176 pg
11/25/68

538

SC-RR-68-547

October 1968

MASTER
**AEROSPACE
NUCLEAR
SAFETY**

THERMAL AND ABLATION RESPONSE
OF SMALL SPHERICAL PARTICLES IN
REENTRY--SUMMARY REPORT

B. W. Marshall, 9511

AEROSPACE NUCLEAR SAFETY DEPARTMENT 9510

SANDIA LABORATORIES

OPERATED FOR THE UNITED STATES ATOMIC ENERGY COMMISSION BY SANDIA CORPORATION | ALBUQUERQUE, NEW MEXICO; LIVERMORE, CALIFORNIA

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Issued by Sandia Corporation,
a prime contractor to the
United States Atomic Energy Commission

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

October 1968

SC-RR-68-547

THERMAL AND ABLATION RESPONSE
OF SMALL SPHERICAL PARTICLES IN
REENTRY--SUMMARY REPORT

B. W. Marshall, 9511
Sandia Laboratories, Albuquerque

Approved by:

L. A. Hopkins, Jr.
L. A. Hopkins, Jr., 9500

V. E. Blake
V. E. Blake, 9510

ABSTRACT

The reentry response of small spherical particles includes the thermal response of the particle and the melting of and mass loss (ablation) from the body. Results are presented of (1) theoretical calculations to determine the conditions of melt for release in reentry, (2) analytical studies to determine liquid layer and liquid droplet stability conditions in reentry, and (3) experiments on small particle ablation conducted in a hyperthermal arc tunnel, a hypersonic shock tunnel, and a free-flight gun range.

The cutoff date for this information
in this report is September 15, 1968.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:
A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.
As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Blank Page

SUMMARY

The use of nuclear materials in SNAP systems for space applications requires a knowledge of the potential safety hazards resulting from the use of the radioactive material. One area of potential hazard is the reentry response of the material in an aborted mission. Extensive effort was extended in investigating the reentry response of small (50 to 250 micrometers diameter) spherical particles (microspheres) of plutonium dioxide.

As a result of extensive calculations of the reentry thermal response of microspheres, a melt envelope was generated. This melt envelope defines the combinations of reentry release altitudes and velocities which result in melting of the particles. Estimations, based on no shape change during melting, of conditions which result in 50 and 100 percent melt of a microsphere are presented.

To define the mass loss rate following melting of a microsphere and the size distribution of any resulting debris, theoretical studies of liquid layer and liquid droplet instabilities were initiated. An analysis by Aerotherm Corporation found that the critical Weber number (ratio of dynamic pressure forces to surface tension forces) for mass loss decreased as the thickness of the liquid layer decreased and that critical Weber numbers near and below 1 were possible for the configuration considered in their analysis. In contrast, General Electric/Valley Forge Space Technology Center found that the critical Weber number for a partially molten sphere is greater than that for a totally molten sphere.

Experimental investigations of the thermal and mass loss characteristics of small spherical particles were performed using plutonium dioxide in a hyperthermal arc tunnel, iron oxide in a hypersonic shock tunnel, and gold in a free-flight gun range. The arc tunnel experiments resulted in mass loss at Weber number conditions near unity. Analysis of the impact samplers used in the experiments indicated that the debris generated during the exposures can be characterized as being from less than 0.01 micrometer to approximately 14 micrometers in diameter. The results of the experiments done in the Cornell Aeronautical Laboratory

hypersonic shock tunnel suggest that the critical Weber number for breakup of the partially molten particles is higher than the number for breakup of fully molten particles. For the gold microspheres fired in the ACDRL free-flight light gas gun, debris data were obtained for mass loss during flight from partially and fully molten particles for Weber numbers from near 7.5 to approximately 45. Attempts are made to interpret the data from both analytical and experimental sources, with areas of inconclusive data pointed out.

ACKNOWLEDGMENTS

In a program of the scope reported in this report many different organizations and people made contributions. The cooperation and assistance provided by G. P. Dix and his associates in SNS were essential. Many people were involved in the arc tunnel experiments. H. L. Hodges, J. F. Hudson, M. D. DeVore, and H. Romero provided characterization of the material tested; R. E. Elrick performed analyses to determine the efficiencies of the arc tunnel samplers and, together with B. T. Kenna, was responsible for the debris samplers in the free-flight gun range experiments; I. B. White and K. L. Romine were responsible for the operation of the arc tunnel. L. D. Tyler provided assistance in the liquid instability analyses. Finally, H. E. Hansen provided many helpful suggestions in the overall program.

