$ diameter range. Hence only hypersonic flow will be considered here. Possible release conditions for the microspheres are expected to vary from about 250,000 ft., where the mean free path of the air molecules is roughly 10^{-2} ft., to about 100,000 ft. where the mean free path is approximately 10^{-4} ft. When these figures are compared with the diameter of a 250μ microsphere (8.2×10^{-4} ft.), it is clear that flow regimes ranging from free molecule to continuum are possible.

In the free molecule flow regime, where air molecules may travel relatively large distances before colliding with another molecule, the microscopic or thermal motion of the molecule may become as important as the mean flow. Consider, for example, the case where these velocities are of the same order of magnitude.

Then collision between any body placed in such a flow and the air molecules are just as likely to result from the thermal motion as the mean flow field. Such is not the case, however, when the mean velocity is much larger than the thermal velocity, for here the former will dominate. It is to be expected, then, that the momentum and energy exchange between a solid body and an incident free molecule flow field will be dependent on the molecular "speed ratio," which is defined as the ratio of the mean flow velocity to the molecular thermal velocity, and may be expressed by Reference II-1.

(II-1)

$$S = \sqrt{\gamma/2} M$$

γ = ratio of C_p/C_v of the gas

M = Mach number

In addition to the speed ratio, drag and heat transfer coefficients are also functions of the manner in which a molecule is reflected from a solid surface. If the collision is "specular" (elastic), the component of the molecule's velocity which is normal to the surface is reversed, while the tangential component is unchanged. On the other hand, if the molecule is trapped or adsorbed on the surface for a brief period, the re-emittance velocity will depend on the thermal speed rather than the mean flow speed. This type of reflection is known as diffuse reflection, and it is commonly assumed that molecules undergoing diffuse reflection leave the surface with a Maxwellian velocity distribution (Reference II-2). Consequently, when the type of reflection is specified, the momentum and energy of the re-emitted molecules is known, and the drag and heat transfer coefficients can be determined.

An inherent feature of free molecule flow is that reflected molecules collide with other molecules at relatively large distances from the body. The reflected molecules have no influence on the incident molecules and viscous boundary layers and shock waves do not exist. As the air density increases, however, the mean free path of the molecules decreases, and it is possible for a reflected molecule to collide with an incident molecule, thereby altering the energy with which the incident molecule strikes the surface. This interaction results in the formation of shock waves and viscous layers and must be taken into account when the mean free path is small compared to the size of the body; i.e., in continuum flow.

In general, there are two separate theories for continuum and free molecule aerodynamics, and common practice is to use a

bridging function which gives a smooth transition from one regime to the other. These will be discussed in connection with the various theories below.

b. Spinning and Non-Spinning Spheres

As was mentioned earlier, the dynamics of a re-entering microsphere have a direct effect on its thermal history. The same can be said for the microsphere's motion about its own center of gravity. Since the microsphere is symmetric, spinning will not alter the deceleration because the drag coefficient remains unchanged. The total heat flux to the particle is also unaffected, but the manner in which the heat is absorbed does in fact depend on whether the microsphere is spinning. In the case where stable entry occurs, the major aerodynamic heating will be the highest near the stagnation point, and the sides and back surface of the microsphere will remain relatively cool. Hence, with most of the heating concentrated at the stagnation point, melting can occur rather quickly. If, however, the microsphere is tumbling randomly, the stagnation point will sweep around the surface of the microsphere and cause it to be heated uniformly at a much slower rate even though the total heat flux is the same.

In the seven analyses considered here (Reference II-3 to II-9), the G.E. and Sandia analyses (References II-3 and II-9) considered both spinning and stable microspheres. Curiously enough, the G.E. study shows very little difference in the temperature response of the front and rear stagnation points for the stable case, and also that the spinning microsphere heats up quicker. No explanation is given although the results of the G.E. analysis are inconsistent with the Sandia work. The Sandia studies show the more reasonable results that melting initiates faster for the non-spinning spheres at the same release conditions; i.e., faster temperature response at the stagnation point.

c. Comparison of Drag Coefficients in Free Molecule, Continuum and Transition Flow

To date, the major re-entry analyses of PuO_2 microspheres may be found in References II-3 to II-6. Although Reference II-3 does not present any information on the theory used to obtain the drag coefficient for the microspheres, the others do, and these are given below for comparison.

(1) Aerotherm Corporation (Reference II-4)

Free molecule flow

(II-2)

$$C_{D_{F_M}} = \frac{2}{\sqrt{\pi}} \frac{e^{-s^2}}{s} \left(1 + \frac{1}{2s^2} \right) + 2 \left(1 + \frac{1}{s^2} - \frac{1}{4s^2} \right) \text{erf}(s) + 2 \frac{\sqrt{\pi}}{s}$$

where s is the speed ratio given by Eq. (II-1)

erf is error function

Continuum flow (II-3)

$$C_{D_C} = C_{D_0} \left[1.0 + 1.29 \exp - \left(\frac{4.27}{M^{4.53}} + \frac{370}{R_e^{1.25}} \right) \right]$$

where C_{D_0} is the subsonic continuum drag coefficient

M is the free stream Mach No.

R_e is the Reynolds No. which results in a Knudsen No. of .01.

Transition (II-4)

$$C_D = C_{D_C} + [C_{D_{F_M}} - C_{D_C}] [\sin^2 \pi (1/3 + 1/6 \log_{10} \text{Kn})]$$

where Kn is the Knudsen No.

(2) Sandia Corporation (Reference II-5)

Free molecule flow (II-5)

$$C_{D_{F_M}} = 2.0$$

Continuum flow (II-6)

$$C_{D_C} = 1.0$$

Transition

Figure 1 of Reference II-5 shows a plot of C_D vs. Kn which was taken from Reference II-7. The data show a smooth variation of C_D from a value of 1.0 at $\text{Kn} = .001$ to 2.0 at $\text{Kn} = 10$.

(3) Martin Company

Free molecule flow (II-7)

$$C_{D_{F_M}} = \frac{e^{-s^2/2} (1+2s^2)}{\sqrt{\pi} s^3} + \frac{4s^4 + 4s^2 - 1}{2s^4} \text{erf}(s) + \frac{2\sqrt{\pi}}{3s} \sqrt{\frac{T_1}{T_\infty}}$$

where T_1 is the body temperature and T_∞ is the free stream static temperature.

Transition and Continuum

(II-8)

$$C_D/C_{D_{FM}} = .8917 - .0922\lambda - .113\lambda^2 + .0032\lambda^3 - .168 \times 10^{-3}\lambda^4$$

$$\text{where } \lambda = \ln \frac{R_e}{M}$$

Equations (II-2) and (II-7) are for diffuse reflections, and both approach a limit of 2.0 for large speed ratios. Since large speed ratios are involved here (See Equation (II-1)), there is very little difference between Equations (II-2), (II-5), and (II-7) over the significant portion of the microsphere trajectory. There is a slight difference in the hypersonic continuum drag coefficients given by Equations (II-3) and (II-8), resulting in values of .95, 1.0, and .85 for the Aerotherm, Sandia and Martin theories. The free molecule--continuum flow bridging functions used in References (II-4 to II-6) are plotted in Figure II-1 and, as can be seen, there is little difference among them. It appears, then, that although there are some slight differences in the drag coefficients which are predicted by the various investigators, these are not significant in the resulting trajectories.

d. Comparison of Aerodynamic Heating Rates

In order to determine the temperature response of the microsphere during re-entry, it is necessary to calculate the appropriate heat transfer coefficient (free molecule, transitional or continuum). This is then used to calculate the heat flux at the solid surface of the microsphere, where melting, mass transfer and vaporization may have to be taken into account.

The technique used by G.E. differs significantly from the usual approach and will be discussed first. The analysis is quite complex and can be only briefly summarized here; however, the details may be found in Reference II-3. The method considers heat blockage effects due to the flow of the melt layer and provides a unique way of calculating the surface temperature of the microsphere during melting without recourse to a finite-difference solution to the heat conduction equation.

While this method is unique in that it circumvents using a digital computer, it was obviously done to conserve time, and its accuracy is open to question!

The heat flux at the gas-liquid interface is calculated by means of correlations with the mass transfer rate and gas velocity gradient given in Reference II-7. The correlations are valid for high Reynolds number laminar continuum flow, so correction factors must be applied in the free molecule and low Reynolds number regimes. During the course of the computations, it was found that the gas phase heat transfer rate was independent of the surface velocity of the melt layer (but dependent on the gas layer velocity gradient) and this effect was neglected. The velocity gradient of the gas layer is given by:

$$\frac{R_B}{V_\infty \cos \phi} \frac{du_e}{dx} = 1.0 \quad \text{free molecule flow} \quad (\text{II-9})$$

$$= \sqrt{2\epsilon} \quad \text{continuum flow}$$

where R_B is particle radius

V_∞ is free stream velocity

ϕ is the angle measured from the stagnation point

ϵ is the density ratio across a normal Rankine-Hugoniot shock wave

Once the shear stress and heat transfer rate at the gas-liquid interface have been found, the liquid layer equations can be solved using these for boundary conditions at the outer edge. The results of the calculations show that for both free molecular and high Reynolds number flow, all of the solutions can be correlated in terms of four parameters: (1) the ratio of the heat transfer rate in the solid to that in the liquid, (2) the melting rate, (3) the liquid layer thickness, and (4) the solid surface temperature.

Only the gas phase velocity gradient correlations are used until melting begins, after which the melt correlations are used as well. This requires knowledge of the first parameter, which must be estimated and the other three are determined directly from the correlations.

This is the biggest drawback of this particular method. In their report, G.E. gives "two possible variations" of the heat transfer parameter with time, but gives no further information as to which was used and how.

A more conventional approach to the microsphere heating was taken by the Aerotherm Corporation (Reference II-4). The heat transfer coefficients are given by the following expressions:

Free molecule flow

$$(\rho_e U_e C_H)_{FM} = \alpha_T \rho_\infty U_\infty \sin\theta \quad (II-10)$$

where $\alpha_T \approx .9$ = thermal accommodation coefficient

θ is the angle measured from the tangent point

Continuum flow

The stagnation point heat transfer coefficient is

$$(\rho_e U_e C_H)_o = \frac{.985 \times 10^{-3}}{R_e^{1/2}} (U_\infty \times 10^{-4})^{.193} (P_o)^{1/2} \quad (II-11)$$

where P_o is the total pressure at stagnation point,

and the distribution around the microsphere is given by:

$$\frac{(\rho_e U_e C_H)_c}{(\rho_e U_e C_H)_o} = \frac{\rho_e U_e \mu_e r}{2 \int_o^s [\rho_e U_e \mu_e r ds]^{1/2}} (P_o \mu_o \frac{du_e}{ds})^{1/2} \quad (II-12)$$

where s is the distance measured on the surface of the microsphere

r is the radius

M_e is the Mach number

ρ_e is the density

U_e is the velocity

and the "e" subscript denotes conditions at the edge of the boundary layer.

For the case of a sphere, the heat transfer distribution given by Equation (II-12) is very nearly sinusoidal (as for free molecule flow), so this was used in all calculations.

Bridging

$$\rho_e U_e C_H = (\rho_e U_e C_H)_C \left[1 - \exp \left(- \frac{(\rho_e U_e C_H)_{FM}}{(\rho_e U_e C_H)_C} \right) \right] \quad (II-13)$$

Subscripts: C = continuum

FM = free molecular

The heating rates on the surface of the microsphere are expressed in terms of the heat transfer coefficient as

(II-14)

$$q_{\text{surface}} = \rho_e U_e C_H H_r$$

for cold wall heating, when re-radiation and the wall enthalpy are taken into account,

(II-15)

$$q_{\text{surface}} = \rho_e U_e C_H (H_r - H_w) - \sigma \epsilon T_w^4$$

where $H_r = U_\infty^2 / 2$ gJ is the recovery enthalpy

H_w is the wall of enthalpy

σ is the Stefan-Boltzmann constant

ϵ is the surface emmissivity

T_w is the wall temperature

Equation (II-15) is coupled to a finite difference solution to the transient heat conduction equation as the surface boundary condition.

Surface melt movement as well as vaporization and surface chemical reactions are calculated, but the effect of these on the heat transfer to the surface of the microsphere is not estimated, although it is expressly stated in the report that these will have a significant effect once melting begins. What this means is that the heat transfer calculations can be considered accurate only up to the initiation of melting. After this point has been reached, vaporization and melt movement will reduce the level of heat transfer such that neglecting these effects can lead to a significant error in the heat transfer rate to the solid portion of the microsphere.

The Sandia Corporation (Reference II-5) has chosen to use essentially the same method as Aerotherm, with the exception that blockage effects due to vaporization are included. The stagnation point heat transfer coefficient is expressed by an equation analogous to Equation (II-13); i.e.,

(II-16)

$$G_o = \psi G_{oc} [1 - \exp(-G_{o|M} / \psi G_{oc})]$$

where $G = \rho_e U_e C_H$

ψ is the "blowing correction factor" for vaporization

The Chapman equation (Reference II-8) is used for ψ

$$\psi = \frac{.94}{C_1/C_2} + .06 \quad (\text{II-17})$$

$$\text{where } C_1 = \frac{.75}{\bar{M}^{.7}} \quad (\text{II-18})$$

$$C_2 = \frac{P_0 \cos \theta}{P_v} - 1 \quad (\text{II-19})$$

where $\bar{M}$ is the ratio of the molecular weights of the blowing gas to the external flow (air).

P_θ is the pressure at any angle (θ) from the stagnation point.

P_v is the equilibrium vapor pressure.

The heat transfer distribution around the sphere is taken to be of the form,

$$G = G_o \phi(\theta) \quad (\text{II-20})$$

$$\text{where } \phi_{FM} = \cos \theta + .0113 \sin \theta \quad (\text{II-21})$$

$$\phi_c = \frac{2 \sin \theta \cos^2 \theta}{\sqrt{\theta^2 - \theta \sin \frac{4\theta}{2} + \frac{1 - \cos 4\theta}{8}}} \quad (\text{II-22})$$

and in the transition region,

$$\phi = \phi_{FM} - [\phi_{FM} - \phi_c] \frac{G_o}{\psi G_{oc}} \quad (\text{II-23})$$

These differ very little from the distribution considered by G.E. and Aerotherm and have been shown to give good results with experiment.

In addition to blockage, vaporization also lowers heat transfer rates due to mass transfer and sublimation. These effects were also calculated from the theory in reference II-8 as,

$$q_{\text{vap}} = \dot{m} \Delta h_v \quad (\text{II-24})$$

where $\dot{m}$ is the mass flow rate

Δh_v is the heat of vaporization

The mass loss rates are given by

$$\dot{m}_{\text{FM}} = \frac{\alpha_L P_v}{\sqrt{2\pi RT}} \quad (\text{II-25})$$

$$\dot{m}_c = \frac{\psi G_c}{\bar{M} C_2} \quad (\text{II-26})$$

$$\dot{m} = \frac{\dot{m}_{\text{FM}} \dot{m}_c}{\dot{m}_{\text{FM}} + \dot{m}_c} \quad (\text{II-27})$$

where α_L is the Langmuir coefficient

R is the gas constant

The heat flow to the surface of the microsphere is essentially the same as Equation (II-15) with the additional term of Equation (II-24),

$$q_{\text{surface}} = G (H_w - H_r) - \sigma \epsilon T_w^4 - \dot{m} \Delta h_v \quad (\text{II-28})$$

and this is used as the boundary condition for the heat conduction computer program.

A direct comparison of the aerodynamic heating rates predicted by the various investigators is virtually impossible. To begin with, the G.E. analysis is a combination graphical-analytical method, and although the "boundary layer" heat flux is presented in Reference II-3 for the re-entry cases considered, this must be corrected for Reynolds number and free molecule flow effects to obtain the actual heat flux to the particle. The situation is further complicated by the fact that the expressions used by Aerotherm and Sandia for the heat flux (Equations (II-15) and (II-28)) are highly non-linear; hence the heat flux cannot be obtained without actually doing the problem, since the particle temperature must be known as a function of time.

There is one point, however, that can be compared. A sample calculation is given by G.E. for the case of the re-entry of a

250 μ particle at a speed of 25,200 ft./sec. and an altitude of 244,000 ft. The stagnation point heat flux to this particle at injection is given in Reference II-3 as 906 BTU/ft.²/sec. Equation (II-15) can be used to calculate the heat flux corresponding to the Aerotherm theory as follows:

From Figure 10 of Reference II-4,

$$G_o = .065 \text{ lb./ft.}^2 \text{ sec.}$$

$$H_r = 12.0 \times 10^3 \text{ BTU/lb.}$$

Assuming an initial wall temperature of 1,000° R (as G.E. did) and an emissivity of .9 for PuO₂,

$$H_w = 450 \text{ BTU/lb.}$$

$$\sigma \epsilon T_w^4 = .432 \text{ BTU/ft.}^2 \text{ sec.}$$

and Equation (II-15) gives,

$$q_{\text{wall}} = .065 (12,000 - 450) - .432 \\ \approx 750 \text{ BTU/ft.}^2 \text{ sec.}$$

For this case, which is the only one for which a comparison can be made, the G.E. heat flux differs from the Aerotherm value by approximately 17%. The Sandia equation (Equation II-28) would give the same result as the Aerotherm value since melting has not begun yet.

e. Conclusions

- (1) For most atmospheric release conditions, the aerodynamic flow regimes will vary from free molecular to transitional. Only releases at very low altitudes will re-enter into pure continuum flow.
- (2) The drag coefficients and aerodynamic heating coefficients in the transition region must be calculated from one of several "bridging techniques."
- (3) The analyses performed by all investigators to predict drag coefficients in the transition regime are in good agreement.
- (4) There is a 17% difference in the aerodynamic heating coefficients between Aerotherm and G.E. in the one case where a direct comparison can be made.