TABLE OF CONTENTS

	<u>Page</u>
LIST OF SYMBOLS	6
INTRODUCTION	7
ANALYTICAL STUDIES	9
Thermal Response	9
Mass Loss	12
EXPERIMENTAL STUDIES	18
Arc Tunnel Experiments	18
Flow Calibration	21
Test Material	27
Test Conditions and Results	38
Ablation Debris	51
Discussion of Arc Tunnel Results	61
Study of Breakup of Partially Molten Spheres in a Hypersonic Shock Tunnel	69
Test Equipment and Instrumentation	70
Test Material	71
Test Procedure and Conditions	73
Test Results	77
Discussion of Results--Fully Molten Spheres	90
Discussion of Results--Partially Molten Spheres	94
Conclusions	99
Small Particle Ablation Experiments in a Free-Flight Gun Range	100
Discussion of Gas-Gun Experiments	110
DISCUSSION OF RESULTS	120
APPENDIX A -- Sample Calculation for Heat Flux to a Microsphere in the Hyperthermal Arc Tunnel	125
APPENDIX B -- Estimate of Mass Loss by Vaporization in Arc Tunnel Experiments	133
REFERENCES	137

LIST OF SYMBOLS

A - area, ft^2
C - specific heat
d - diameter, feet
F - heat of fusion, Btu/lb
Fr - Froude number = V^2/gR
g - 32.2 ft/sec^2 , constant
H - enthalpy, Btu/lb
L - length, feet
Le - Lewis number
M - fluid property parameter, $\rho\sigma R/\mu^2$
Pr - Prandtl number
 P_{t0} - nozzle plenum pressure, atmosphere
 P_{t2} - model pressure, atmosphere
q - heat flux, $\text{Btu}/\text{ft}^2\text{-sec}$
R - radius, feet
Re - Reynolds number
S - surface tension, lb/ft
T - temperature, $^{\circ}\text{R}$
t - time, seconds
U - volume, ft^3
V - velocity, fps
W - Weber number = $\rho V^2 R/S$
 ϵ - surface emissivity
 ρ - density, lb/ft^3
 σ - Stefan Boltzman constant
 μ - viscosity, $\text{lb}/\text{ft-sec}$

Subscripts

e - edge of boundary layer
s - total value
w - evaluated at wall temperature
 ∞ - free stream conditions
F - furnace condition

THERMAL AND ABLATION RESPONSE OF SMALL SPHERICAL PARTICLES IN REENTRY--SUMMARY REPORT

INTRODUCTION

In the use of nuclear power supplies in space, one of the problems that must be studied is the potential health hazard created by the use of radioactive fuel. Several different types of safety problems exist; however, the only one considered in this report is the reentry of the fuel. Generally, the fuel is located within one or more containers, and the system may be designed either to burn up during reentry or to reenter intact. In the first case, the fuel is intentionally exposed and dispersed in the atmosphere; in the latter case, only an unexpected failure of the container will result in exposure of the fuel. In either case, a knowledge of the fuel response in the reentry environment is desired.

If a body enters the earth's atmosphere with sufficient velocity, the kinetic energy will result in partial or total melt and/or vaporization of the body. Once a body melts interest centers on the breakup of the melt into smaller size particles, called debris. It is the characteristics of this debris that are desired. If one could express the quantity and size distribution of this debris as a function of the release conditions, calculations could be made of the dispersion, fallout, and other atmospheric phenomena that affect the quantity of debris deposited at the earth's surface. It is the deposited debris which is potentially a hazard.

To be able to accurately predict the final state of the fuel, several interdependent analyses must be performed. The aerodynamics of the body must be analyzed and expressed in relationships from which parameters such as flow regimes, body forces, time of flight, deceleration, aerodynamic heating, and trajectory can be determined. One can then take the trajectory and the aerodynamic heating information obtained

from the aerodynamic analyses and apply these to the thermal response of the reentering body. From the thermal analyses, one can determine under what conditions a reentering object will melt.

Once a body undergoing reentry melts, describing the resultant melt becomes of primary concern. An analytical treatment of this phenomenon is a more complex problem, and, in fact, the author knows of no theories which will describe mass loss by discrete particles of mass from a melting body. Vaporization can be treated and is included in the thermal response portion of the overall problem.

A combined theoretical and experimental program was initiated by Sandia Laboratories' Aerospace Nuclear Safety Department to investigate the complete thermal and ablative response of spherical particles in the reentry environment. The purpose of these studies was to develop an analytical technique, verified by experimental data, which would allow one to confidently predict the temperature response and mass loss from small spherical particles for a wide range of reentry environments.

ANALYTICAL STUDIES

Thermal Response

Stephenson^{1,2} performed the original analyses at Sandia on the thermal response of small diameter (50 to 250 micrometer) spherical particles. A spinning microsphere with its point mass attaining a uniform temperature throughout was initially considered. Stephenson later improved the calculations to include nonspinning (stable) reentry with finite thermal conductivity for the material but with no property variation with temperature or vaporization losses. In both of these analyses, a parametric velocity-altitude study was performed, and as a result a reentry region in which release would cause particle melting was produced. The resulting "melt envelopes" are shown in Figure 1. It can be seen that the assumptions used in the nonspinning reentry resulted in a definite increase in the region of release where stagnation point melting would occur.