- (5) The stagnation point heating rates for a non-spinning sphere are considerably higher than the surface-averaged heating rates of a spinning sphere.
- (6) A theory was developed by G.E. to account for the heat transfer through a thin molten layer at the stagnation point. However, its usefulness is limited since it can only be solved parametrically with certain variables of the hypersonic flow field.
- (7) The aerodynamic heating rate calculations performed by Sandia, Aerotherm and Martin are reasonably accurate until melting and flow deformation start.

B. THERMAL TEMPERATURE RESPONSE OF MICROSPHERES DURING RE-ENTRY AND ITS IMPLICATIONS ON THE MELTING, FRACTURE AND MASS LOSS PHENOMENON

To answer the fundamental question of "Is there melting of microspheres for various atmospheric release points?" a number of detailed thermal analyses of a single PuO_2 microsphere were performed by Sandia, Aerotherm Corporation, the Martin Company, and General Electric-Valley Forge. The initial approach of both Sandia (Reference II-8) and Martin (Reference II-6) was to assume an infinite thermal conductivity of PuO_2 and thus a constant temperature throughout the sphere. An additional assumption made by Sandia was that the sphere tumbled randomly and did not have a constant stagnation point. Utilizing this approach, which simplified considerably the temperature vs. time calculations, a "melt envelope" was generated in velocity-altitude coordinates. The envelope is a line at which the peak microsphere temperature just equals the melt temperature of PuO_2 and is shown in Figure II-2 for both spinning and non-spinning spheres based on the work done by Stephenson (References II-8 and II-9).

The second approach taken by Sandia to generate the upper curve in Figure II-2, was to assume a non-spinning sphere, which had a stationary stagnation point with respect to the flow. Melting initiated earlier (from time of release) for the non-spinning sphere since the stagnation point heating rates are 3 to 4 times higher than the surface-averaged heating rates. The melting initiation curve is somewhat higher in altitude at the same velocity for this case as shown in Figure II-2. An outgrowth of the non-stable analysis was the presence of large temperature gradients and high ΔT 's throughout the sphere. This meant that the assumption of infinite thermal conductivity was not valid. To solve for the temperature vs. time at various locations in the sphere required that the internal heat conduction be considered. The assumption of infinite thermal conductivity is made in the latest Martin and G.E. thermal analyses and limits the accuracy of their melt initiation curves in Figure II-3.

FIGURE II-2. RELEASE CONDITIONS FOR MELTING OF STABLE AND SPINNING SPHERES
1966 ANALYSIS FOR $T_{\text{MELT}} = 4175^{\circ}\text{F.}$

FIGURE II-3. RELEASE CONDITIONS FOR MELTING OF STABLE AND SPINNING SPHERES FROM VARIOUS INVESTIGATORS

Another effect of the presence of large ΔT 's throughout the sphere during re-entry are large thermal stresses which could possibly cause cracking and fracture. Unfortunately, the mechanical properties of $\text{Pu}^{238}\text{O}_2$ such as Young's Modulus and maximum allowable tensile and compressive stress are not known. It is therefore difficult to analytically predict (1) the magnitude of the thermal stress for a given temperature distribution in the sphere and (2) the allowable stress before cracking occurs. Some experiments were performed at Battelle Memorial Institute at Columbus on plasma-fired microspheres in a rapidly decreasing external pressure environment with no observable fracturing. The external pressure was changed from 2500 psi to 1 atmosphere in approximately 25 milliseconds after steady state conditions at 2500 psi had been achieved in the microsphere. The microsphere temperature was 1300° F.

The latest melt initiation curves or "melt envelopes" which have been generated by Sandia, Martin and G.E., are compared in Figure II-3. It is concluded, based on the thermal studies performed and covering various assumptions such as spinning vs. non-spinning, etc., that there is a band of release conditions for which melting initiates. Above this band, no melting would occur on release; below the band some melting would result. However, more confidence should be placed in the Sandia melt curves as the analysis is more exact and accounts for internal conduction.

It should be noted that no dynamic tumbling analysis of a microsphere, or cloud of microspheres, has been performed to justify spinning or non-spinning at the point of release from a thermally-destroyed fuel capsule. In reality, some mode of rotating between fully stable and complete random spinning could occur.

Sandia has generated curves of constant percentages of mass melted in terms of altitude vs. velocity release conditions, and this is illustrated in Figure II-4 for 50% and 100% melting and compared with the melt initiation curve. However, little confidence can be placed in the 50% and 100% melting curves due to the assumptions made in the thermal model.

Before proceeding further into a more detailed discussion of the various thermal studies, a few general remarks pertaining to the problem of microsphere melting and ablation should be made to get perspective. First, the existing theoretical studies can fairly accurately predict the point at which melting initiates, if spinning vs. non-spinning is defined, as the solid state (no deformation) heat flow is well approximated by the finite difference solutions to the Fourier conduction equation. However, once melting initiates the liquid mass flow by ablation and decelerating body forces will redistribute the heat from the stagnation point to the cooler sphere sides and back; partial or total resolidification is possible with a large change in shape resulting. This phenomenon has not been duplicated in the Sandia arc tunnel experiments (Reference II-10), as discussed in Section II-F, but some bulging on the sides has been observed in the Sandia experiments. More bulging would most likely

FIGURE II-4. PARTIAL MELTING CURVES FOR 2 SPHERE SIZES
(REFERENCE II-10)

occur in the real flight environment with an associated increase in particle drag which could be significant in reducing heating. An approximate solution to the heat transfer of a liquid layer on a solid core was made by G.E. (Reference II-3), but is only valid near the stagnation point and does not cover the region of interest, which is the entire surface of the sphere. A review of the literature indicated there are no valid theories and very little experimental data to describe the thermal behavior of microspheres once the front face begins to melt and ablate. Thus the mass loss mechanisms during the partially molten stage cannot be accurately described by any existing theory since the particle deformation is not yet known; all existing mass loss theories in the partially molten state make basic and restrictive assumptions on the shape and thickness of the liquid layer (it must be very thin).

1. Latest Thermal and Physical Properties of PuO_2

A brief summary of the latest available thermal properties of PuO_2 will be presented to (1) list the values used in the thermal analyses performed by past investigators and (2) point out where values for thermal properties are lacking and require the performance of additional experiments and/or analyses to obtain them. The latest tabulation of a wide range of properties for $^{238}\text{PuO}_2$ is given in Reference II-11 and the thermo-physical data will be summarized below in Table II-2.

TABLE II-2
THERMAL PROPERTIES OF $^{238}\text{PuO}_2$

a. Specific Power	0.40 w/g for 80% ^{238}Pu density
b. Bulk Density	~ 6.7 g/cc and varies with individual microsphere depending on void space
c. Thermal Conductivity	See Figure II-5. Values have been determined up to approximately 3300°R by thermal diffusivity experiments on $^{239}\text{PuO}_2$ (References II-11 and II-12). No experimental values on the liquid material are anticipated in the near future.
d. Specific Heat	See Figure II-5. Values have been determined up to approximately 2900°R (Reference II-12).

(continued)

TABLE II-2 (continued)

e. Melting Temperature	$>2150 \pm 80^\circ \text{ C.}^*$
f. Heat of Fusion	101 BTU/lb.
g. Total Hemispherical Emittance	Not measured. Values used in Sandia calculations varied from 0.55-0.75.

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* Unpublished Mound Laboratory data.

The most significant lack of data is for the emittance. Experiments at Mound Laboratory are planned for the latter part of 1968 to obtain emittance for PuO_2 (Reference II-12). A parametric study to account for variations in emittance between 0.55 and 0.75 was made by Sandia in Reference II-8 and is presented as Table II-3. The peak temperatures attained were calculated for the two values of emittance for various points of release. The differences in peak temperature are reasonably small ($<300^\circ \text{ F}$); however, the emittance has not been experimentally determined to be within the 0.55-0.75 band, and the errors in peak temperature could be somewhat larger if the value for emittance is outside the 0.55-0.75 band. In general, the effect of emissivity for the lower altitude releases will not be significant in determining the initiation of melting.

The thermal conductivity and specific heat data is limited to roughly 1500° F . below the melting temperature of PuO_2 . However, as can be seen in Figure II-5, the curves are fairly flat, and values can be extrapolated to the melt temperature. As the material melts, there is no knowledge as to how thermal conductivity and specific heat change. This could become significant for the heat transfer during the partially molten state; it has been assumed in past analyses, that the thermal conductivity and specific heat remain constant at the values below the melting point after the phase change.

Since the problem involved in analyzing the heat transfer to the microspheres once melting begins is complicated by the fact that the flow deformation is not known, an accurate value for thermal conductivity would be insufficient in describing the melting behavior in the partially molten state. If more theoretical work is to be done, the theories must be refined to account for this phenomenon. Since the experimental approach, such as the NOL light gas gun experiments, appears more reasonable, the time and effort involved in determining the properties of molten PuO_2 may not be justified at present.

FIGURE II-5 Thermal Properties - PuO_2

TABLE II-3

PEAK TEMPERATURE OF P_uO_2 MICROSPHERES WITH
AN EMITTANCE OF 0.75 COMPARED TO THOSE
WITH AN EMITTANCE OF 0.55

DIAMETER (microns)	RELEASE ALTITUDE (kft)	EMITTANCE	RELEASE VELOCITY (ft/sec)	PEAK TEMP. (°R)	TIME TO PEAK TEMP. (Sec.)
50	250	0.55	24,000	3841	0.08
50	250	0.75	24,000	3583	0.08
50	210	0.55	21,000	4653	0.025
50	210	0.75	21,000	4506	0.025
50	190	0.55	19,050	4745	0.018
50	190	0.75	19,050	4638	0.019
50	160	0.55	14,950	4438	0.018
50	160	0.75	14,950	4279	0.018
70	250	0.55	24,000	3847	0.12
70	250	0.75	24,000	3585	0.10
70	210	0.55	21,000	4636	0.04
70	210	0.75	21,000	4501	0.04
70	190	0.55	19,050	4727	0.025
70	190	0.75	19,050	4603	0.03
70	160	0.55	14,950	4344	0.03
70	160	0.75	14,950	4186	0.025

* Note: Initial temperature = 1000°R.

2. Analysis Techniques Used by Various Investigators

A review of the assumptions and methods of solution for the temperature response of microspheres as performed by Sandia, Martin, Aero-therm and Sandia was made during the course of this study. As illustrated by Figure II-3, the melt initiation curves agree reasonably well with each other. However, when a comparison of the temperature response at the stagnation point is made for the same particle size and release condition, large differences exist between the various analyses as illustrated in Figures II-6 and II-7. The conditions selected for comparison are release at 244,000 ft., $V = 25,200$ ft./sec and 181,000 ft., $V = 21,000$ ft./sec. and diameter = 250μ . Unfortunately, a Martin analysis was not made, for these conditions, and could not be used for comparison. It is apparent, in comparing Figures II-6 and II-7 with Figure II-3, that differences in temperature

response with time do not yield significantly different melt initiation curves.

A logical explanation of this phenomenon could be due to the fact that different methods in calculating the aerodynamic heating rates and in constructing the exact thermal model are small compared to the changes in atmospheric density (and resulting heating rates) with changes in altitude, at the same velocity, between the various melt curves. For example, the average difference between the Martin and Sandia envelopes for stable spheres is approximately 10,000-20,000 ft. If free molecular flow is assumed, the cold-wall heating rate is:

$$\dot{q}_s = \frac{\beta \rho V^3}{2gJ}$$

V = velocity, ft./sec.

g = acceleration of gravity

J = mechanical equivalent of heat - 778 ft.-lb./BTU

β = accommodation coefficient

Although the flow for most release conditions is not completely free molecular, the above equation is approximately correct. Thus, the aerodynamic heating rates are roughly proportional to the atmospheric density at release. Using the 1962 Standard Atmosphere Tables, the density at an altitude of 240,000 ft. and 255,000 ft. are given below:

$$h = 240,000; \quad \rho = 3.535 \times 10^{-6} \text{ lb./ft.}^3$$

$$h = 255,000 \quad \rho = 1.792 \times 10^{-6} \text{ lb./ft.}^3$$

For an altitude change of only 15,000 ft. (at the same flight velocity), the density ratio is $3.535/1.792 = 1.96$. Thus, the heating rates are nearly doubled if one releases at 240,000 ft. vs. 255,000 ft. It is likely that the large change in density with altitude may account for having significant differences in time vs. temperature between the various analyses as seen in Figures II-6 and II-7 while having the melt initiation curves appear reasonably close in altitude vs. velocity coordinates.

It must be again kept in mind that all of the thermal studies only predict the initiation of melting. Once melting begins and the liquid PuO_2 starts to flow, the temperature calculations can be quite inaccurate. (This is discussed in Section II-D) The theoretical melting rates predicted by the various analyses, such as the

SPHERE 2 code, are quite optimistic. The actual melting rates should be considerably lower when the effects of vaporization, liquid flow and chemical reactions are accounted for.

A brief review of the various thermal models is given below.

Sandia SPHERE 2 Code Analysis

The SPHERE 2 Code analysis (Reference II-5) computes the deceleration trajectory at the point of insertion into the atmosphere, the resulting aerodynamic heating rates for the continuum, free molecular and transition flow regimes and the resulting temperature response at various stations in the sphere. A stable orientation with respect to the flow is assumed. This code was used to calculate the melt initiation curve in Figure II-3 and the curves of 50% and 100% melting of Figure II-4. The spherical model is divided into 40 nodal segments. Vaporization, and the reduction in aerodynamic heating due to vaporization, were accounted for by utilizing the technique of Chapman and Matting (Reference II-13). The approximate bridging technique as devised by Chapman, also from Reference II-13, was used to predict the aerodynamic heating rates in the transition flow regime between continuum and free molecular flow.

Martin Thermal Analysis

Two models to calculate the thermal response of a microsphere upon release have been generated by Martin (Reference II-6). The first analysis, or "equilibrium model," neglected the heat capacity. It was found that the particle temperatures calculated using this model were much too conservative when compared with an accurate thermal model where conduction is considered. Therefore, in the second phase the "non-equilibrium model", the particle heat capacity was considered and the thermal conduction term neglected. The basic assumptions of the two models are given below. The non-equilibrium model was used to generate the melting initiation curve in Figure II-3.

Basic Conditions

a. Equilibrium model

- (1) The particle is a stable, non-rotating sphere throughout the entry.
- (2) The flow around the particle may be free molecular, transitional or continuum.

- (3) Heat of fusion and the particle heat capacity are negligible compared to the heat of vaporization and radiation. (The assumption of negligible heat capacity reduces the thermal problem to a quasi-steady on.)
- (4) Internal heat generation due to nuclear disintegration is considered.
- (5) Heat blockage due to vaporization is accounted for.
- (6) Atmospheric quantities are given by the 1962 U. S. Standard Atmosphere.
- (7) There may be a temperature difference between the front and rear surfaces of the particle. For purposes of calculation of this heat conduction effect, the body is treated as a slab of thickness equal to the radius.

b. Non-equilibrium model

- (1) Same as Conditions 1 to 2.
- (2) The temperature difference between the front and the rear of the particle is negligible.
- (3) Heat of fusion, particle heat capacity, heat of vaporization, internal heat generation and radiation are considered.
- (4) Atmospheric quantities are given by the 1962 U. S. Standard Atmosphere.

Aerotherm Thermal Analyses

The thermal analysis performed in Reference II-4 utilized the Aerotherm arbitrary transient temperature prediction computer code, which incorporates an explicit finite difference solution to the heat conduction equation for general axisymmetric shapes. The general assumptions for the microsphere model are:

- (1) The material properties, such as thermal conductivity, specific heat and density, are input as a function of temperature.
- (2) The particles are and remain spherical; no transport of molten material is considered.
- (3) The production of gaseous chemical species at the material-atmosphere interface is zero.

A 12-node mesh was constructed to simulate one-half of a spherical particle. Aerotherm did not calculate a melt initiation curve as this was not contractually required. Temperature calculations were made for four release points as specified by Sandia. A summary of the times to melt at the front (stagnation point) for the four release points and for two diameters and for total melting are given in Table II-4.

While the Aerotherm 12-node model is not as accurate as the 40-node Sandia SPHERE 2 Model, the temperature vs. time response at the stagnation point agrees reasonably well for one release point that was checked - a 250 μ particle released at 24,000 ft. and velocity of 25,200 ft./sec.