Figure 1. Release conditions for melting of stable and spinning spheres

As an outgrowth of the nonspinning microsphere model, the computer program used to calculate the reentry thermal response was improved. The program presently being used, SPHERE 2, is essentially a numerical approximation technique for the general unsteady-state Fourier heat conduction equation. However, restrictions placed on the computer calculations are such that the results should represent a very close approximation to the closed form solution. The sphere, which can be of any radius of interest to the present Aerospace Nuclear Safety Program, is divided into 40 equal volume nodes. A network is set up throughout the sphere such that heat is transferred between all connecting nodes as well as stored in all nodes according to their specific heat properties. All surface nodes radiate energy from the body according to the fourth power temperature law and the surface emissivity value. Variable material properties are included in the calculations as is the material heat of fusion. The program also accounts for the vaporization from the surface of the body according to a relationship given by Chapman.³

The aerodynamic heating at the stagnation point is calculated according to a relationship given in Reference 4 for the continuum regime, by kinetic theory relationships in the free molecular regime, and by a bridging relationship given by Chapman³ for the transition regime. The heating distribution about the body has been well established by Lees⁵ in the continuum flow regime and is taken as a cosine distribution in the free molecular regime. A bridging relationship is used for the transition regime. The microsphere is considered to fly in a stable orientation. With velocity, gas density, particle size, drag coefficient, and material properties as input data, the computer will calculate velocity, stagnation heat flux, Weber number, displacement, and the temperature of each of the 40 nodes as a function of time. Details of this program can be found in Reference 6.

A parametric study of release velocity and altitude was performed using SPHERE 2 to determine a revised melt envelope. The results of this study are shown in Figure 2. Comparison of this melt envelope with Stephenson's nonspinning model shows very little difference in the results. The results of these thermal calculations are thought to be very reliable until the particle begins to melt. Once substantial melting occurs (> 10 to 15 percent), the program is in error in that the shape change and mass redistribution and/or loss cannot be accommodated. In all calculations, the particle is assumed to remain spherical.

Figure 2. Microsphere response in reentry

Shown on Figure 2 are curves for 50 and 100 percent melt for 250- and 50-micrometer diameter particles. Because of the previously mentioned lack of accommodation of shape change, these results should be considered only as approximate guides to the actual events.

Concurrent with the development of the Sandia computer analyses, studies were conducted by General Electric/Valley Forge Space Technology Center (GE) and by Aerotherm Corporation (AC) under Sandia contracts. A portion of the overall purpose of these studies was to provide an independent check on the Sandia melt envelope. Complete reports on these studies were published,^{4,7,8} and only the results of the thermal analyses are discussed here.

Four release conditions, designated 1, 2, 3, and 4, are shown on Figure 2. All four conditions were given to Aerotherm, while GE considered only Conditions 1 and 2. Both Aerotherm and GE concluded that release at point 1 would result in melting for nontumbling reentry.

This point lies on the edge of the Sandia melt envelope, and the Aero-therm/GE results are consistent with this. Condition 2 release lies well within the melt region, with both Aerotherm and GE agreeing that melting would result. The results of the Aerotherm analyses of Conditions 3 and 4 do not contradict the Sandia results. Condition 3 was a release condition near the edge of the melt zone, and Aerotherm concluded that 50-micrometer particles would totally melt and 250-micrometer particles would partially melt. Condition 4 represented a release outside the melt zone, and Aerotherm showed no melt for 250-micrometer particles and melting only at the stagnation point for 50-micrometer particles. These results are considered consistent with the Sandia melt envelope and provide confidence in the program being used to calculate the reentry thermal response.

Included in the Aerotherm analyses was a calculation to determine the "thermal stopping time" for the reentry heating of the microspheres. This is defined as the length of time during reentry where the heating is sufficient to overcome all losses and maintain the temperature at or above melting. This can be calculated by determining the point in reentry where the radiation loss at the particle melting temperature becomes equal to the aerodynamic heating. In all calculations, the particle was assumed to remain spherical. Aerotherm found that in general the thermal stopping time was an order of magnitude longer than the time required to completely melt the particle. In effect, much excess heat is available. The point that can be made from this is that the effect on the heating that could result from the shape change of the particle must be very great. One might then suggest that since any excessive shape change will occur only after substantial melting has occurred, this effect may not be sufficient to counter the total melt conclusion.

Mass Loss

The author does not know of any complete analytical treatment of molten material in the reentry environment. Various studies and experiments have been performed regarding conditions under which wholly molten particles will break up. Luna and Klikoff⁹ surveyed the data available on this subject and found that breakup was generally correlated as a

function of Weber number. Weber number is defined as the ratio of the dynamic pressure acting on a body to the surface tension forces of the body. This relationship is normally expressed as

$$W = \frac{\rho V^2 / 2}{S/d} = \frac{\rho V^2 R}{S}.$$

Breakup data have been correlated as a function of critical Weber number. If the flight Weber number exceeds some critical value, the particle will break up; if the number is not exceeded, the liquid particle is stable. However, the survey showed that critical Weber numbers from 1 to 10 have been reported in the literature. When applied to the case of the reentry of small particles, this range of uncertainty provides results which range from little or no mass debris formed to the total fuel inventory released as debris. As a result of this variation and since the thermal calculations indicate that a partially molten particle (a liquid layer on the solid substrate) could exist in the reentries of interest, further study was undertaken.