TABLE II-4

PREDICTED MELTING TIMES

AEROTHERM MODEL

RE-ENTRY CONDITIONS

Material	Injection altitude feet	Injection velocity ft./sec.	Particle size microns	Time to melt seconds	
				front face	rear face
PuO ₂	244,000	25,200	250	.125	.425
PuO ₂	244,000	25,200	50	.0155	.0275
PuO ₂	200,000	16,500	250	.12	---
PuO ₂	200,000	16,500	50	.0085	.0195
PuO ₂	181,000	21,000	250	.010	.075
PuO ₂	181,000	21,000	50	.0015	.0045
PuO ₂	118,000	12,000	250	---	---
PuO ₂	118,000	12,000	50	.0012	---

General Electric Analysis

The basic assumptions are:

- (1) The two-degrees of freedom trajectory was used to represent the motion of the released particle.
- (2) The effects of a variable drag coefficient are continuously determined throughout the trajectory.
- (3) Vaporization and melting mass loss mechanisms are not considered.
- (4) The aerodynamic heating rates to the particle are based on work done in Reference II-14 and II-15 to include heat transfer in free molecular, near-free molecular, transition and low Reynolds number, flow regimes.
- (5) The net heat conducted into the particle causes a uniform temperature change throughout the particle. A spin factor (ϕ) of 0.218 was utilized to obtain the surface averaged heating rates.

The details of the thermal model, other than the basic assumptions above are not given in Reference II-3. On page 47 of Reference II-3 reference is made to more detailed analysis where conduction effects were accounted for but found to be negligible. This is illustrated in Figures 26 and 29 of the same report. However, no details of the model to obtain results in Figures 26 and 29 are given. It also appears that the temperature response for the more refined analysis is much slower in time than the one-dimensional analysis as seen in both figures. The melt initiation curve in Figure II-2 of this report was computed with the simpler model.

3. Conclusions

- (a) The assumption of spinning vs. non-spinning gives two values for a melt initiation curve as seen in Figures II-2 and II-3.
- (b) A collation of melt initiation curves from Sandia, Martin and G.E., is given in Figure II-3. Since spinning or non-spinning cannot be defined for a real atmospheric release, Figure II-3 gives a "band" of altitude vs. velocity coordinates for which some melting will occur.
- (c) While there are some apparent differences in the thermal analysis as seen in Figures II-6 and II-7 the melt initiation curves are reasonably close.

- (d) Thermal properties for molten PuO_2 are nonexistent.
- (e) No value for total emittance is available for PuO_2 microspheres.
- (f) Once melting initiates at the stagnation point, all of the thermal models are grossly inaccurate. In general, the actual microspheres should melt at a slower rate than the theoretical models.

C. STABILITY AND/OR MASS LOSS OF PARTIALLY AND FULLY MOLTEN PARTICLES

As discussed in Section II-B, the first question to be answered by analysis and experiment is: "Is there melting for actual fuel release points?" The second question to be answered is: "Does this melting result in mass loss due to removal of some of the liquid mass by any physical mechanism?" As illustrated by Figure II-3, there will be some melting for most release points; therefore the technical investigator must address himself to the second question to evaluate the survival of microspheres during re-entry.

The sequence of physical events at release, if one assumes a non-spinning microsphere are:

1. Aerodynamic heating gives a rapid temperature rise in the sphere. Since the heating rates are very high for the higher density (lower altitude) releases, the stagnation point reaches the melt temperature well before the remainder of the particle. This effect is more pronounced on the larger microspheres ($D \sim 200\mu$); however, the larger spheres are more critical from a molten state breakup standpoint.
2. The front face of the microsphere continues to melt while the remainder of the particle is solid. During this time the liquid material will be dragged and pushed away from the stagnation point by the shear and pressure forces into the flow field. As more material melts, the particle, with the additional effect of deceleration forces, will become distorted from a spherical shape. Solidification of some of the molten PuO_2 is possible on the sides.
3. As the particle continues to receive aerodynamic heating, it may become fully molten. In general, the behavior of fully molten particles is better understood in terms of breakup than semi-molten particles.
4. The particle continues to heat up by aerodynamic heating. Either it breaks up due to aerodynamic and deceleration forces or it remains intact and eventually resolidifies, decelerates further and

falls to the ground intact.

As also discussed in Section II-B and will be discussed in Section II-D, the present theoretical thermal analyses are inaccurate once the stagnation point of a microsphere starts to melt. The processes occurring during Step 2 above are only known qualitatively at present for the real flight conditions. Thus, it is difficult to assess the actual conditions, in terms of altitude and velocity at release, for which complete melting will take place. The approach taken to date was to calculate the conditions, based on the so-called critical Weber number, for which the particle would remain whole. This concept is based on a large number of experiments indicating that a particular material in the molten state will not break up if the Flight Weber number is less than the critical value. However, this critical value is in fact not one number, but a wide band which makes the concept only marginally useful for real flight analysis.

In the partially molten state, however, the effects of deceleration and aerodynamic forces (Weber No.) are much less known, both quantitatively and qualitatively. The same critical parameters that have been used for fully molten particles have been assumed to apply for this problem, such as the Weber number, Froude number, Bond number, Ohnesorge number and Reynolds number. (See Table II-6). It is not yet clear which of the parameters, or combinations of parameters, fully describe the partially molten breakup or mass loss.

An estimate of the real time that the microsphere is in the partially molten state is given by Reference II-4 and is listed in Table II-5.

TABLE II-5
PREDICTED MELTING TIMES (Reference II-4)

Material	Injection Altitude (feet)	Injection Velocity (ft./sec.)	Particle Size (microns)	Time to melt (seconds)		Thermal Stopping Time (sec.)
				front face	rear face	
PuO ₂	244,000	25,200	250	.125	.425	3.7
PuO ₂	244,000	25,200	50	.0155	.0275	0.75
PuO ₂	200,000	16,500	250	.12	--	0.75
PuO ₂	200,000	16,500	50	.0085	.0195	0.13
PuO ₂	181,000	21,000	250	.010	.075	0.6
PuO ₂	181,000	21,000	50	.0015	.0045	0.1
PuO ₂	118,000	12,000	250	--	--	0.04
PuO ₂	118,000	12,000	50	.0012	--	0.01

TABLE II-6

VARIOUS NON-DIMENSIONAL PARAMETERS
TO DESCRIBE LIQUID DROPLET BREAKUP

$$\text{Froude Number} = \frac{V^2}{Da} = \frac{\text{Aerodynamic Forces}}{\text{Acceleration Forces}}$$

$$\text{Weber Number} = \frac{\rho_g V^2 D}{2 \sigma} = \frac{\text{Aerodynamic Forces}}{\text{Surface Tension Forces}}$$

$$\text{Reynolds Number} = \frac{\rho_g V D}{\mu_g} = \frac{\text{Aerodynamic Forces}}{\text{Viscous Forces}}$$

$$\text{Ohnesorge Number} = \left[\frac{\mu_l^2}{\rho_l D \sigma} \right]^{1/2} = \frac{\text{Viscous Forces}}{\text{Inertial Forces} \times \text{Surface Tension Forces}}$$

$$\text{Bond Number} = \frac{\rho_l D^2 a}{\sigma} = \frac{\text{Acceleration Forces}}{\text{Surface Tension Forces}}$$

$$\text{Capillary Number} = \frac{\mu V}{\sigma} = \frac{\text{Viscosity Forces}}{\text{Surface Tension Forces}}$$

D = sphere diameter

V = Velocity

ρ_g = gas density

σ = surface tension

μ_g = gas coefficient of viscosity

μ_l = liquid coefficient of viscosity

ρ_l = liquid density

a = acceleration

The time difference from front face to rear face melting can be compared with the so-called "thermal stopping time" at which point the heat flux is offset by the surface radiation, and no more melting takes place.

It is apparent from Table II-5 that for many flight conditions, both the partially molten and fully molten situations must be analyzed. Generally, two mass loss criteria--one for fully molten and one for partially molten spheres--must be satisfied for atmospheric release conditions where significant melting results.

1. Fully Molten Particle Stability

To date, various investigators have discovered, via experiments, four rather well-defined breakup modes. (Reference II-16). They are, in order of increasing relative velocity:

- a. Vibrational or rotational breakup
- b. Bag breakup
- c. Shear or stripping breakup
- d. Chaotic breakup

The vibrational form of breakup occurs when the drop is flattened by the gas stream and oscillation develops at the drop's natural frequency. The natural frequency as given by Renolds, et al (Reference II-4) is:

$$f = \frac{1}{2\pi} \sqrt{\frac{8\sigma}{\rho_l r^3}} \quad \text{seconds}^{-1} \quad (II-29)$$

where

σ = surface tension

ρ_l = liquid density

r = drop radius

It is believed that this mode of breakup can occur when the relative velocity (and corresponding Weber number) is slightly above some critical value to cause instability. For PuO_2 , the period of oscillation is given as (Reference II-4),

$$T = 1.0128 \times 10^{-7} \sqrt{D^3}$$

D = particle diameter in microns

For a 200μ diameter particle, the period is approximately 2×10^{-4} seconds. The time for breakup to occur by the vibrational mode (Reference II-4) is approximately 3 times the period of oscillation

or roughly 6×10^{-4} seconds for a 200μ PuO_2 microsphere. Thus, there is sufficient time for most release cases for the vibrational breakup to occur if the fully molten particle becomes unstable.

The second mode of breakup is bag breakup and has been observed during experiments in both steady and transient flow fields. The gas stream increasingly flattens the drop as in the vibrational breakup. Instead of oscillating, the center portion of the drop's front surface first becomes concave and then is blown out downstream to form a hollow bag attached to a rim. The bag finally bursts, producing many fine drops; later, the heavy rim divides into much larger drops, which could constitute roughly 75 per cent of the liquid originally in the drop. In the presence of a high deceleration body force, this type of breakup will occur at a higher relative velocity than it would if no deceleration was involved.

At still higher relative velocities, the third mode of breakup is observed. In this case the drop deforms in a manner nearly opposite to that of bag breakup; that is, the drop flattens on the rear side and becomes convex on the front or upstream side (Reference II-16). Then, after a period of time, the drop loses mass from the upstream surface either in the form of thin sheets, filaments, or droplets depending on the liquid properties and relative velocity. This form of drop breakup, termed shearing or stripping breakup, has been observed only during transient flow experiments (Reference II-16). The qualitative physical interpretation of this phenomenon is fairly clear. Motion of the liquid on and near the drop's front surface is caused by shear forces resulting from gas flowing over the liquid surface. When the shearing force is slightly larger than the resisting force of surface tension, cylindrical sheets of fluid are removed from the drop at the downstream edge of the convex surface. Subsequently ligaments divide into small drops. For greater aerodynamic shear, the initial deformation process is unchanged, but mass removal is accomplished by wave crest stripping. It is well known that a gas stream flowing over a liquid surface will contribute to the generation of surface waves and that the higher the relative velocity between the gas and the liquid, the more pronounced is this effect. An increase in the size of the disturbance results finally in detachment of liquid from the surface and ultimately in disruption of the drop.

At extremely high relative velocities, the chaotic breakup has been observed. The drop, when suddenly exposed to a flow of high aerodynamic pressure, shatters into a cloud of very small particles. It is questionable whether this type of breakup could occur for the re-entry release of PuO_2 microspheres since the molten material could be stripped off as soon as melting begins at the stagnation point before a fully molten particle was formed. If it could occur, it would be due to a relatively low altitude, high velocity release.

A thorough survey of experimental and theoretical data by Reference II-16 has concluded that the Weber number ($\rho U^2 D / \sigma$) is the best non-dimensional parameter to express the breakup of a liquid drop. A review of the various non-dimensional parameters is given in Table II-6. The values of Weber number for which breakup is possible have been primarily obtained experimentally and range from approximately 1.5 to 25. It is also concluded in Reference II-16 that, based on the relatively wide spread of experimental data for critical Weber number, a single value for all conditions may not exist.

Much of the experimental data has been for subsonic flow. It has been argued in Reference II-17 that the aerodynamic shear forces in supersonic and hypersonic flow are phased differently with respect to the pressure forces; therefore, the breakup in hypersonic flow could be different than breakup in subsonic flow at the same Weber number. A relatively small amount of experimental work has been performed in hypersonic flow. Two theoretical studies have been performed by Aerotherm Corporation (Reference II-4) and G.E. (Reference II-3) to obtain an instability/breakup parameter in hypersonic flow. The parameter obtained to correlate breakup in all cases was the Weber number. The critical Weber numbers from the various sources both experimental and theoretical are listed in Table II-7.

TABLE II-7

CORRELATION OF APPROXIMATE VALUES OF CRITICAL WEBER NO.
OBTAINED FROM VARIOUS SOURCES FOR FULLY
MOLTEN PARTICLES IN HYPERSONIC FLOW

<u>Reference</u>	<u>Method Obtained</u>	<u>We CRIT</u>
Aerotherm Corp.	Analysis	10
General Electric	Analysis	2
Haas (CAL)	Experiment	3.3
Beal (CAL)	Experiment	~ 6

a. Theoretical Studies

The theoretical approach by G.E. considers the distortion of a hypothetical cylindrical plug in the presence of pressure, deceleration, viscous and surface tension forces. A differential equation expressing deformation velocity is written combining all terms (top of page 42 in Reference II-3):

$$\frac{dV}{dt} = \frac{3}{2\rho_1 D} \left[\rho_\infty U^2 - \frac{8\sigma}{D} - \frac{16\mu V}{D} \right] + G$$

V = distortion velocity

ρ_1 = material density

D = particle diameter

U = free stream velocity

ρ_∞ = free stream density

σ = surface tension

μ = coefficient of viscosity

G = aerodynamic deceleration (ft./sec.²)

This equation, when integrated twice, gives the deformation. The critical Weber number is obtained by solving for the flow conditions at which the deformation starts to increase with time; i.e., become unstable. The breakup, according to this theory, is time dependent, and the approximate critical value of 2 for W_e in free molecular flow is the value for which this instability initiates. The analysis does not account for aerodynamic shear forces that the actual spherical particle would see due to the assumption of a fictitious cylindrical model. In general, the agreement with experimental data is reasonable. It should be pointed out that this analysis would be expected more to give an order of magnitude value rather than an absolute one.

The theoretical study by Aerotherm considers a spherical model with the following assumptions:

- (1) Constant surface tension
- (2) Uniform liquid density
- (3) Zero surface shear.
- (4) No viscous forces are considered

The external pressure field is non-uniform but axisymmetric. The model is similar in approach to the G.E. model in that deceleration causes breakup but it more closely approximates the actual sphere in shape. Viscous forces are not included (although G.E. has concluded that viscosity has no effect on the critical value). The main difference between the G.E. and Aerotherm analyses is that the G.E. analysis predicts the time-dependent conditions for which the

deformation becomes unstable while the Aerotherm analysis predicts the change in shape in a steady-state flow field and deceleration field. The limiting condition in the Aerotherm analysis occurs when the deformed shape cannot remain whole due to surface tension. The approximate value of critical Weber number (10) derived by Aerotherm agrees moderately well with experimental data (Reference II-16) but is a factor of 5 higher than the G.E. value.

Based on a review of the two theoretical analyses, it is clear that the values for critical Weber number calculated are order of magnitude numbers. More confidence should be expressed in the experimental data for hypersonic flow listed in Table II-7 than in the analytically determined values.

b. Hypersonic Droplet Breakup Experiments

The experiment at Cornell Aeronautical Laboratory (CAL) on droplet breakup were performed on zirconium droplets formed by passing electrical current through a thin zirconium wire (Reference II-18). It was found that by passing current at roughly 30 volts, through the wire in a vacuum, the molten wire exploded into approximately 15-20 drops of fairly uniform size.

A shock tunnel was then triggered to pass hypersonic air over the molten particles. Breakup was recorded photographically. The calculated Weber number based on the shock tunnel conditions was 7.25. Some of the drops broke up while others did not, drop distortion was not great, and the number of new drops formed was small. All of these observations indicate that the critical condition was just slightly exceeded. Beal then concluded that the critical Weber number was close to six.

Another set of experiments at CAL was performed by melting a zirconium wire in a plasma jet tunnel (See Figure II-8). As the end of the rod melted, a drop of the metal was carried downstream and examined photographically. Unfortunately, the plasma jet conditions, unlike the previously discussed shock-tunnel conditions, were not well defined. Therefore, the conclusions of the arc-tunnel tests were that the breakup process was only qualitatively the same as for the shock-tunnel experiments. A third set of experiments on Fe_2O_3 spheres, primarily to obtain data for partially molten particles, concluded that the critical Weber number for fully molten particles was near 6.

The critical value of Weber number that CAL has found via several experiments for hypersonic, continuum flow is near six. The theoretical model used by G.E. (Reference II-3) has indicated that the critical value is strongly dependent on the drag coefficient (C_D), which goes from approximately 2 in free molecular flow to .9 in continuum, hypersonic flow. The value of

FIGURE II-8 PLASMA JET TEST APPARATUS FOR MELTING ABLATION
(NOT TO SCALE)

W_{eCRIT} in continuum flow, with the G.E. model, is 2.67 as opposed to 2 in free molecular flow. Utilizing this argument, the CAL value of $W_{eCRIT} = 6$ could be closer to 4 by ratioing the G.E. theoretical values in free molecular flow. However, this has not been established experimentally.

It was also concluded at CAL that the critical value is not a highly precise number. It has been observed that droplets at the same size and same Weber number slightly above the critical value behave differently; i.e., some breakup while others do not.

c. Application to Flight Conditions

Based on the experiments performed at CAL on zirconium and Fe_2O_3 droplets, a critical Weber number in the range of 4-6 is felt to be reasonable. A plot of the lines corresponding to breakup and/or mass loss for both $W_{eCrit} = 4$ and 6 and diameter of 100 μm and 200 μm is presented in Figure II-9. Also plotted is the Sandia melt initiation curve. As discussed in Section II-B, the theoretical curve corresponding to complete melting, Figure II-4, is grossly inaccurate and complete melting cannot be predicted. The actual area in Figure II-9 corresponding to complete melting but no breakup cannot be accurately defined. The only positive statement that can be made is that if the critical value of W_e is in the range of 4-6, no breakup for fully molten droplets would occur below the lines shown in Figure II-9.

d. Character of Debris Resulting from Particle Breakup

Beal (Reference II-18) has reported some data on the breakup of zirconium droplets in hypersonic flow. The initial droplet diameter was 500 μm , and the test conditions are:

$$M_{\infty} = 7.48$$

$$\rho_{\infty} = 40.9 \times 10^{-6} \text{ slugs/ft.}^3$$

$$U_{\infty} = 8,061 \text{ ft./sec.}$$

$$\text{Simulated altitude} = 95,000 \text{ ft.}$$

The resulting debris impacted on a lead target, and the crater sizes were measured. It was found that the drop diameter is approximately one-half the crater diameter, although this had not been accurately determined. The weight fraction of the debris as a function of crater diameter is plotted in Figure II-10. The droplet diameter was assumed to be one-half the crater diameter. The peak in debris size appears to be approximately 120 μm or 24% of the initial diameter. It was expressed by Beal that, in general, the peak in diameter would go down as the Weber number conditions are increased; i.e., the particle would head toward chaotic breakup with a large number of very small particles. The data in Figure II-10 is for a droplet slightly in excess of the critical value of Weber number.

FIGURE II-10 WEIGHT DISTRIBUTION BY CRATER SIZE
CAL EXPERIMENTS

e. Conclusions

- (1) For some flight conditions, total melting of PuO_2 microspheres could result although this cannot be theoretically predicted in terms of flight conditions. Breakup determined by the fluid mechanics of fully molten drops is possible.
- (2) There are four observed mechanisms of droplet breakup. The actual physical mechanisms occurring are, in general, quite complex.
- (3) The best correlation for droplet breakup is the Weber number.
- (4) There is not as much experimental data on supersonic flow breakup as there is for subsonic flow. There is evidence to suggest the breakup in supersonic flow could occur at a higher Weber number than in subsonic flow (Reference II-4).
- (5) A reasonable estimate based on past experiments and analyses at the critical Weber number for molten droplets in hypersonic free molecular flow is in the 4 to 6 range.
- (6) The breakup area for fully molten particles is shown in Figure II-9 as a function of size--100 μ and 200 μ diameters--and for Weber numbers of 4 and 6.

2. Partially Molten Particle Stability

As discussed earlier, the behavior of partially molten droplets is not as well understood as the breakup of fully molten droplets. Complicating this even further is the fact that a PuO_2 microsphere, upon release, starts to melt on its front face if no rotation takes place and that appreciable flow of the liquid is involved. There has been some theoretical analysis done on this problem (References II-3 and II-17) and two experiments that are pertinent--the CAL shock tunnel experiments of Fe_2O_3 spheres and the Sandia hyperthermal arc tunnel experiments of $^{238}\text{PuO}_2$ microspheres. In addition, the work done by Chapman on earth entry tektites (Reference II-19) is applicable.

A collation of results of the two experimental programs and the theoretical studies are, in general, conflicting. The parameter selected for comparison is the Weber number, although Reynolds in his analysis (Reference II-17) has shown that the critical Weber number for a thin layer is highly dependent on viscosity. The G.E. analysis, however, shows that the critical value is independent of viscosity (Reference II-3) although the mechanisms of instability are different in the two studies. Since the effect of viscosity is not yet clear on stability, the Weber number remains the only approximate way to predict breakup.

A summary of the approximate critical Weber numbers determined for partially molten spheres is shown in Table II-8.

TABLE II-8
APPROXIMATE CRITICAL WEBER NUMBERS AS DETERMINED
EXPERIMENTALLY AND THEORETICALLY FOR
PARTIALLY MOLTEN SPHERES

<u>Source</u>	<u>We_{CRIT}</u>	<u>Method Obtained</u>
Stanford University	0.15-0.20* PuO ₂ micro- spheres	Analysis
General Electric	4.18	Analysis
Beal - CAL	~10	Experiment - Fe ₂ O ₃ spheres
Sandia [See Section II-F]	~ 4.6**	Experiment - 238PuO ₂ microspheres
Chapman - Ames	>10	Experiment

* The Stanford analysis assumes that waves travel to the back of the sphere through a thin layer. As discussed in the next section, this is not feasible with a non-spinning sphere in a deceleration field. In general, this value is inconsistent with the experimental data and is only included for completeness.

** Sandia has not determined a critical value for Weber number. They have observed some mass loss at values close to 1, although stripping or breakup is not evident until $We = 4.6$.

In addition, Chapman (Reference II-19) has stated qualitatively that observations of tektites that have survived re-entry indicates that breakup did not occur at a Weber number where breakup would result for a molten drop. His conclusion is that partially molten spheres are more stable than fully molten drops.

a. Application of Hydrodynamic Instability Analysis to PuO₂ Microspheres

In the absence of an external gas flow, waves on the surface of a liquid layer can be amplified by a body force field (atmospheric deceleration) directed away from the liquid. This is the instability usually attributed to Taylor (Reference II-20). Without

surface tension, such a system is inherently unstable to wave disturbances of all wave numbers; surface tension stabilizes waves above a certain "cut-off" wave number, α_c , and viscosity reduces the rate of amplification of the unstable modes. The analysis performed by Liner (Reference II-3) considers this Taylor type instability acting on a thin liquid layer on a solid particle near the stagnation point.

The presence of aerodynamic flow around the sphere has two effects. First, a shear flow is established in the liquid film through the shear at the surface. Second, the appearance of waves on the surface can now lead to alterations of the forces which the gas exerts on the film; these effects provide additional instability mechanisms of which the most important is the Kelvin-Helmholtz (K-H) instability. The analysis performed by Reynolds (Reference II-17) deals with the instability due to aerodynamic shear and is a derivation of the K-H mechanism. The analysis is based on studies done by Craik (Reference II-21) and Benjamin (Reference II-22) and is referred to by Reynolds as the C-B instability mechanism. Craik's analysis revealed the fact that very thin liquid films are more unstable than thick layers under some circumstances. This situation arises when the external flow exerts a disturbance shear stress on the interface in phase with the wave slope.

Thus, the two theoretical studies as performed by Liner of G.E. and Reynolds of Stanford University to predict instability of partially molten PuO_2 microspheres assume two different phenomena and, not surprisingly, show a different value, for critical Weber number. However, it must be remembered that the assumptions in both analyses are quite restrictive and are expected more to show order of magnitude at the very best values and the relative effect of the two different stability mechanisms. A refined analysis would consider both effects, although Reynolds has shown that shear instability occurs at a much lower W_e for small spheres in free molecular flow.

b. The Character of Reynolds/Aerotherm Stability Theory and its Significance to Real Microsphere Flight

The analysis considers the hydrodynamic stability of a liquid layer on a solid body in the presence of a hypersonic free molecule external flow. Since the basic equations are written in two dimensional cartesian coordinates, curvature effects have been neglected, which implies that the liquid layer has been assumed to be small relative to the dimensions of the body, although this is not expressly stated in the report.

Specifically, the problem has been defined by Reynolds as:

If the surface of the liquid is given a small displacement, is this disturbance damped or amplified as time goes on?

In order to answer this question, Reynolds has chosen to use a perturbation method and this requires knowledge of the undisturbed velocity in the liquid layer. In this analysis, it has been assumed that the undisturbed liquid layer velocity varies linearly from zero at the body surface to some value at the gas-liquid interface. The surface disturbance has been assumed to be of the form:

$$\eta(x,t) = \hat{\eta} \exp [i\alpha (x-ct)] \quad (\text{II-30})$$

$$\text{where } c = c_r + ic_i$$

and α is the disturbance wave number which may be specified

The problem is to find out whether C_i is positive (amplification) or negative (damping) under the following conditions:

1. The surface displacement is consistent with the perturbation velocity field.
2. There is a balance among the gas phase normal stress, liquid phase normal stress, and surface tension at the liquid surface.
3. There is no discontinuity of the tangential shear stress across the liquid-gas interface.
4. The perturbation velocities are zero at the solid surface. The perturbation stream function was assumed to be of the form:

$$\psi(x,y,t) = \phi(y) \exp [i\alpha(x-ct)] \quad (\text{II-31})$$

where the perturbation velocities are given by,

$$u = - \frac{\partial \psi}{\partial y} \qquad v = \frac{\partial \psi}{\partial x}$$

Substitution of II-31 into the Navier-Stokes equations results in the well known Orr-Sommerfeld equation for ϕ

$$\{ (D^2 - \alpha^2) - i\alpha R [(\bar{u} - c) (D^2 - \alpha^2) - D^2 \bar{u}] \} \phi = 0 \quad (\text{II-32})$$

where $R = U_s \delta / \nu$ Liquid Reynolds Number

δ = liquid layer thickness

$\bar{u}$ = y Mean velocity distribution in liquid layer

The velocity profile in the liquid layer was obtained by assuming that the axial component of the deceleration force could be neglected, in which case the linear velocity profile is an exact solution to the Navier-Stokes equations. The solution for the eigen-values, c , were obtained for two cases:

- (a) For small αR , *Crail's solution (Reference II-21) was used, where the characteristic equation for the eigen-values is obtained from the determinant of the boundary conditions. A value of Weber number to achieve instability was not calculated in the Reynolds study.
- (b) For large αR , a technique similar to that used by Miles (Reference II-23) was employed, where the characteristic equation is obtained from the normal force balance.

The characteristic equations may be solved to determine R as a function of α for any value of c . The neutral stability curve is of greatest interest because it represents the boundary between stability ($c_i < 0$) and instability ($c_i > 0$).

The solutions for neutral stability ($c_i = 0$) were carried out for both the large αR and small αR cases, and were joined together on a single plot, and a mathematical curve fit was used to obtain an expression for the neutral stability curve. This expression forms the basis for Reynold's microsphere stability criterion, and in his notation it is:

$$\alpha > 1.6R^{.3} [W / (1 - \beta / \alpha^2)]^{.4} \quad (\text{II-33})$$

where $W = R / \rho \sigma \delta / \mu^2$

$B = -\rho g_y \delta^2 / \sigma$ deceleration parameter

This curve fit is valid under the following conditions:

$$\alpha R > 100 \quad .5 < s < 4.0 \quad (\text{II-34})$$

where s is a parameter related to the surface speed, U_s , and the surface perturbation shear.

---

* An examination of the small αR case is given in Appendix B.

Equation II-33 was used by Reynolds to establish a stability criterion for re-entering microspheres by considering waves of $1/2$ the circumference of the body, in which case equation II-33 becomes:

$$M^3 W_e^{.7} [1 - 3/16 W_e]^{-.4} < 2 \text{ for stability} \quad (\text{II-35})$$

$$\text{where } W_e = \frac{\rho_{\infty} u_{\infty}^2 r}{\sigma}$$

$$M = \frac{\rho_{\infty} r}{\mu} \quad \text{is a dimensionless liquid property parameter}$$

c. Discussion of Analysis

Since this problem is a rather complex one, a number of assumptions and simplifications have been made in order to facilitate the analysis. These are:

1. The problem has been assumed to be two dimensional when it is actually three - dimensional.
2. The actual physical boundary conditions have been used but these have been linearized.
3. The axial deceleration forces have been neglected, which results in a linear liquid layer velocity profile.
4. It was assumed that the liquid flows all around the sphere.

Naturally, these assumptions impose some limitations on the analysis in that the problem which has been solved is only an approximation to the real physical situation. Just how much of a credibility gap this creates is difficult to say, but certainly the results must also be considered to be approximate.

The first two assumptions are reasonable to get order-of-magnitude values provided that the liquid layer thickness is small relative to the size of the body. The third assumption raises some questions; namely whether or not the use of a linear profile is justifiable, and if not, what effect does this have on the results of the theory?

In fact, this is a direct result of using a two dimensional analysis. For the real microsphere, the deceleration force does act normal to the liquid layer at the stagnation point, but the

direction varies around the surface of the microsphere such that it is in the axial direction 90° from the stagnation point. To carry this effect over into a two dimensional analysis, one would have to impose a sinusoidal (in x) body force on the liquid layer and this would complicate things considerably. In this case, the velocity profile would be parabolic in y and would also vary with x , thereby making the solution of the Orr-Sommerfeld equations quite difficult.

Since this force has been neglected in the analysis, the approximation should be considered to be valid only in the region near the stagnation point, where for all practical purposes, the deceleration force is normal to the surface of the liquid layer.

It is interesting to note that if equation II-35 is applied to the stagnation point, where $\theta_0 = 0$, a critical Weber Number of $16/3$ is obtained.

With regard to the application of the theory to the microsphere re-entry problem, two points are significant. First, the final stability criterion is based on the assumption that the longest possible wave (lowest possible wave number) exists on the liquid surface, and according to the theory, this wave is highly unstable. What should be pointed out, however, is that the theory also says that the lower wave lengths (higher wave number) are more stable. This means that the critical Weber Number for a given microsphere and release conditions is a function of the disturbance wave length on the surface. This can best be illustrated by the following example:

If one considers wave numbers of $= \frac{n(2\delta)}{r}$ where $n = 1, 2, 3 \dots$

this corresponds to disturbance wave lengths of $1/2, 1/4, 1/6 \dots$ of the microsphere circumference. In this case, equation II-33 becomes,

$$2n > m^3 [W_e \sin 2\theta_0]^{.7} \left[1 - \frac{3}{16n^2} W_e \cos \theta_0 \right]^{-.4} \quad (\text{II-36})$$

where θ_0 is the angle from the stagnation point. Following Reynold's example, if we use the fact that $\sin 2\theta_0$ and $\cos \theta_0$ can be no larger than 1.0, the stability criteria can be expressed by,

$$\frac{m^3 W_e^{.7}}{n \left| 1 - \frac{3W_e}{16n^2} \right|^{.4}} < 2 \quad (\text{II-37})$$

which is the same as Reynold's result if $n = 1$.

Equation II-37 was plotted for P_uO_2 microspheres of 50 and 250 microns in diameter to obtain $(W_e)_{crit}$ as a function of n , and the results appear in Figure II-11.

As can be seen from this plot, the higher wave number disturbances are much more stable than the lower wave number ones, and this must be interpreted to mean that a single critical Weber Number stability criterion does not exist. Although it is true that using the critical Weber Number corresponding to $n = 1$ will give an absolute lower bound (if $W_e < (W_e)_{crit}$

then stability is assured), but this does not necessarily mean that instability will result if the Weber number is larger. Such would be the case only if the low wave number disturbances were created during the re-entry. In other words, the critical Weber number for $n = 1$ can be used to predict stability but not instability. In all fairness, there may be no other recourse except to use the critical Weber number in this fashion, but it should be recognized that it may be extremely conservative in certain cases.

The assumption that the longest wave length is $1/2$ the circumference also implies that the thin liquid layer spreads evenly around the sphere. Since the aerodynamic shear force goes to zero at the tangent point in free molecular flow and the deceleration force acts forward for a non-spinning sphere, fluid would not be able to flow rearward at the tangent point. In fact, fluid would tend to flow forward. Based on this theoretical argument and based on Chapman's observations on tektites (Reference II-19), there would be a mass buildup in the vicinity of the tangent point, and no liquid mass would flow to the back side. Thus, the stability criterion of equation II-37 would be valid if n is allowed to correct for the fact that the liquid is restricted to the front surface. For $n = 2$, the longest wave length would be $1/4$ the circumference or from the stagnation point to the tangent point; from Figure II-11, the critical Weber number is approximately 1 for the 250 μm spheres and 0.5 for the 50 μm spheres (or approximately a factor of 3 higher than for $n = 1$).

The second, and more subtle, aspect of the Reynolds Theory is the restriction that $\alpha R > 100$. Although the liquid layer thickness drops out of the stability criteria, it is directly related to this restriction since for $\alpha = \frac{2\delta}{r}$ it becomes,

$$(\delta/r)^3 \left(\rho \frac{r^2}{\mu^2} \right) (P_\infty U_\infty^2) > 100 \quad (II-38)$$

If the microsphere size and material are specified, then equation II-38 can be used to determine the range of free stream dynamic pressure for which the theory is valid as a function of δ/r . This has been done for 50 and 250 μ diameter microspheres using the following P_{UO_2} properties,

$$\rho = 17.86 \text{ slug/ft.}^3$$

$$\mu = 6.68 \times 10^{-4} \text{ lb./sec./ft.}^2$$

and the results appear in Figures II-12 and II-13. Figure II-14 gives dynamic pressure as function of altitude and velocity so that it may be used to relate dynamic pressure to release conditions to use Figures II-12 and II-13.