Sandia initiated study contracts with Aerotherm and GE to study the stability of totally molten spheres and of a liquid layer when they are acted upon by forces typical of the reentry conditions of present interest. Details of these studies are found in References 4, 7, and 8, and only a summary and the conclusion of the studies are presented here. The release conditions given to Aerotherm and GE are given in Table I.

TABLE I
Release Conditions Given to Aerotherm and GE

Condition	Diameter (μ m)	Velocity (fps)	Altitude (ft)	Comments
1	50 250	25,200	244,000	Both Aerotherm & GE
2	50 250	21,000	181,000	Both Aerotherm & GE
3	50 250	16,500	200,000	Aerotherm only
4	50 250	12,000	118,000	Aerotherm only

In performing the stability analysis, GE considered both a wholly molten particle and a liquid layer configuration. For the wholly molten particle, GE considered the dynamic pressure acting on the front of the particle; this pressure causes the particle to flatten, inflate in the manner of a parachute, and finally burst. An analysis of the particle when acted upon by the dynamic forces, inertial forces, surface tension forces, and retarding forces resulting from viscosity resulted in a critical Weber number of 2 for the free molecular conditions of interest.

For the liquid layer analysis, GE analyzed the situation of constant liquid layer thickness on a body of such proportions that the radius of curvature was neglected. Among other things, the shear stress at the liquid/gas interface was assumed to be zero whereas body forces and external pressure gradients were included. Physically, this model might be said to best represent the growth or decay of waves in the liquid layer at the stagnation region. The results of these analyses showed that the stability criterion for the liquid layer was given approximately as

$$\frac{Fr}{W} \geq \frac{1}{\pi}.$$

Solving this for the case of interest (i.e., small particles in free molecular flow) revealed

$$W(\text{liquid layer}) \simeq 4.18.$$

Based on these analyses, a trend of increasing stability for decreasing percentage of melt can be supported. However, it should be remembered that many approximations were made in solving the equations and that the analyses represent only first approximations. Based on the results of these analyses, GE concluded that small particles, when released at the lower altitudes and higher velocity combinations within the melt zone, become molten relatively quickly and will lose mass by means of particle breakup.

In stability analyses of reentering, melting bodies, Aerotherm Corporation also pursued two separate problems: (1) an investigation of the stability of equilibrium liquid drops in free-molecule flow, neglecting surface shear, and (2) a study of the stability of a liquid

layer subjected to an impinging free-molecule flow. In the former case, the Aerotherm model assumed constant surface tension, uniform liquid density, zero surface shear with a nonuniform but axisymmetric external pressure field, and an acceleration in the direction of the resultant pressure force equal to the integrated normal force divided by the drop mass. This analysis in effect considers only a Taylor instability and neglects others, such as Kelvin-Helmholtz and Craik-Benjamin instabilities. The Aerotherm report points out this is probably invalid for the very high speed flow situations of interest but does delineate a region of interest for instabilities. The results of this analysis are shown as Figure 3 which is taken directly from Reference 4.

Figure 3. Stability conditions for liquid PuO_2 drops

Calculations for peak decelerations for both 50- and 250-micrometer particles, for the release conditions shown in Table II, when compared with Figure 3 indicate that all release conditions except number 4 for a 250-micrometer particle are stable. Moreover, thermal calculations show no melt for this particular release, and one can conclude from these results that all considered release conditions which cause particle melting are insufficient to cause breakup of the molten droplet.

TABLE II
Release Conditions

Condition	Injection Altitude (ft)	Injection Velocity (fps)	Peak Deceleration (ft/sec ²)	
			250 μ m	50 μ m
1	244,000	25,200	0.6×10^4	3.1×10^4
3	200,000	16,500	1.3×10^4	7.7×10^4
2	181,000	21,000	3.5×10^4	3.0×10^5
4	118,000	12,000	1.0×10^5	6.5×10^5
Critical deceleration (ft/sec ²)			0.9×10^5	2.35×10^6

For the latter part of the analysis the problem was formulated by Reynolds,⁸ with the following principal assumptions: a flat, liquid layer lies over a solid substrate; the liquid viscosity, density, and surface tension are constant; the external flow field is free molecular, having an arbitrary angle of impingement; and a body force exists in the direction opposite the flow. Reynolds⁸ performed the analysis and postulated that a disturbance wavelength on the liquid layer of one-half the particle circumference is the greatest that can be supported. Using these assumptions, an equation for stability can be written as

$$\frac{M^{0.3} W^{0.7}}{\left(1 - \frac{3}{16} W\right)} 0.4 < 2 \quad \text{for stability} \quad (2)$$

where

M = dimensionless fluid property parameter, $\frac{\rho S R}{\mu^2}$

W = Weber number based on particle radius, $\frac{\rho v^2 R}{S}$.

The above equation is plotted on Figure 4. The conditions for reentry release given in Table I are also plotted on this figure, and it is seen that all but two conditions indicate an unstable situation. However, it should be remembered that this relationship was derived for a flat plate and the boundary conditions mentioned previously. The use of this relationship for spherical particles in reentry requires certain extrapolations which, because of the lack of experimental data, cannot be verified.

Figure 4. Stability regimes for a liquid layer in free-molecule flow

EXPERIMENTAL STUDIES

Arc Tunnel Experiments

To study the response of actual fuel material (plutonium-238 dioxide microspheres), Sandia obtained a 40-kilowatt arc tunnel facility together with a glove box encompassing the test chamber and generator. Hyperthermal arc tunnels have been used successfully for some time to study the heating, melting, and ablation phenomena that occur when a body is exposed to the atmosphere at high velocity. The arc tunnel permits enthalpy and pressure combinations which are typical of a wide range of reentries, and the response of the model can be photographed with movie cameras to detail the response of the surface during melting. However, it should be remembered that a simulation of the complete reentry environment cannot be produced in this laboratory facility. In the arc tunnel, reentry deceleration forces on a body are not reproduced, local flow conditions away from the stagnation region are not necessarily reproduced, and the variable reentry pulse is not reproduced. In addition, various nitrogen and oxygen species, in addition to slight electrode contamination, are present in the gas flow. Nevertheless, when these limitations are properly recognized, the arc tunnel is one of a very limited number of experimental test facilities which can provide any degree of reentry ablation simulation. In most cases, the lack of complete simulation requires one to be conscious of and allow for the nonsimulated forces which are of importance in a particular study and to be cautious in extending the resulting data. A schematic of the arrangement of the arc tunnel facility used in these experiments is shown in Figure 5. The design has evolved somewhat in use and is shown as it exists at this time.

The experiments are conducted by placing a single microsphere on the tip of a conical sting of special design; the sting is placed in the centerline of the 1/2-inch-diameter stream. The microspheres are exposed to the supersonic high temperature gas flow for preselected periods of time by means of an electrically controlled water-cooled shield. Nitrogen, which is passed through the dc arc, and oxygen, which is mixed in the plenum in the proper proportion to obtain reconstituted

air, are expanded through the converging-diverging nozzle to provide supersonic flow in the test stream. A partial vacuum (a few millimeters) is maintained in the test chamber to provide properly expanded flow in the nozzle. The initial sting was a 200- μm diameter hypodermic needle on which concially shaped ablation material was placed to prevent needle melting. As a design improvement, the hypodermic needle was replaced with a fabricated, tungsten conical needle with a smaller cone angle and a smaller 160- μm diameter tip. To retain the microspheres on the needle, a very small amount of epoxy cement was placed on the needle tip, the needle was touched to the microsphere, and a vacuum was drawn on the microsphere through the hole in the needle. The combined vacuum and epoxy were sufficient to center the particle on the needle and to retain it during most test exposures.

Figure 5. Schematic of arc tunnel facility

A high speed camera with 1000 cps timing marks placed on the film was used to record the response of the microsphere during heating and ablation. Initially, the camera was operated at 500 frames per second, but the speed was increased in later tests to 6000 frames per second. By placing the lens of the camera close to the microsphere and focusing the image on a plane many times further from the lens, enlarged-image photographs were generated. Accurate real time resolution of events recorded in enlarged detail on the film was possible. To determine time zero in the test, a mark (high-intensity lamp) indicating when the flow deflector was retracted was placed along the edge of the film on the opposite side from the timing marks.

Originally, attempts to sample debris generated by the ablation of a microsphere consisted of placing a thin 0.75-inch diameter copper disc approximately 0.75 inch behind the microsphere. In subsequent modifications to the test equipment, the sampler was placed approximately 4 inches behind the microsphere. All samplers used to date were basically impact samplers, with the several designs used consisting of the original 0.75-inch diameter disc with eight 3/4-inch long by 1/8-inch wide arms extending from the disc, solid 3.5-inch diameter thin copper plates, and 3.5-inch diameter thin copper plates with sixty-four 3/16-inch diameter holes placed symmetrically on the plate. All designs successfully collected debris, but because of the unknown efficiencies of the impact samplers, no mass balance can be determined directly from the data. However, extensive effort was devoted to determining the quantity and size distribution of the debris collected on the samplers, and these data are discussed in a later section.

The hazardous nature of the material tested placed certain constraints on the test procedure. Procedures formulated with the cooperation of Sandia Health Physics personnel were used in all test operations. All microspheres were individually characterized, as is discussed later, and were brought into the test area in individual containers. To perform an experiment, the particle was removed from the container, mounted on the sting, and centered very carefully in the test stream. The camera was aligned, focused, and loaded with film. The arc tunnel was started and adjusted to the predetermined conditions with the flow deflector remaining in front of the microsphere. Once the test conditions were established, the camera was started, the deflector withdrawn and

returned, and the camera stopped by engaging a switch on the control console. The debris sample was removed for analyses, as were the remains of the microsphere, when possible. Figure 6 shows the operational facility.