It is important to remember here that the entire theory rests on the fact that δ/r must be small. As can be seen from the results of the calculations, that for small values of δ/r , acceptable release conditions do not correspond to the region of greatest interest. Essentially, the microsphere re-entry problem is associated with small particles which means small Reynold's numbers; hence, small values of αR . Since the curve fit on which the stability criteria is based is a large αR approximation, there is a rather limited range of microsphere release conditions for which the results are valid as presented in the form of the stability criteria.

In order to clarify this point, consider a typical release point, say 20,000 ft./sec. at 180,000 ft. altitude. From Figure II-14, this corresponds to a free stream dynamic pressure of about 220 lb./ft.². As can be seen from Figures II-12 and II-13, δ/r must be between 1/5 and 1/2 for a 250 μ particle, and greater than 1/2 for a 50 μ particle if αR is to be greater than 100. Since the theory is based on a thin liquid layer assumption, the validity of the stability criteria is questionable under these conditions..

Clearly, if the large αR restrictions were relaxed, more confidence in the theory could be obtained.

d. Conclusions

1. Although the importance of hydrodynamic instability for thin molten layers in hypersonic flow has been found by past investigators, the theory performed by Reynolds is not applicable to the problem of semi-molten microspheres.
2. The concept of a single Weber number stability criterion is questionable without more knowledge of the wave pattern in the molten layer.

FIGURE II-11
APPLICATION OF THE REYNOLDS STABILITY THEORY WHEN THE
DISTURBANCE WAVE LENGTH IS A FRACTION OF THE MICRO-
SPHERE CIRCUMFERENCE.

FREE STREAM DYNAMIC PRESSURE ~ PSF.

II-51

FIGURE II-14
DYNAMIC PRESSURE AS A
FUNCTION OF VELOCITY AND
ALTITUDE

3. The solution of the neutral stability equations makes the approximation that $\alpha R < 100$, which is not true for most release conditions where a thin molten layer exists.

e. The Character and Significance of Liner's Stability Analysis (G.E.)

The analysis performed by General Electric at Valley Forge considers potential mass loss of partially molten particles. The phenomena considers body deceleration forces acting on very thin liquid layers on a solid core. This is similar to the so-called Taylor instability (Reference II-17). For thin liquid layers the inertial forces resulting from the particle deceleration inhibit the flow of liquid into the wake so that the melt tends to accumulate. When this occurs, the dominant mode of instability would be the growth of small disturbances on the surface of the liquid. Such disturbances could grow into spikes which break off to form small droplets.

It should be noted that the G.E. study does not consider instability due to the so-called Craik-Benjamin (C-B) phenomenon (Reference II-17). This is caused by the supersonic shear forces on the microsphere surface in the area from $\phi = 45^\circ$ to 90° , where ϕ is the angle from the stagnation point-center axis as shown in Figure II-15. The shear stresses at the gas-liquid interface have been assumed to be zero. This fact had been recognized and discussed by G.E. in Reference II-3, and it was brought out by G.E. that the analysis was valid only in the region near the stagnation point. However, the analysis performed by Stanford (Reference II-17) has indicated that the zone near the sphere tangent point ($\phi = 90^\circ$) is the area at which instability first initiates. This is also the zone at which the G.E. analysis is least valid. Additional comments by Chapman indicates that mass loss will occur near the tangent point. However, the G.E. results appear to agree moderately well with the hypersonic tunnel tests run at Cornell Aeronautical Laboratory (Reference II-18). The Cornell tests indicate that breakup can be initiated in a small number of microspheres of a large statistical sample for partially molten droplets at Weber numbers from approximately 6 to 10, depending on the percent of the particle which is molten. The G.E. analysis predicts instability occurring at Weber number greater than 4.18.

f. Stability Analysis

The physical model used by G.E. is based on a study by Chang and Russell (Reference II-25) with modifications to account for the spherical geometry. The primary approximations are that the liquid layer thickness is constant over the front face of the particle and is very thin compared to the particle radius. This assumption, in addition to neglecting the aerodynamic shear

FIGURE II-15
COMPARISON OF INSTABILITY MODELS
USED BY STANFORD AND G.E.

forces, probably constitutes the biggest limitation to the analysis. Experience with tektites indicates that the liquid layer on a partially molten particle is neither thin nor of constant thickness. However, this approximation allows the curvature to be neglected so that the equations of motion can be written in cartesian coordinates. The present mathematical tools available can only solve the resultant differential equations in cartesian coordinates. Thus, the approximation of a thin layer is a necessary evil at present to obtain solutions but does cast doubt as to the accuracy of the conditions required to achieve instability.

It has also been assumed that the particle remains spherical. The mean velocities are assumed to be small so that the non-linear, inertial terms in the momentum equations can be neglected. The problem is treated as quasi-linear in the sense that the free stream conditions are treated as being constant.

The stability criterion derived by G.E. is given as equation (III-68) in Reference II-3 as:

$$\frac{F_r}{W_e} \geq \frac{1}{\pi} \quad , \quad \text{stability}$$

where F_r = Froude number

W_e = Weber number based on liquid density

The ratio $\frac{F_r}{W_e}$ is given by equation (III-69)

$$\frac{F_r}{W_e} = \frac{T}{\rho_1 G a^2}$$

where G = deceleration in g's

ρ_1 = liquid density

a = particle radius

T = surface tension

The expression for F_r is given in equation (III-76) as

$$\frac{4\rho_1}{F_r \rho_\infty} = \frac{3}{2} C_D$$

ρ_∞ = free stream density

The free stream critical Weber number can then be expressed as:

$$W_{e\infty} > \frac{8}{3} \pi \frac{1}{C_D}$$

For free molecular flow, $C_D = 2$ and $W_{e\infty} = 4.18$

The above value is the critical Weber number based on free molecular flow for a slightly molten particle. In continuum flow, the value for $W_{e\infty}$ would be approximately 8.4. For fully molten particles, the critical Weber number for microspheres for free molecular flow, was found to be equal to 2.

g. CAL Experiments on Fe_2O_3 Spheres (Beal)

A series of experiments were run (at CAL) on partially molten spheres inserted into a hypersonic flow of air (Reference II-18). The flow regime was transitional; between continuum and free molecular flow. CAL initially wanted to use Al_2O_3 particles to simulate PuO_2 microspheres but switched to Fe_2O_3 due to its lower melting temperature and the very good simulation of the viscosity and surface tension of PuO_2 by Fe_2O_3 . The particles were energized by a quartz heater as they dropped through the heater tube. When the spheres fell into the shock tube, they triggered and exploded the diaphragm which released the stored gas into the shock tube and created the hypersonic flow necessary for the test. Breakup, if it occurred, was recorded photographically for the 7 - 15 spheres visible in most tests. The percent melting of the spheres was controlled by the heater temperature and is correlated by post-test analysis.

The CAL test conditions are summarized in Table II-9 as reproduced from Reference II-18. The results are, in general, applicable to the release of PuO_2 microspheres as the flow and deceleration forces are encountered. The melting on an actual melting microsphere would be more on the front surface than evenly distributed as in the CAL experiments if the actual particles are non-spinning. Figure II-16 summarizes some of the test data; Weber number vs. radius fraction melted is compared. Slight breakup means that a few of, but not all, the spheres of a given test gave evidence of breakup.

h. Conclusions

Based on a collation of the analyses and experiments performed by Aerotherm (Reynolds), G.E. and CAL the following conclusions have been reached.

TABLE II-9
CAL TEST CONDITIONS*

RUN NO.	1	4	6	7	8	9	11	12	13	15
RESERVOIR CONDITIONS										
P_o , PSIA	2971	2894	6304	7392	6602	6253	6603	2802	2843	1747
H_o , FT ² /SEC ² x 10 ⁻⁶	52.58	69.04	79.98	74.30	81.86	81.50	81.68	69.70	70.36	64.37
T_o , °R	7076	9007	10,352	9648	10,514	10,500	10,534	9131	9185	8443
STEADY FLOW TEST TIME										
UNCONTAMINATED, ms	2.45	1.78	1.88	1.88	1.82	1.84	1.83	1.77	1.75	1.78
TOTAL, ms	6.00	5.60	5.35	5.50	5.30	5.32	5.32	5.60	5.58	5.70
STREAM CONDITIONS										
P_o' , PSIA	--	3.268	5.796	8.339	5.619	5.787	5.763	3.146	3.271	2.024
M_{∞}	11.87	11.00	10.90	10.38	11.00	10.90	10.93	11.05	11.00	10.54
u_{∞} , FT/SEC	10,079	11,530	12,390	11,918	12,539	12,506	12,535	11,572	11,625	11,100
P_{∞} , PSIA	.01242	.01859	.03693	.06111	.03534	.03626	.03688	.01702	.01779	.01411
T_{∞} , °R	290	441	520	530	523	530	529	441	449	446
ρ_{∞} , SLUGS/FT ³ x 10 ⁶	3.472	3.420	5.762	9.345	5.482	5.552	5.651	3.128	3.211	2.565
Re_{∞} /FT x 10 ⁻⁶	0.1566	0.1056	0.1927	0.3026	0.1888	0.1687	0.1928	0.1135	0.1154	0.07737
SPHERE CONDITIONS**										
$Re_{\infty, \phi}$	149	101	183	288	180	179	183	103	109	73
We	4.07	5.24	10.4	15.3	9.82	10.0	10.25	4.83	5.00	3.65
$K\eta_{\infty}$ ***	.118	.162	.088	.0536	.093	.090	.088	.152	.149	.213

* BASED ON THE PROPERTIES OF REAL NITROGEN

** BASED ON A SPHERE DIAMETER OF 283 MICRONS. THE VARIATION IN DIAMETER WAS $\pm 5\%$

*** COMPUTED BY $K\eta_{\infty} = 1.486 \frac{M_{\infty}}{Re_{\infty, \phi}}$

FIGURE II-16 BREAKUP PHENOMENA AS A FUNCTION OF ESTIMATED RADIUS FRACTION MELTED

1. There is a difference by more than an order of magnitude in the critical Weber number generated by G.E. and Aero-therm (4.18 vs. 0.1 - 0.2).
2. The assumptions of both analyses are very restrictive, and the value for critical Weber number cannot be used to make engineering judgements of the breakup of partially molten microspheres.
3. Based on the CAL tests, the critical Weber number for fully molten spheres is near, but slightly lower than, the value of six customarily accepted for continuum flow.
4. The critical Weber number for partially molten particles is higher than that for fully molten particles.

i. Application to Flight Conditions

The significance of the critical value for Weber number in terms of release conditions is shown in Figure II-17 and is compared to an orbital decay trajectory re-entry of a "typical" SNAP generator. The curves generated, for both 100 μ m and 200 μ m diameter particles, use three values of We_{crit} :

1. The 0.2 theoretical value of Reynolds (Stanford)
2. The 4.18 theoretical value of Liner (G.E.)
3. A slightly conservative value of 6 as obtained experimentally by Beal (CAL) for partially molten spheres.

The curves in Figure II-17 represent the band of conditions to predict potential mass loss based on the existing reported data on partially molten spheres. It can be seen that, if the critical value in flight is approximately 4-6, a release above 160,000 - 180,000 feet for an orbital decay re-entry would not result in mass loss regardless of melting. If the Stanford value is correct, mass loss will occur, providing there is melting, for all releases below 240,000 - 260,000 feet. However, since there has been no experimental evidence to substantiate the exact wave pattern of Reynold's analysis, and significant theoretical evidence to indicate that it just does not apply to real flight conditions, more dependence must be placed on the CAL experimental data ($We_{crit} > 6$). This would roughly explain any breakup or shear stripping which might occur in flight. However, it would not account for any mass loss due to vaporization, boiling of metal oxide impurities in the PuO_2 microspheres, the pressure buildup in small pockets, or some other process to remove molten mass not accounted for in the CAL experiments.

FIGURE II-17 PARTIALLY MOLTEN PARTICLE INSTABILITY DETERMINED BY CRITICAL WEBER NUMBER

In our opinion at present it is these second order effects rather than the classical breakup which predominate in the real flight case.

This is shown to some degree by the Sandia Arc Tunnel tests discussed in section II-E.

D. HEAT TRANSFER AND MASS FLOW DURING THE LIQUID/SOLID PHASE CHANGE

As discussed in Section II-B, the thermal analyses performed by Sandia, utilizing the SPHERE 2 Code, to predict the temperature response of microspheres after the initiation of melting, as shown in Figure II-4 does not account for any shape change. Analyses by G.E. and Aerotherm have shown that much liquid flow takes place. Therefore, the temperature predictions are inaccurate once melting starts. This effect could be significant if the SPHERE 2 Code is to be utilized during the NOL light gas gun program to predict melting vs. distance in the tunnel for various test conditions. This limitation also applies to the Martin and Aerotherm analyses.

The analysis performed by Aerotherm (Reference II-4) was to assess the effects of pressure and shear forces on the liquid flow during melting. The time to remove mass at the stagnation point was calculated and compared to the estimated time for complete melting (assuming no deformation) and time for breakup of a molten particle. Consider the physical system described in the following sketch:

A liquid layer of thickness δ is formed at the stagnation point of a body of local radius of curvature R_c . The surface recession rate at the stagnation point can be expressed as (see Appendix D of Reference II-4):

$$\dot{S} = \frac{(\rho v)_w}{\rho^*} + 2\delta \int_0^1 \frac{\partial u}{\partial x} d\eta \quad (II-39)$$

where

- $\dot{S}$ rate of surface recession (ft./sec.)
- $(\rho v)_w$ rate of gas phase mass removal at the surface (lbs./ft.²-sec.)
- ρ^* density of liquid (lb./ft.³)
- $\frac{\partial u}{\partial x}$ velocity gradient in the liquid layer
- $\eta = \frac{y}{\delta}$

The local velocity gradient can be expressed as:

$$\frac{\partial u}{\partial x} = \delta^2 \frac{\partial^2 p}{\partial x^2} \int_1^\eta \frac{\eta}{\mu} d\eta - \delta \frac{\partial \tau_w}{\partial x} \int_1^\eta \frac{d\eta}{\mu} \quad (II-40)$$

where

- μ = liquid viscosity
- τ_w = wall shear
- p = surface pressure

Equation II-39 was solved, for the stagnation point only, in an approximate manner by Aerotherm to obtain the recession rate for several release conditions. The time for one microsphere diameter to be ablated was calculated by integrating equation II-39 and compared to the calculated melting time. These results are illustrated in Table II-10.

By integrating $\dot{S}$, the distance ablated at the stagnation point parallel to the flow can be approximated. When $\int \dot{S} dt$ is equal to the diameter, all of the particle has been removed or ablated. These numbers are listed in column four of Table II-10. Comparing the time to melt and time to remove mass shows that liquid removal rates are very high and the time to remove mass is short (or of comparable magnitude) to the elapsed melting time for the release points studied. This analysis is solely used to illustrate the fact that much of the liquid will flow away from the stagnation point once melting begins and that the mass removal rates can be high enough to cause a significant redistribution of heat due to the liquid flow.

TABLE II-10

COMPARISON OF TIME TO REMOVE LIQUID MASS AT STAGNATION
POINT TO TIME FOR MELTING

Injection altitude (feet)	Injection velocity (ft./sec.)	Particle size (microns)	Time to ablate one diameter (seconds)	Calculated time to melt (seconds)	
				front face	rear face
244,000	25,200	250	0.012	0.125	0.425
244,000	25,200	50	0.0045	0.0155	0.0275
200,000	16,500	250	0.007	0.12	--
200,000	16,500	50	0.015	0.0085	0.0195
181,000	21,000	250	0.0032	0.010	0.075
181,000	21,000	50	0.0008	0.0015	0.0045
118,000	12,000	50	0.0004	0.0012	--

In a hyperthermal environment, the physical effects on a partially melting particle are extremely complex. Some vaporization and sublimation is possible, depending on the surface temperature and total aerodynamic pressure. A substantial amount of flow due to pressure and shear will most likely occur. Vaporization will be discussed in Section II-E. The present section is concerned with the problem of liquid flow and its effects on the overall heat transfer and the implications toward mass loss due to pressure and shear forces. This generally results in particle sizes on the order of microns, while vaporization results in debris in the molecular size range. A present estimate of the resulting radiological health hazards of $Pu^{238}O_2$ (Reference II-26) indicates that the worst size range from an inhalation standpoint is on the order of microns as opposed to molecular size.

The aerothermodynamic environment of a liquid layer requires the simultaneous solutions to the hydrodynamic, thermodynamic and heat transfer equations of the entire microsphere. The most difficult term to include in a "closed form" solution is the heat conduction in the solid-liquid state. Present solutions for the heat conduction exist only for a solid sphere.