Figure 6. View of the hyperthermal tunnel

Flow Calibration

To determine the flow conditions in the stream and the resulting heat flux on a particle, considerable effort was expended in performing calibration measurements in the arc tunnel. Generally, a heat flux calorimeter the same size and shape as the model to be tested is placed in the stream, and the heat flux is measured directly. However, because of the extremely small size of the microspheres tested, this was not feasible. Therefore, calorimeters differing in size and shape from the

microspheres were used, with the result that the heat flux to the microspheres was calculated from the calorimeter data and appropriate relationships.

To determine the stream conditions, a blunt-nosed 0.125-inch diameter copper calorimeter and a water-cooled pitot probe were used. The calorimeter operates on a transient basis and can be used to measure the stagnation heat flux on the calorimeter (not the microsphere). The theory underlying the response of this type of calorimeter is explained in detail by Sonnenschein and Winn.¹⁰ The temperature response of the copper measuring slug in the calorimeter was measured with a chromel-alumel thermocouple and used in the appropriate equations to calculate the stagnation heat flux on the body.

The stagnation heat flux on a body is related to the gas enthalpy and model total pressure by

$$q\sqrt{R} = 0.039\sqrt{Pt_2}(H_s - H_w) \quad (3)$$

which is derived from the Fay-Riddell analysis. For these studies, R was taken as four times the geometric radius of the flat-faced blunt body. With the quantity q measured by the calorimeter, $q\sqrt{R}$ can be calculated for each condition. The model total pressure, Pt_2 , was measured directly by the water-cooled pitot.

Using these data, the total gas enthalpy for a cold wall condition (i.e., $H_w = 0$) was calculated as

$$H_s = q\sqrt{R}/0.039\sqrt{Pt_2}. \quad (4)$$

This quantity establishes the enthalpy of the gas at the centerline of the stream in contrast to an enthalpy calculated by a heat balance on the system. The latter method gives only a bulk, average enthalpy and does not recognize any gradients in the stream cross section. Since gradients are known to exist, all references to enthalpy in this report will be to values determined from the calorimeter data. Table III gives the calorimeter and pitot data for each test condition together with the gas enthalpy calculated by the method described above. The tolerance

values reflect the scatter in data resulting from the numerous measurements made at each test condition.

TABLE III
Flow Conditions Measured by Copper Calorimeter

Condition	Stagnation Heat Flux, q (Btu/ft ² -sec)	$q\sqrt{R}$	Pt ₂ (atms)	H _s (Btu/lb)	Pt ₀ (atms)	P _{chamber} (atms)
A	675 ± 70	97 ± 10	0.0485	11,000	0.31	0.0055
B	750 ± 70	108 ± 10	0.0737	10,000	0.40	0.00619
C	600 ± 70 - 100	87 ± 10 - 15	0.0593	9,500	0.35	0.0048
K	1110 ± 100	160 ± 10	0.0987	13,000	0.55	0.0088
M	1700 ± 150	245 ± 20	0.276	12,000	1.15	0.0138
L	145 ± 20	21 ± 3	0.0237	3,600	0.49	0.0019

Because of the nature of the flow in an arc tunnel and the potential effect of this flow on the heating to various types of surfaces, copper calorimeters with the front surfaces coated with other materials were also used for heat flux measurement purposes. Experiments previously reported in the literature have shown that certain types of surfaces will reduce the heat flux to a body in dissociated, ionized flow conditions. As classes of materials, metals are reported to be catalytic to gas recombination, silicon oxide is reported to be noncatalytic, and metal oxides and ceramics are reported to fall in between these limits. Since the fuel material is a metal oxide, the coated calorimeters were deemed necessary.

At each test condition, repeated heat flux measurements were made with an uncoated copper calorimeter, with a calorimeter coated with silicon oxide, and with a calorimeter coated with plutonium dioxide. Pitot pressure measurements were also made for each calorimeter measurement. Since the conditions in the facility were repeated for all calorimeters, the effect on the heat flux to the calorimeter was attributed directly to the surface material. An analysis showed that a micrometer thickness of the coating material would have an insignificant effect on the calorimeter thermal response.

Table IV shows the measured heat flux data for each test condition for all three calorimeters. It can be seen that coating the calorimeter surface with either silicon oxide or plutonium oxide significantly reduces the heat flux to the surface of a body. The tolerances shown in the table reflect the scatter in the measured data. Reductions of 30 to 40 percent in heat transfer were effected by coating the surface with the nonmetallic materials. The data show clearly that the gas in the test stream contains some amount of disassociated species. The data also suggest that, for these conditions, plutonium dioxide reduces the surface heat flux in approximately the same proportion as the silicon oxide, a reported noncatalytic material. The conclusion drawn from these data, as it relates to this microsphere program, is that the plutonium dioxide acts as a noncatalytic surface in the arc tunnel flow.