An additional complication to the problem is the fact that some resolidification is probable on the back surface or sides of the sphere due to the large temperature differences from front face to back face. To illustrate the magnitude of the temperature differences throughout the

microsphere, results of the Aerotherm thermal analysis (Reference II-4) indicate that ΔT 's on the order of 2,000-3,000° F. at the initiation of melting from stagnation point to back surface for 250 μ diameter spheres exists for a release at 181,000 ft. altitude and a velocity of 21,000 ft./sec. Resolidification of the first liquid mass would depend on the liquid boundary layer flow rates. Certainly if the flow rates are fairly high, resolidification would occur somewhere near the flow tangent point (90° from the stagnation point).

As discussed in Section II-F, the effects of body forces from the drag deceleration are important in calculating the flow of the liquid layer. For test condition B of the Sandia arc tunnel experiments, a reduction by approximately 50% in aerodynamic shear results when body forces are accounted for.

A good understanding of the partially molten case is necessary for two reasons:

- a. In order to assess the mass loss due to body forces and aerodynamic shear, the existing theories have assumed a thin liquid layer distributed evenly around the microsphere. The approximations should not be made in a more sophisticated mass loss/instability analysis.
- b. A theory could be used to correlate the empirical mass loss data of the forthcoming NOL light gas gun experiments.

1. Chapman and Matting Melting Analysis

Two analyses are presently available to theoretically predict the thermal behavior and mass removal rates of a partially molten particle at the stagnation point of a blunt shape. (References II-13 and II-3). The analysis performed by Chapman and Matting considers the mass removal by flowing and vaporization or sublimation of a non-charring material for a blunt surface. The analysis has been machine-programmed for numerical solutions using a finite difference scheme. The velocity of the mass removal at the stagnation point is expressed by Equation 5 of Reference II-13 as (Figure II-18):

$$\bar{V} = \bar{V}_w - 2 \int_0^y \bar{u} dy_1 \quad (\text{II-41})$$

where

$\bar{V}_w$ = recession velocity of the liquid-solid interface in the particle

$\bar{u}$ = partial derivation of velocity in the x direction (Equation 9 of Reference II-13).

$$\bar{u}(y, t) = p'' \int_y^{y_{BF}} \frac{y_1 dy_1}{\mu} + \tau_w^1 \int_y^{y_{BF}} \frac{dy_1}{\mu}$$

$$\tau_w^1 = \frac{\partial \tau_w}{\partial x} = \text{partial derivation of shear}$$

p'' = the second derivation of P with x with a correction for body forces

μ = coefficient of viscosity

FIGURE II-18

Coordinate System
for Reference II-13
Analysis

If the value of $\bar{V}_w$ can be expressed independently of the liquid mass removal, such as by vaporization or by depolymerization (Arrhenius equation), the value of $\bar{V}$ can be calculated fairly accurately. Application of this theory to glassy materials and teflon gives good correlation with arc tunnel test data, as illustrated in Figures II-19 and II-20 (Reference II-13).

However, for application to PuO_2 microspheres, the value of $\bar{V}_w$ is dependent on the thermal conduction interior to the microsphere as described in Section II-B. Therefore, $\bar{V}_w$ cannot be calculated readily. The value of $\bar{V}_w$ in this case represents the melting rate at the front face and is, in turn, related to the liquid removal rates. Perhaps the biggest limitation of the theory is that it only predicts mass removal rates at the stagnation point by ablation and does not account for possible shear stripping at other areas on the front face. Also, the mass buildup near the sphere tangent point is not accounted for.

2. G.E. Melting Analysis

The analysis presented in Reference II-3 is applicable to stagnation point melting in the absence of gas-liquid interface re-radiation and vaporization mass transfer. It is further assumed that the thickness of the liquid layer is much smaller than the sphere diameter ($\delta \ll R$) so that the high Reynolds number laminar boundary layer equations can be used. This assumption is necessary to simplify the hy-

FIGURE II-19 - COMPARISON OF CALCULATED AND MEASURED SURFACE RECESSION OF TEKTITE GLASS IN A WIND TUNNEL.

FIGURE II-20 - COMPARISON OF CALCULATED AND MEASURED SURFACE RECESSION OF TEFLON IN WIND TUNNEL.

drodynamic equations to obtain numerical solutions. In reality, however, the liquid layer may be reasonably thick on certain portions of the sphere.

The biggest limitation of this theory, along with the Chapman and Matting theory, is that it can only be applied at the stagnation point. It does not consider the flow of liquid material over the entire sphere.

3. Experimental Correlations

An experiment is now being set up at Cornell Aeronautical Laboratories that may give some insight to the problem of partially molten thermal and dynamic behavior of PuO_2 microspheres. The tests will have spheres on the order of 1/4" diameter suspended by an upward flowing stream of helium. The flow will be transitional (near free molecular), and the melting point of the spheres will be low enough to observe melting. The liquid flow and breakup or stripping will be photographically measured during melting.

The most useful approach to assess the behavior of microspheres in the partially molten state is the forthcoming NOL light gas gun program. While it is not certain that breakup or stripping can be photographically recorded, the resultant debris in the tunnel will be collected and measured. This is the key information required from any analysis or experiment; namely: What is the resultant debris spectrum from PuO_2 for given real flight conditions?

4. Use of Tektite Analyses

Some observations on recovered tektites and meteors that have survived re-entry have been made by Chapman (Reference II-27). It was

concluded that much liquid flow and subsequent flattening out by aerodynamic heating, pressure forces and deceleration forces took place. The flattening and shape change effects were verified in hyperthermal arc tunnel experiments (Reference II-27), although the shape change in the arc tunnel experiments were not as distorted as those of recovered tektites since the deceleration forces were not present. It has also been noted that no apparent distortion similar to the tektites has been observed in the Sandia experiments on PuO_2 microspheres (Reference II-10) in the absence of deceleration forces. As discussed in Section II-F, the deceleration forces should cause molten buildup on the sides on semi-molten microspheres. The paradox of why the arc tunnel experiments on tektites result in much distortion and why the PuO_2 arc tunnel experiments do not could be attributed to the difference in flow conditions-continuum for tektites vs. near free molecular for PuO_2 . In free molecular flow there is zero pressure behind the protuberance or bulge, and any buildup would be blown around to the back surface in a static test. However, in continuum flow the expansion around the bulge could result in the buildup of local higher pressure regions behind the bulge, resulting in a stable position near the tangent point as was observed.

Chapman has observed critical Weber numbers much in excess of 10 for the entry of tektites. There has been some thought that this could be attributed to the higher viscosity of tektites over PuO_2 (Reference II-30). Opik (Reference II-31) has reported that at the melting point of meteoric stone, the viscosity is very high but decreases rapidly as temperature increases. This is shown in Table II-11 for several meteoric materials and compared with PuO_2 .

TABLE II-11

VISCOSITY OF SEVERAL STONY METEORIC MATERIALS IN POISE
(Extrapolated values are in parentheses)

T, °K	1383	1423	1473	1573	1673	2000	2500	3000
Olivine I	--	37900	3180	296	137	(18)	(2.4)	(1.0)
Olivine II	--	--	732	173	120	(18)	(2.4)	(1.0)
Basalt	3550	--	--	126	55	--	--	--
Techenite	21500	--	--	1820	851	--	--	--
Pu2390 ₂ *							.32	

*Measured only at the melting temperature of ~ 2240°C

COMPOSITION OF OLIVINE MATERIALS (%)

	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	FeO	MgO	CaO
Olivine I	49.29	18.49	2.38	6.77	6.09	8.14
Olivine II	49.24	16.81	6.16	3.60	8.02	9.49

Theoretically, the analyses of G.E. and Stanford (References II-3 and II-17) differ in conclusions on the effect of viscosity toward instability. G.E. concluded that the point of instability (Weber number) is independent of viscosity (the rate of instability depends on viscosity) while Stanford concludes that the Weber number is strongly dependent on viscosity. The stability criterion developed by Reynolds indicates that the We_{crit} is related to $1/\mu^2$; thus tektites would be much more unstable at their melting temperature than PuO_2 microspheres according to Figure 9 of Reference II-17. In general, the effect of viscosity on instability of the molten layer is not clearly defined. The work done on tektites is useful in showing an increase of stability of partially molten particles over fully molten spheres but cannot be applied directly to PuO_2 microspheres without a better understanding of the role of viscosity on hydrodynamic instability and subsequent mass removal.

5. Conclusions

- a. For most release conditions, liquid mass will flow away from the stagnation point once melting initiates.
- b. Due to the large ΔT 's in the sphere, some resolidification is possible on the sides and back surface.
- c. The thermal conduction analyses, such as SPHERE 2, the Aero-therm thermal code and the Martin code are not applicable once melting initiates at the stagnation point.
- d. The aerothermodynamic environment of a partially molten sphere is extremely complex. To date, no theory adequately predicts the heat transfer and deformation in the partially molten state. Thus, the stability analyses of Section II-C cannot be refined to account for a finite liquid layer thickness and a resulting shape change.
- e. The effects of deceleration forces during the microspheres release are significant on the liquid layer deformation and its removal potential from the surface. The integrated effect is beneficial i.e., inhibits shear stripping.
- f. The NOL light gas gun experiments appear to be the only method of determining real flight mass loss potential for fully and partially molten microspheres. It does not seem likely that a theory sophisticated enough to fully explain the thermal, dynamic and mass loss behavior for partially molten spheres can be developed in the near future. Hence, an experimental approach is the only sensible path to take.
- g. The CAL levitation experiments may yield some empirical data to partially explain the liquid layer flow phenomena, which could be incorporated into an expanded theory.

E. MASS LOSS BY VAPORIZATION

At the relatively high melting temperature of PuO_2 , surface vaporization will occur if the total aerodynamic pressure is less than the vapor pressure. For most atmospheric release altitudes, this will be the case. The vaporization has two effects on the thermal/mass loss behavior of the melting microspheres:

1. Mass is lost due to vaporization of molecular size particles.
2. The aerodynamic heat transfer is slightly reduced by the "blocking" effect of the vaporizing PuO_2 .

The first effect is of more significance as it results in mass loss; mass loss, in general, is to be avoided if possible. The evaporation of material from a surface is categorized as rate-controlled or diffusion-controlled. The rate-controlled regime is valid for free-molecular flow and will represent the mechanism by which most of the material vaporizes. As the external flow becomes less rarefied, the diffusion controlled mechanism is more pronounced.

An estimate of the surface vaporization in the rate-controlled regime is given by the Langmuir equation (Reference II-30):

$$\dot{m}_{FM} = \frac{P_v}{\sqrt{2\pi M \bar{R} T}} \quad (II-42)$$

where

$$\begin{aligned} \dot{m}_{FM} &= \text{surface mass loss rate in free molecular flow} \\ P_v &= \text{vapor pressure} \\ M &= \text{molecular weight} \\ \bar{R} &= \text{gas constant} \\ T &= \text{temperature } ^\circ\text{ absolute} \end{aligned}$$

For PuO_2 , equation II-42 can be written as (Reference II-30):

$$\dot{m}_{FM} = 0.1601 P_v \text{ (mm of H}_g\text{)} \sqrt{\frac{M}{T(^{\circ}R)}} \text{ (lb/ft}^2\text{ - sec)}$$

The vapor pressure of PuO_2 is given by Reference II-30 as:

$$\log_{10} P_v = 8.072 - \frac{29.40}{T} \quad (II-43)$$

where P_v is in atmospheres and T is in degrees Kelvin.

In the diffusion-controlled regime, the vapor mass loss is given by Reference II-13 as:

$$\dot{m}_d = \frac{\dot{q}_{\psi_c}}{\frac{28.9}{M} \left(\frac{P}{P_v} - 1 \right) (H_s - H_w)} \quad (\text{II-44})$$

$\dot{q}_{\psi_c}$ = aerodynamic heating rate, with a correction for vapor blockage

P = total aerodynamic pressure at sphere surface

H_s = free stream enthalpy

H_w = total enthalpy at wall

In the transition vaporization regime between rate-controlled and diffusion-controlled, an approximate "bridging" equation is given by Reference II-13 as:

$$\frac{1}{\dot{m}_T} = \frac{1}{\dot{m}_{FM}} + \frac{1}{\dot{m}_d} \quad (\text{II-45})$$

$\dot{m}_T$ = total vaporization rate

The thermal analyses performed by the Martin Company (Reference II-6) considered both the mass and heat lost due to vaporization and the effect of the vapor blockage on the aerodynamic heating rates. The Sandia SPHERE 2 Code (Reference II-5) also accounts for the same effects, i.e., mass and heat transfer by vaporization. Both analyses use equations similar to II-42 through II-46. However, the earlier Sandia analyses performed by Stephenson (Reference II-9) did not consider vaporization. The Aerotherm Corporation (Reference II-4) performed an approximate analysis based on equation II-42 that was not coupled to the transient melting analysis.

The equation for aerodynamic heating when vapor blocking is considered, used by both Martin and Sandia, is given by Reference II-13 as:

$$\dot{q}_{\text{blowing}} = \psi \dot{q}_{\text{no blowing}} \quad (\text{II-46})$$

$$\psi = \frac{0.94}{1.0 + \frac{c_1}{c_2}} + 0.06$$

$$c_1 = \frac{0.75}{(28.9/M)^{0.7}}$$

M = molecular weight of vapor

$$c_2 = \frac{P_\theta \cos^2 \theta}{P_v} - 1$$

P_θ = stagnation aerodynamic pressure

θ = angle measured from stagnation point

1. Application to Flight Conditions

The analysis performed by Martin In Reference II-6 indicates that some vaporization will occur for all conditions where melting takes place, but the relative hazard due to vaporization is much less (1-2 orders of magnitude) than mass loss due to stripping or breakup (if this occurs). As an example, Martin has computed the expected tumor incidence for a SNAP-19 mission due to several potential sources of debris (plus the original microsphere) for various points on the launch vehicle trajectory. This is illustrated in Table II-12.

A value of 0.0 for melt debris indicates no breakup has occurred, utilizing a critical value of Weber number of 5.0. It can be seen from Table II-12 that a finite tumor incidence will result from vaporization, but the effect is considerably smaller than the tumor incidences resulting from breakup.

The approximate calculations by Aerotherm (Reference II-4) have shown that vaporization may occur for all release conditions where the microspheres start to melt (see Figure II-3), although the amount of mass vaporization was not accurately calculated.

2. Application to Sandia Arc Tunnel Experiments

As will be discussed in Section II-F, the resultant debris due to testing the $^{238}\text{PuO}_2$ microspheres in the plasma arc tunnel (Reference II-10) contained a large number of very small particles ($<0.01\mu$, See Figure I-1). The exact character of the debris below approximately 0.01μ is unknown as the diameter cannot be accurately measured below this size. Since debris appears for test conditions A, B, C and K and no stripping or breakup is evident, it was speculated by Weiner Associates, Inc. (in view of the Martin and Aerotherm calculations for real flight conditions) that vaporization could account for some of the debris caught on the target. Some subsequent preliminary calculations by Sandia have indicated that the vaporization rates are the same order of magnitude as the amount of small debris caught by the arc tunnel target. Thus, some of the mass loss, especially for conditions A, B, C and K, could be due to vaporization. However, other mechanisms of mass loss, such as boiloff of impurities or helium pressure blowout, are postulated to account for some of the large debris ($>0.05 - 0.10\mu$) as discussed in the next section.

TABLE II-12
(Reference II-6)
Expected Tumor Incidence from Inhalation, Critical Weber Number = 5.0
Expected Tumor Incidence

	Fuel Release Condition	1 Melt Debris	2 Vapor	3 Production Fines	4 Microspheres	5 Total
Agena first burn abort	1+1	2.8×10^1	1.3×10^{-2}	9.0×10^{-4}	0.0	2.8×10^1
	2+2	4.7×10^1	1.7×10^0	9.0×10^{-4}	0.0	4.9×10^1
	3+3	1.8×10^0	1.3×10^0	9.0×10^{-4}	0.0	3.1×10^0
	4+4	0.0	4.0×10^0	1.0×10^{-3}	0.0	4.0×10^0
	5+5	0.0	4.3×10^0	1.0×10^{-3}	0.0	4.3×10^0
	6+6	0.0	1.9×10^0	1.0×10^{-3}	0.0	1.9×10^0
	7+7	0.0	4.8×10^{-3}	4.0×10^{-4}	0.0	5.2×10^{-3}
Short orbit abort	8+8	0.0	6.2×10^{-2}	1.5×10^{-3}	6.1×10^{-6}	6.4×10^{-2}
	9+9	0.0	5.4×10^{-1}	7.3×10^{-4}	3.0×10^{-6}	5.4×10^{-1}
	10+10	0.0	1.8×10^0	7.3×10^{-4}	3.0×10^{-6}	1.8×10^0
	11+11	0.0	0.0	2.9×10^{-3}	2.4×10^{-5}	2.9×10^{-3}
	12+12	0.0	0.0	1.5×10^{-3}	1.2×10^{-5}	1.5×10^{-3}
Agena second burn deorbit	1+4	2.5×10^{-1}	6.6×10^{-2}	2.9×10^{-6}	2.8×10^{-8}	3.2×10^{-1}
	1+6	1.5×10^{-2}	2.1×10^{-2}	4.3×10^{-7}	4.1×10^{-9}	3.6×10^{-2}
	2+6	8.5×10^{-2}	1.7×10^{-1}	2.9×10^{-6}	2.9×10^{-8}	2.6×10^{-1}
	3+6	7.2×10^{-2}	1.3×10^{-1}	2.9×10^{-6}	3.1×10^{-8}	2.0×10^{-1}
	4+6	0.0	4.5×10^{-2}	2.9×10^{-6}	3.1×10^{-8}	4.5×10^{-2}
Nonnominal cases applicable to short orbit and Agena second burn deorbit aborts	1+3	3.7×10^1	2.20×10^0	3.4×10^{-4}	0.0	3.9×10^1
	2+1	2.5×10^0	1.2×10^{-2}	3.4×10^{-4}	0.0	2.5×10^0
	2+4	5.1×10^1	1.6×10^1	6.8×10^{-4}	0.0	6.7×10^1
	3+4	1.7×10^1	7.1×10^0	6.8×10^{-4}	0.0	2.4×10^1

3. Conclusions

- a. For all release conditions where melting occurs, some vaporization will result.
- b. The tumor incidence for vaporized debris for all aborts postulated for the SNAP-19 mission is considerably less than the tumor incidence resulting from breakup of the molten microspheres. This conclusion would generally be true for an earth orbiting RTG mission that would utilize PuO_2 microspheres.
- c. Vaporization could account for some of the debris caught in the hyperthermal arc tunnel.