TABLE IV
Effect of Surface Coating
on Heat Flux to Calorimeter

Condition	$q \sqrt{R}$ Copper	$q \sqrt{R}$ SiO	$q \sqrt{R}$ PuO ₂	Percent Reduction	
				SiO	PuO ₂
A	97 ± 10	60 ± 7	60 ± 3	38	38
B	108 ± 10	74 ± 7	65 ± 2	32	40
C	87 ± 10 - 15	61 ± 3	62 ± 10	30	29
K	160 ± 10	120 ± 13	119 ± 15	25	25
M	245 ± 20	193 ± 7	---	21	--
L	21 ± 3	---	20 ± 2	--	5

No calorimeter as small as the microsphere could be obtained, and it was necessary to rely on calculations to determine the stagnation heat flux on the microsphere. To do this satisfactorily, several phenomena which affect the heat transfer to the surface must be considered. Some of these phenomena are: corrections for noncontinuum flow; real gas effects of the test gas, including dissociation and ionization; catalytic effects of the surface of the plutonium dioxide microspheres; and nonequilibrium flow effects in the supersonic nozzle.

Calculations which account for these effects in some manner are made for the heat flux. To do this, it is necessary to make assumptions which are based on limited information available in the literature. A common method of expressing the heat flux at low Reynolds number (transition and free molecular flow) is with the expression

$$\frac{q}{q_{BL}} = f(Re). \quad (5)$$

To calculate the boundary layer heat flux, the equation of Rosner¹¹ is used:

$$q_{BL} = 0.763 \left(\frac{\rho_w \mu_w}{\rho_e \mu_e} \right)^{0.1} \left(\beta \rho_e \mu_e \right)^{0.5} (Pr)^{-0.6} \Delta H \left\{ 1 + \phi [Le^{0.6} - 1] \frac{\Delta H_c}{\Delta H} \right\} \quad (6)$$

where

ΔH_c = enthalpy of dissociation

ΔH = enthalpy difference across boundary layer =

$$(H_s - H_w) - (1 - \phi) \Delta H_c$$

ϕ = extent of recombination at surface

β = inviscid velocity gradient at nose =

$$\frac{V_\infty}{R} \left\{ \frac{\rho_\infty}{\rho_2} \left[2 - \frac{\rho_\infty}{\rho_2} \right] \right\}^{0.5}.$$

To use this equation, the properties of the gas after it expands through the nozzle must be determined, and a computer program was written which accomplishes this.¹² The program calculates these properties for both local thermodynamic equilibrium and for the composition frozen at the nozzle plenum composition. These two limits represent the extremes in the possible variation of properties, and it is reasonable to expect that the actual properties will lie within these bounds. Literature available suggests that, at the pressures typical of these experiments, frozen calculations best reflect data measured. Table V gives

the calculated flow conditions after expansion through the supersonic nozzle for both frozen and equilibrium flow for all conditions except L.

TABLE V
Calculated Properties in Test Stream

Condition	Frozen				Equilibrium			
	Velocity (ft/sec)	Density (lb/ft ³)	Static Pressure (atms)	Static Temperature (°R)	Velocity (ft/sec)	Density (lb/ft ³)	Static Pressure (atms)	Static Temperature (°R)
A	13,120	2.03×10^{-5}	0.00167	2085	14,500	1.83×10^{-5}	0.00569	8910
B	12,840	2.69×10^{-5}	0.0022	2156	14,160	2.43×10^{-5}	0.00716	8760
C	12,600	2.44×10^{-5}	0.00196	2160	13,870	2.21×10^{-5}	0.00627	8570
K	14,000	3.13×10^{-5}	0.00275	2070	15,500	2.79×10^{-5}	0.00977	9460
M	13,760	6.13×10^{-5}	0.00545	2200	15,180	5.59×10^{-5}	0.0188	9470

From these calculated conditions and others, such as viscosity, the stagnation heat flux conditions for each test condition were calculated as shown in Appendix A; the test conditions are shown in Table VI.

TABLE VI
Test Conditions

Condition	Enthalpy (Btu/lb)	Model Pressure (atms)	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

Some limited work, detailed in References 13 through 17, has been done on estimating and measuring the nonequilibrium gas and the non-catalytic surface effects for arc tunnel flow conditions. A general conclusion of these papers is that arc tunnel flow is in nonequilibrium,

particularly for densities and enthalpies typical of these experiments. It is concluded that noncatalytic surfaces can significantly affect the surface heat flux. Wethern¹⁵ states that a calculation based on completely frozen nonequilibrium flow with a noncatalytic surface best represents data taken in his experiments. Carden¹³ and Windler, et al,¹⁶ both report a reduction of heat flux approaching 2 for the use of oxide surfaces in the stream. Based on this information, the heat flux to the plutonium dioxide surface was based on frozen flow through the supersonic nozzle and a noncatalytic surface.