F. SANDIA ARC TUNNEL EXPERIMENTS

A series of experiments were conducted in the Sandia Corporation 40-kW hyperthermal arc tunnel on the hypersonic melting and ablation of 238PuO_2 microspheres (Reference II-10). The thermal and dynamic response of the microspheres (nominal diameter of $250\mu\text{m}$) was observed with high speed movie cameras, and the debris removed by the hot airstream was collected on samples placed downstream of the particles. The test objective was to measure the size and amount of the removed debris and correlate this to real atmospheric release conditions by simulating the heating and aerodynamic environment in the arc tunnel to those that would be encountered in real flight. The Sandia test conditions are summarized in Table II-13 and are called A, B, C, K, L, M for reference purposes with condition M being the most severe.

The dynamic and thermal response of a microsphere in a hypersonic environment depends (in addition to its own thermophysical properties) on (1) the aerodynamic heating rates, (2) total aerodynamic heat input, (3) total enthalpy, (4) static and dynamic pressure (Weber number), (5) aerodynamic shear stresses and (6) deceleration body forces.

TABLE II-13
SANDIA ARC TUNNEL EXPERIMENTAL CONDITIONS

Condition	Enthalpy BTU/lb.	Model Pressure Atm.	Stagnation Heat Flux (BTU/ft ² -sec)	Weber Number
A	11,000	0.0485	650	1.0
B	10,000	0.0737	950	1.6
C	9,500	0.0593	800	1.2
K	13,000	0.0987	1800	2.2
L*	3,600	0.0237	100	--
M	12,000	0.276	2800	4.6

* No melting observed

SANDIA ARC TUNNEL EXPERIMENTS
 COMPARISON OF LINES OF CONSTANT HEAT FLUX
 WITH LINES OF CONSTANT WEBER NO. TO ILLUSTRATE
 MISMATCH OF HEATING RATES AND WEBER NO. AT THE FLIGHT
 CONDITIONS OF INTEREST (20,000 - 25,000 FT./SEC. OR HIGHER)

The aerodynamic heating rates, total heating and enthalpy determine the amount of melting taking place within the microsphere. The static and dynamic pressure, shear stresses, body forces and total enthalpy govern the ablation, stability of the liquid layer and breakup of the partially molten particle. An inherent feature of the arc tunnel is that it is impossible to match the heating rates, Weber number and enthalpy at the same simulated release condition. This is illustrated in Figure II-21, where lines of constant aerodynamic heating and Weber number, taken from Table II-13, are plotted. Also the flight velocity which matches the test enthalpy is indicated. As can be seen, all three conditions are not matched at any one real flight condition. The actual flight Weber number at the point of simulated enthalpy is considerably lower. Another way of interpreting Figure II-21 is that at velocities between 20,000 and 25,000 ft./sec., the Weber number and subsequent shear forces would be much lower in flight than in the arc tunnel. In general, the 20,000 - 25,000 ft./sec. velocity range is where most atmospheric release conditions would occur and where an accurate simulation of aerodynamic heating and flow conditions is most desirable (higher velocities up to 36,000 ft./sec. are required for the SNAP-27 mission). For test conditions A, B and C the flow and heating conditions match between 10,000 and 15,000 ft./sec.

An additional drawback of arc tunnel simulation is that the aerodynamic deceleration is not simulated. There had been some thought in Reference II-10 that the deceleration forces would be further destabilizing. This is true at the stagnation point; however, as the molten mass flows away from the stagnation point, the deceleration forces would oppose the aerodynamic shear forces, and the net force tending to strip away mass on the front of the microsphere would be less*. An approximate solution to the effective shear with the presence of body forces was derived during the program. The ratio of shear stress in flight to shear stress in the tunnel is given by Equation II-51.

The results of the Sandia experiments have shown that some debris was lost for all test conditions with the exception of condition L. However, as discussed in the preceding paragraphs it is difficult, if not impossible to correlate the Sandia results to any real flight conditions. In reviewing the Sandia test results, Section II-F is further divided into the following subsections:

1. Effect of hyperthermal environment on mass loss.
2. Character of the debris as measured by Sandia, and a rational explanation of its origin.

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* Flow runback was observed for conditions K and M, the two most severe points. Stripping was only thought to occur for condition M. Thus, it will be hypothesized that breakup or instability - not necessarily mass loss - will be due to aerodynamic shear stripping. The distinction between instability and mass loss is made because for conditions A, B and C there is a small amount of mass loss with no correlation to stripping or breakup and could be due to other reasons.

3. Conclusions on the application of the test data to real flight conditions

1. Effect of Hyperthermal Environment on Mass Loss

Mass loss may be experienced through one or several of the following mechanisms.

a. Gaseous Ablation

As discussed in Section II-E vaporization is generally represented as rate-controlled or diffusion-controlled. The equations representing mass loss (Equation II-42 and II-44) for the two mechanisms are, respectively:

$$\dot{m}_{FM} = \frac{P_v}{\sqrt{2\pi MRT}} \quad (\text{rate-controlled})$$

$$\dot{m}_d = \frac{q_{\psi c}}{\frac{28.9}{M} \left(\frac{P}{P_v} - 1 \right) (H_s - H_w)} \quad (\text{diffusion-controlled})$$

The rate of mass loss for the diffusion controlled regime is dependent on the heating rate, pressure and enthalpy. As discussed previously, all three conditions are not simultaneously simulated in the arc tunnel. The total mass loss due to diffusion-controlled vaporization is a function of the total heat input. The rate-controlled vaporization, however, is dependent of P_v and temperature, which are accurately simulated in the arc tunnel. Thus, the vaporization mass loss is only approximately duplicated in the arc tunnel with the difference being in the diffusion-controlled regime.

b. Spallation and/or Ablation Mechanical Stripping

As the particle melts, a liquid layer forms initially in the stagnation region and is moved downstream by the shear and pressure forces exerted by the oncoming air. As this liquid reaches between 45° to 90° from the stagnation point, these shear and pressure forces have a tendency to strip it away from the solid portion of the particle. The shear force between a solid and the liquid is influenced by the body force acting on the particle. The effect of this influence and the corresponding difference between the shears experienced by the microsphere in flight and while being mounted in the tunnel was investigated analytically as shown below. Let us assume that locally the liquid layer may be represented by a Couette type flow, which is an approximate solution to the boundary layer equations. However, at the liquid gas interface, the boundary condition is changed to that of equal shear stress. Further, we shall assume

FIGURE II-22
 COORDINATE SYSTEM FOR APPROXIMATE SOLUTION TO
 BOUNDARY LAYER EQUATION FOR A LIQUID LAYER IN
 FREE MOLECULAR FLOW

that both the shear and pressure exerted by the air are given by the free molecular values which are consistent with the Sandia experiments or are more severe, hence conservative. The complete derivation is given in Appendix A. The differential equation governing the problem is (see sketch, Figure II-22):

$$\frac{d^2u}{dy^2} = \frac{1}{\mu} \frac{dp}{dx} - \frac{F}{\mu} \quad (\text{II-47})$$

where:

- u = velocity
- y = coordinate normal to flow
- μ = coefficient of viscosity
- p = pressure
- x = coordinate parallel to flow
- F = body force due to deceleration

The boundary condition at the solid-liquid interface is:

$$\text{at } y = 0 ; \quad u = 0$$

and at the liquid-gas interface:

$$\text{at } y = \delta ; \quad \tau = \tau_\infty = \rho_\infty U_\infty^2 \sin\left(\frac{x}{R}\right) \cos\left(\frac{x}{R}\right) \quad (\text{II-48})$$

44

where (Reference II-31 for free molecular flow):

δ = liquid layer thickness

τ = shear stress

ρ = liquid density

ρ_{∞} = air density

U_{∞} = free stream velocity

The pressure is given by,

$$P = \rho_{\infty} U_{\infty}^2 \cos^2 \left(\frac{x}{R} \right) \quad (\text{II-49})$$

In the tunnel $F = 0$; while in flight,

$$F = -\frac{3}{4} \frac{\rho_{\infty} U_{\infty}^2}{R} \sin \frac{x}{R} \quad (\text{II-50})$$

Solving this boundary value problem for the flight and the tunnel situations, the ratio of the shear stresses experienced at the solid-liquid interface may be written as:

$$\frac{\tau}{\tau} = \frac{\tau_{\text{flight}}}{\tau_{\text{tunnel}}} = \frac{W_{e \text{ flight}}}{W_{e \text{ tunnel}}} \left[1 - \frac{\frac{3}{4} \frac{1}{\cos \left(\frac{x}{R} \right)} \frac{\delta}{R}}{1 + \frac{2\delta}{R}} \right] \quad (\text{II-51})$$

The first term on the right hand side of the above equation shows the effect of Weber number simulation, and the second term shows the effect of body force simulation. It may be seen that both can have an important effect on the shear experienced by the microsphere in flight as compared to the tunnel, with the effect of the body force increasing in importance as the melt layer thickens.

The effect of the deceleration forces only, and not of Weber number, is shown in Figure II-23 as a function of δ/R and position on the microsphere (x/R) based on the bracketed term of equation II-51. It can be seen that the deceleration effect is relatively unimportant near the stagnation point but reduces significantly the shear stress as the flow approaches the tangent point ($\pi/2$). At some angle close to $\pi/2$, as a function of δ/R , the total shear goes to zero. At this point the liquid layer tends to pile up and start the "flanges" that have been experimentally found by Chapman.

To illustrate this effect on the Sandia experiments, the effective shear stress for conditions A, B, C, K and M was calculated with the effects of Weber number and deceleration forces

accounted for. The ratio is the actual flight shear stress divided by the arc tunnel shear stresses and is shown in Figure II-24. The test Weber number was taken from data supplied by Sandia (Reference II-10).

The important conclusion to be drawn from this analysis is that in order to observe the particle dynamic characteristics correctly in a wind tunnel, it is not sufficient to simulate the heating rate, but Weber number and body force are of equal importance. To illustrate this fact, we have examined Sandia test condition B and compared it with the flight condition where the heating rate is duplicated; assuming that the melt layer thickness was 10% of the particle radius. It was found that $\bar{\tau} = .56$ which means that the shear in flight is only 56% of the shear experienced in the tunnel for the assumed liquid layer thickness. Therefore, mass removal by stripping is a more critical mass loss mechanism in the tunnel than in flight. However, it must be kept in mind that the analysis in equation II-51 must select a value of δ/R . Real values of this are not known for PuO_2 microspheres. A value of 10% was selected as it was felt that a liquid thickness on the order of 20-30% δ/R could be experienced based on some of Chapman's work on tektites. The significance of this phenomenon is enhanced by the fact that the heating rate simulated by the tunnel is a constant equal to the initial heating rate experienced by the microsphere in flight. Therefore, since the heating rate decreases in flight and consequently the total heat during a given time period is less than in the tunnel, the melt layer thickness in the tunnel is larger and the importance of the stripping phenomenon is even larger.

c. Liquid Layer or Molten Particle Instability

The type of phenomenon considered here is the amplification of waves in the surface of the liquid layer due to the pressure and shear forces exerted by the air stream. The stability analysis is an unsteady analysis and due to mathematical difficulties is normally restricted to a flat plate (no pressure gradient) thin liquid layer type linearized perturbation analysis (Reference II-17) which, under the best of circumstances, provides order of magnitude type estimates. The limitations of the theoretical studies for liquid layer instability are discussed in depth in Section II-C. In this instance, both the zero pressure gradient and thin liquid layer assumption are grossly violated and, therefore, it is difficult to assess the quantitative significance of the critical parameters predicted in Reference II-17. However, qualitatively, there is little doubt that the Weber number and the Ohnesorge number are significant parameters for the liquid layer instability and the Weber number and Bond number for the molten particle instability. Therefore, simulating these parameters and especially, the Weber number, is very important in a tunnel test aimed at assessing the dynamics and breakup of the microspheres.

FIG II-23 ~ EFFECT OF DECELERATION FORCES
ON LIQUID LAYER SHEAR

FIG II-24 ~ EFFECT OF WEBER NUMBER AND
DECELERATION FORCES ON LIQUID
LAYER SHEAR.

2. Character of the Debris as Measured by Sandia and a Rational Explanation of its Origin

The $^{238}\text{PuO}_2$ debris on the impact samplers was analyzed for size distribution by Tracerlab, Inc. (Reference II-10). One sample each from tests at condition A, K, and M was analyzed. The lower limit of resolution in particle size, utilizing Tracerlab's hollow star method, was $0.05\mu\text{m}$. Consequently, the size range of the debris counted by Tracerlab using this method extended from $0.05\mu\text{m}$ to the largest particle found on each of the slides.

A collector efficiency analysis of the samplers was performed by Sandia to determine the size range of the particles which could be expected to be collected. This analysis included flow patterns, effect of shock waves and sampler configuration. In general, the analysis indicated that particles between 0.01 and $10\mu\text{m}$ in diameter would be collected with approximately 100% efficiency (although particles as large as $14\mu\text{m}$ were caught). Particles less than $0.01\mu\text{m}$ in diameter would be trapped in the flow and diverted around the sampler. Since the microspheres were weighed and measured before and after the test, the approximate total mass loss was determined. The total debris mass on the targets was also determined. For conditions A and K the target mass was considerably less than the measured microsphere mass loss and was attributed to particles less than $0.01\mu\text{m}$; no particles bigger than $10\mu\text{m}$ were expected for A and K as the largest particles measured on the target were 1 – $1.6\mu\text{m}$. For condition M, approximately 93% of the mass is attributed to particles greater than $10\mu\text{m}$ as the particle mass in the sampler contains only 6.94% of the measured microsphere mass loss and breakup and stripping were observed. However, some of the 93% of the unaccounted for mass loss could be in particles smaller than $0.01\mu\text{m}$ as these would have also escaped detection.

Table II-14 shows the approximate size distribution of the ablated debris, based on the Tracerlab analysis and modified by the Sandia sampler efficiency analysis (Reference II-10). Although the data in Table II-14 are based on one test for each re-entry condition, Sandia believes that the size distribution of the debris is reasonably representative, especially for conditions A and K. The condition M data are questionable because the molten runback material was considered as large ablated particles. The final fate and debris size of the runback material in an actual re-entry cannot be determined in the arc tunnel facility.

As seen from Table II-14, approximately 91% of the debris for condition K is below $0.01\mu\text{m}$ and 47% for condition A is below this size. Since $0.01\mu\text{m}$ (100 Angstroms) is a size approaching the PuO_2 molecular diameter (metallic radius = $3\text{\AA}$), it is Weiner Associates, Inc. opinion that much, if not all, of the debris below $0.01\mu\text{m}$ for conditions A and K is due to vaporization. Some preliminary calculations by Sandia have shown that the theoretical mass loss by

vaporization is close to the mass loss as summarized in Table II-14. It has been further stated by Sandia, on page 10 of Reference II-10, that the total mass loss for condition K represents approximately 5-10% of the original mass. Thus, if 90% of this mass loss is due to vaporization, only 1/2 - 1% of the original PuO_2 microsphere mass is due to other mass loss mechanisms. For condition A, vaporization could account for 47% of the mass loss with the remaining 53% due to other mechanisms. However, the total mass loss for condition A is considerably less than that for condition K so that the approximately 1/2 - 1% of the original mass removed by mechanisms other than vaporization seems reasonably correct.

Only for condition M, the most severe at a Weber number of 4.6, has stripping of the liquid material been theorized. However, Sandia has observed the presence of small bubbles or protuberances in conditions A, B, C and K that seem to "erupt" between movie frames. Since only a very small amount of mass (1/2 - 1% of the original microsphere mass) is unaccounted for, these "erruptions" could be a very logical cause of this mass removed for conditions A and K and could be due to several phenomena:

- a. Boiling of iron oxide and silica impurities inherent to production PuO_2 microspheres. These impurities could be trapped in the thin liquid layer and vaporize and, in the process of boiling, push or pull a small amount of PuO_2 debris with the gases.
- b. A similar effect as in (a) due to the release of rapidly expanded H_2 gas trapped in the void spaces.
- c. Surface erosion of the PuO_2 microspheres. Since the microscopic surface is very rough with little "tits" hanging on, these "tits" could be rubbed off by aerodynamic shear stresses as the surface starts to melt while the liquid layer film still adheres to the microsphere.

Thus, one can conclude that, based on the photographic records and the arguments presented in the preceding paragraphs, that the PuO_2 debris for conditions A, B, C and K are due to some mass removal mechanism other than classical breakup or stripping. Stripping breakup may have occurred at condition M at a test Weber number of 4.6. As discussed in subsection (1), the shear stripping would be considerably less in the presence of the real flight deceleration forces. When the deceleration forces are accounted for, the "effective" Weber number would be higher than 4.6 and could be in the 6-10 range as obtained experimentally by Cornell Aeronautical Laboratory (Reference II-18) for partially molten Fe_2O_3 spheres.

Whether the same mass loss profile would occur for condition M in real flight is seriously in question due to the fact that the deceleration forces are not accounted for.

3. Conclusions on the Application of the Test Data to Real Flight Conditions

- a. The hyperthermal arc tunnel experiments impose a more severe environment on the microsphere than actual flight conditions because they do not simulate the deceleration forces which are important for evaluating the dynamic behavior and the breakup of the particle. Therefore, until more representative experiments are carried out (such as those planned at NOL in FY 1969), no definite conclusions should be drawn on the particles' re-entry hazard characteristics.
- b. It seems very likely that some vaporization is occurring in the arc tunnel experiments and, if this is true, would probably occur for real flight releases where some melting takes place.
- c. Vaporization probably accounts for the considerable P_uO_2 debris below $0.01\mu m$ that escapes detection in conditions A and K (although a direct correlation to real flight conditions cannot be made).

TABLE II-14
PERCENTAGES OF TOTAL MASS OF
 $^{238}P_uO_2$ ABLATED FROM MICROSPHERES

Size Range of Particle Diameters (μm)	Percent of Ablated Mass	Cumulative Percent
<u>Condition A - Sampler M-3</u>		
0 to 0.01	47.73	47.73
0.01 to 0.3	52.20	99.93
0.3 to 0.9	0.07	100.00
<u>Condition K - Sampler M-35</u>		
0 to 0.01	90.71	90.71
0.01 to 0.05	8.9	99.61
0.05 to 0.1	0.32	99.93
0.1 to 1.6	0.07	100.00
<u>Condition M - Sampler M-29</u>		
0 to 0.01	Very small	--
0.01 to 0.2	6.6	6.6
0.2 to 14.0	0.34	6.94
>10.0	93.06	100.00

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III. ASSESSMENT OF FUTURE EXPERIMENTAL ANALYSES

The results of the collation on past analyses and experiments have been that there is a general lack of data on the mass loss and break-up of PuO_2 microspheres that can be directly applied to real atmospheric release conditions. To correct this situation, the AEC-SNS has initiated a light gas gun experiment at NOL to simulate exactly the complete hyperthermal environment. The results should be able to be directly applied to real atmospheric release conditions since the exact flow and deceleration environment is being seen. Additionally, $^{238}PuO_2$ microspheres will be tested so that extrapolation of data from other materials is not necessary as has been the case in the CAL shock tunnel experiments and the GMRDL ballistic tunnel experiments.

A study was performed by WAI in Reference III-1 to outline a matrix of test points to be used in the NOL experiments. In general, the results of this collation study can be helpful in setting up the instrumentation for the test program. The following pertinent conclusions to the NOL tests will be made:

1. Instrumentation must be set up to catch debris due to two separate phenomena: (a) vaporization and/or blowout of hot gases from void spaces and (b) stripping or breakup. The above conclusion is based on the Sandia experiments where item (a) occurred before breakup was observed. In general, vaporization will probably start to occur as soon as melting begins.
2. The relative position of melting initiation in the tunnel must be known so that the proper instrumentation can be set up.
3. The existing thermal analysis techniques must be modified so that an accurate heat transfer and melting calculation can be made once melting initiates so that the remainder of the instrumentation can be placed. The present thermal analyses are inaccurate once melting begins.

A. CAL Levitation Experiments

It is expected that the tests to be performed at CAL to levitate spheres approximately 1/4" in diameter and observe melting and molten flowback and/or breakup will be helpful in understanding the behavior of small spheres once melting begins.

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109

IV. CONCLUSIONS

1. For the first time, the pros and cons on all the analyses and experiments on the re-entry of P_uO_2 microspheres based on a detailed review of this work, has been documented. A state-of-the-art survey was established.
2. The theories, such as the ones formulated by G. E. and Aerotherm, to predict breakup or instability for both fully and semi-molten microspheres are too approximate to be used. At best, order of magnitude values were generated. Unfortunately, the AEC-SNS requires more accurate information to determine whether breakup (and mass loss resulting from breakup) will occur for all possible atmospheric release points.
3. The most useful data for correlating breakup are the CAL shock tunnel experiments on zirconium and Fe_2O_3 particles and, to a lesser extent, the Sandia arc tunnel experiments on $^{238}P_uO_2$ microspheres. However, the experimental results, for both programs, cannot be directly applied to real atmospheric release conditions.
4. Based on the theoretical studies, melting initiation can be predicted in release coordinates.
5. Once melting begins, the fluid dynamic behavior of the molten P_uO_2 and its effects on the heat transfer is not known. The available thermal models do not take the flow deformation effect into account. Significant deformation will result primarily due to deceleration forces, based on Chapman's tektite studies.
6. The only experiments where deceleration forces were included are the CAL shock tunnel experiments.
7. Based on the Sandia arc tunnel experiments, two criteria for mass loss were observed:
  - a). non-breakup mass loss that can be attributed to vaproization, boiling of impurities or trapped helium blowout
  - b). breakup and/or stripping

The first mechanism results in particles ranging in size from molecular diameter to approximately $1\mu m$. The second mass loss mechanism results in particles from submicron to greater than 10 microns in diameter.

APPENDIX A

ANALYSIS OF THE SHEAR STRESS AT THE
LIQUID - SOLID INTERFACE ON A PARTIALLY
MOLTEN MICROSPHERE.

Consider a two dimensional model of the microsphere in an (x, y) coordinate system, where y is measured normal to the surface and x is measured along the surface. Then,

$$x = R\theta \quad (A-1)$$

where θ is the angle measured from the stagnation point and R is the radius of the microsphere. It will be assumed that the gas-phase flow is free molecular so that the pressure and gas phase shear are given by,

$$p = \rho_{\infty} U_{\infty}^2 \cos^2\theta \quad (A-2)$$

$$\tau = \rho_{\infty} U_{\infty}^2 \sin\theta \cos\theta$$

where ρ_{∞} is the free stream density

U_{∞} is the free stream velocity

hence the pressure gradient will be,

$$\frac{dp}{dx} = - \frac{\rho_{\infty} U_{\infty}^2}{R} \sin\theta \cos\theta \quad (A-3)$$

It will also be assumed that a locally similar analysis will be adequate for our purposes; hence x - derivatives will be neglected in comparison with y - derivatives in the neighborhood of any point x, and the local values of the coefficient in the equations will be used. In this case, the Navier - Stokes equations become,

$$\frac{d^2u}{dy^2} = \frac{1}{\mu} \left(\frac{dp}{dx} - F \right) \quad (A-4)$$

where F is the body force and μ is the liquid viscosity.

Consider first conditions in a wind tunnel, where the body force (deceleration) is not present. Using (A-3), equation (A-4) becomes,

$$\frac{d^2u}{dy^2} = - \frac{\rho_{\infty} U_{\infty}^2}{\mu R} \sin\theta \cos\theta \quad (A-5)$$

which may be integrated twice to give,

$$u(y) = - \frac{\rho_{\infty} u_{\infty}^2}{\mu R} \sin \theta \cos \theta \frac{y^2}{2} + c_1 y + c_2 \quad (A-6)$$

The boundary conditions which must be satisfied are:

1. There is no slip at the liquid-solid interface.
2. The shear stress is continuous across the liquid-gas interface.

These may be expressed mathematically as,

$$u = 0 \quad \text{at } y = 0 \quad (A-7)$$

$$\mu \frac{du}{dy} = \rho_{\infty} u_{\infty}^2 \sin \theta \cos \theta \quad \text{at } y = \delta$$

where δ is the liquid layer thickness.

Application of Equation (A-7) to (A-6) results in,

$$\begin{aligned} C_2 &= 0 \\ C_1 &= \frac{\rho_{\infty} u_{\infty}^2}{\mu} \sin 2\theta \left[\frac{\delta}{R} + \frac{1}{2} \right] \end{aligned} \quad (A-8)$$

so that the velocity profile is,

$$\frac{u}{u_{\infty}} = R_e \sin 2\theta \left[(\delta/R + 1/2) y/R - 1/2 (y/R)^2 \right] \quad (A-9)$$

where $R_e = \frac{\rho_{\infty} u_{\infty}^2 R}{\mu}$ is the Reynolds number.

Using this expression, the shear stress will be,

$$\tau = \mu \frac{du}{dy} = \rho_{\infty} u_{\infty}^2 \sin 2\theta \left[(\delta/R + 1/2) - y/R \right] \quad (A-10)$$

and in particular, the value at the liquid-solid interface ($y = 0$) is then,

$$\tau_{wt} = \rho_{\infty} u_{\infty}^2 \sin 2\theta (\delta/R + 1/2) \quad (A-11)$$

The subscript "t" has been used to indicate that this is the value of the wall shear stress which will result under wind tunnel conditions.

For the case where the microsphere is actually in flight, the body force resulting from the deceleration of the particle must be included. The deceleration is the drag per unit mass of the body, and is expressed by,

$$a = - \frac{3}{4} \frac{\rho_{\infty} u_{\infty}^2}{\rho R} \sin \theta \quad (A-12)$$

where ρ is the particle density and the minus sign indicates that the deceleration acts forward.

The body force is given by,

$$F = \rho a \quad (A-13)$$

hence,

$$F = -\frac{3}{4} \rho_{\infty} u_{\infty}^2 \sin \theta = -\frac{3}{8 \cos \theta} \rho_{\infty} u_{\infty}^2 \sin 2\theta \quad (A-14)$$

When this expression is inserted into Equation (A-4) and the integration is performed, the result is,

$$u(y) = \frac{\rho_{\infty} u_{\infty}^2 \sin 2\theta}{\mu R} \left[\frac{3}{8 \cos \theta} - 1 \right] y^2/2 + c_1 y + c_2 \quad (A-15)$$

The boundary conditions remain unchanged so that use of Equation (A-7) gives for the constants,

$$\begin{aligned} c_2 &= 0 \\ c_1 &= \frac{\rho_{\infty} u_{\infty}^2 \sin 2\theta}{\mu} \left[\frac{1}{2} - \left(\frac{3}{8 \cos \theta} - 1 \right) \delta/R \right] \end{aligned} \quad (A-16)$$

and the velocity profile is,

$$\frac{u}{u_{\infty}} = R_e \sin 2\theta \left\{ \left[\frac{1}{2} - \left(\frac{3}{8 \cos \theta} - 1 \right) \delta/R \right] y/R + \frac{1}{2} \left(\frac{3}{8 \cos \theta} - 1 \right) \frac{y^2}{R^2} \right\} \quad (A-17)$$

The wall shear stress can be determined in the same way as for the previous case, hence,

$$\tau_{wf} = \rho_{\infty} u_{\infty}^2 \sin 2\theta \left[\frac{1}{2} - \left(\frac{3}{8 \cos \theta} - 1 \right) \delta/R \right] \quad (A-18)$$

Here the subscript "f" indicates that this is the expression for the shear stress under real flight conditions.

The ratio of the flight shear stress to the tunnel shear stress is then,

$$\frac{\tau_{wf}}{\tau_{wt}} = \left[\frac{\frac{1}{2} - \left(\frac{3}{8 \cos \theta} - 1 \right) \delta/R}{1/2 + \delta/R} \right] \frac{(\rho_{\infty} u_{\infty}^2)_f}{(\rho_{\infty} u_{\infty}^2)_t} \quad (A-19)$$

and this may be rearranged as,

$$\frac{\tau_{wf}}{\tau_{wt}} = \left[1 - \frac{3}{4\cos\theta} \left(\frac{\delta/R}{1+2\delta/R} \right) \right] \frac{w_{ef}}{w_{et}} \quad (A-20)$$

APPENDIX B

LIQUID LAYER STABILITY FOR SMALL αR

It has been shown in Section II-C that the Aerotherm stability criteria for partially molten microspheres is not valid because, in addition to other limitations, it is based on a high αR theory, while in reality αR will be relatively low if δ/r is small. What is required is a theory that is valid for both small αR and small δ/r . To this end, Reynolds' small αR results in Reference B-1 can be used. The neutral stability curve for $\alpha R < 1$ is given as,

$$R = \frac{\alpha^2 - B}{W} - (3/2)S \quad (B-1)$$

$$\frac{6}{5} \left[\frac{\alpha q}{3 + \frac{27\alpha^2}{5}} \right]^2 + \frac{7}{8} \left[\frac{\alpha q}{3 + \frac{27\alpha^2}{5}} \right]$$

where the following definitions are made:

$$B = \frac{3}{4} \frac{\rho_{\infty} U_{\infty}^2 \delta^2}{r\sigma} \cos \theta_0$$

$$W = \frac{\rho_{\infty} U_{\infty}^2 \delta}{2\sigma} \sin 2\theta_0$$

$$S = -2 \frac{\cos 2\theta_0}{\sin 2\theta_0}$$

$$q = \frac{\sin 2\theta_0}{1/2 \sin 2\theta_0} = 2$$

$$R = \frac{U_s \delta \rho}{\mu}$$

For the sake of continuity, the assumption that $\alpha = 2\delta/r$ will be carried over from Reference B-1. The surface speed, U_s , is given by,

$$U_s = \frac{\delta}{\mu} \rho_{\infty} U_{\infty}^2 \left(\frac{1}{2} \sin 2\theta_0 \right) \quad (B-2)$$

so that the Reynolds number can be expressed as,

$$R = W_e \left(\frac{1}{2} \sin 2\theta_0 \right) M (\delta/r)^2 \quad (B-3)$$

where,

$$W_e = \frac{\rho_{\infty} U_{\infty}^2 r}{\sigma}$$

$$M = \frac{\rho \sigma r}{\mu^2}$$

Recalling Figures (II-12) and (II-13), a stability criteria for small δ/r is desired. From Equation B-3, R is of the order of $(\delta/r)^2$ as is α ; hence, when terms of the order of $(\delta/r)^2$ are neglected, Equation B-1 becomes,

$$\frac{\alpha^2 - B}{W} = \frac{3}{2} s \quad (B-4)$$

or,

$$8(\delta/r) \left(1 - \frac{3}{16} W_e \cos \theta_0\right) + 3W_e \cos 2\theta_0 = 0 \quad (B-5)$$

This may be solved for the critical Weber number such that,

$$W_{e \text{ crit}} = \frac{8 (\delta/r)}{3 \left[\frac{1}{2} (\delta/r) \cos \theta_0 - \cos 2\theta_0 \right]} \quad (B-6)$$

Notice that this expression is a function of both the melt layer thickness and position on the microsphere. It is also interesting to note that

$$\lim_{\delta/r \rightarrow 0} W_{e \text{ crit}} = 0$$

so that according to the theory, the thinner layers are less stable than the thicker ones. Equation B-6 also indicates that the critical Weber number decreases as θ_0 increases (for $\frac{\pi}{4} \leq \theta_0 \leq \frac{\pi}{2}$) and has a minimum at

$\theta_0 = \frac{\pi}{2}$. This means that the position of greatest instability is the tangent point. By way of example, if we choose $\delta/r = 1/10$, then $W_{e \text{ crit}}$ ranges from 7.55 at $\theta_0 = \frac{\pi}{4}$ to .27 at $\theta_0 = \frac{\pi}{2}$.

As is the case with the high αR theory, these results suffer from the restriction that $\alpha = 2\delta/r$, i.e., that the disturbance wave is 1/2 the circumference of the microsphere. It was pointed out in Section II-C that this may not be the case, and that shorter waves may be examined by letting $\alpha = \frac{2n\delta}{r}$. If this is done, the expression for the critical Weber number becomes,

$$W_{e \text{ crit}} = \frac{8 n^2 (\delta/r)}{3 \left[\frac{1}{2} n^2 (\delta/r) \cos \theta_0 - \cos 2\theta_0 \right]} \quad (B-7)$$

From this equation, it can be seen that the critical Weber number is unchanged at $\theta_0 = \frac{\pi}{4}$ but is increased for $\theta_0 > \frac{\pi}{4}$. Once again, the shorter waves are more stable than the longer ones.

REFERENCES

- B-1 Reynolds, W. C., "Hydrodynamic Stability of the Liquid Layer on a Small Object in High Altitude Re-entry", SC-CR-67-2631, 1967