Test Material

The material tested in these experiments was supplied by Monsanto Research Corporation's Mound Laboratory at Miamisburg, Ohio, and was in the form of plutonium dioxide microspheres. Initially, a limited amount of $^{239}\text{PuO}_2$ was tested, but by far the most effort was spent on $^{238}\text{PuO}_2$ and only the latter material will be discussed at length in this report. The initial shipment of $^{238}\text{PuO}_2$ microspheres was received from Mound Laboratory in September 1966. The shipment was divided into two samples: one sample was to contain 1.980 grams of $^{238}\text{PuO}_2$ microspheres, processed, cleaned, etc., in the exact manner as material to be loaded into SNAP-19 and SNAP-27 capsules and was to be representative of the production particle-size distribution; the second sample was to consist of 0.020 gram of $^{238}\text{PuO}_2$ as above, except that the particles were all to be approximately 250- μm diameter, i.e., diameters were to range from about 220 to 300 μm . After receipt of the material, personnel in the Sandia Aerospace Nuclear Safety Department prepared the individual microspheres for the experiments. The particles were cleaned of any surface fines by an ultrasonic cleaning system. The criterion for clean particles was that filters through which aliquots from three successive washings of the particles had been passed would monitor zero counts on an alpha counter. Figure 7 shows the general appearance of the 20-mg sample from which the microspheres for arc-tunnel testing were taken.

a.

Figure 7. Photomicrographs of $^{238}\text{PuO}_2$ microspheres from 20-mg sample (50X)

c. Same view as b. but focused
at a different depth

d.

Figure 7. (continued)

e.

Figure 7. (continued)

Once the particles were determined to be free of surface fines, each microsphere to be used in the arc-tunnel experiments was individually sized, weighed, characterized as to surface texture and sphericity, and photographed. These data, together with the resulting calculated average density, are shown in Table VII. In April, 1967, Sandia received a second shipment from Mound Laboratory and performed similar cleaning and characterization operations. The data on these microspheres are also shown in Table VII and are designated by an M prefix.

A limited number of particles were randomly selected from the cleaned and sized microspheres and used for metallographic cross sections. These cross sections revealed randomly sized, irregularly shaped voids scattered throughout the microsphere. Typical photomicrographs are shown in Figures 8 through 13. Qualitatively, the photomicrographs appear to confirm the calculated gross density to be lower than the theoretical value. Details of the microsphere cleaning procedure and the preparation procedure for the microspheres prior to cross sectioning are given in Reference 18.

TABLE VII

Data on Microspheres
for Arc Tunnel Experiments

Particle Number	Size (μm)	Weight (μg)	Density (g/cc)
1-24-6	226.8	66	10.3
1-24-7	225.4	65	10.8
1-24-8	234.4	69	10.2
1-24-12	221.1	57	10.1
1-24-14	235.4	71	10.4
1-24-15	253.5	75	8.8
1-24-17	235.9	70	10.2
1-24-19	221.1	59	10.4
1-25-1	216.3	60	11.3
1-25-1	247.8	76	9.5
1-25-3	234.4	69	10.2
1-25-5	230.0	65	10.2
1-25-9	224.9	53	8.9
1-25-16	254.0	74	8.6
1-25-20	253.0	87	10.3
M-2	230.9	63.0	9.8
M-3	217.9	51	9.4
M-4	149.4	19.0	10.9
M-6	223.6	56.1	9.6
M-10	225.2	53.8	9.0
M-15	235.3	77.0	11.3
M-16	220.9	60.0	10.6
M-22	218.9	61.7	11.2
M-29	235.4	66.8	9.8
M-31	217.6	56.4	10.4
M-32	227.0	51.0	7.4
M-33	220.4	57.6	10.3
M-35	222.7	63.3	10.9
M-37	227.2	65.3	10.6

a. 212-micrometer particle
before cross sectioning
(70X)

b. 6-micrometer
diamond polished
cross section
(500X)

Figure 8. Photomicrographs of $^{238}\text{PuO}_2$ microsphere 1-24-4,
 $\rho = 10.8 \text{ g/cc}$

a. 214-micrometer particle
before cross sectioning
(70X)

b. 6-micrometer
diamond polished
cross section
(500X)

Figure 9. Photomicrographs of $^{238}\text{PuO}_2$ microsphere 1-24-2,
 $\rho = 10.7 \text{ g/cc}$

a. 209-micrometer particle
before cross sectioning
(70X)

b. 6-micrometer
diamond polished
cross section
(500X)

Figure 10. Photomicrographs of $^{238}\text{PuO}_2$ microsphere 1-24-10,
 $\rho = 11.1 \text{ g/cc}$

a. 221-micrometer particle before cross sectioning (120X)

b. 6-micrometer diamond polished cross section (500X)

Figure 11. Photomicrographs of $^{238}\text{PuO}_2$ microsphere 1-25-7, $\rho = 10.0 \text{ g/cc}$

b. 6-micrometer diamond polished
cross section (500X)

Figure 12. Photomicrographs of $^{238}\text{PuO}_2$ microsphere 1-25-14,
 $\rho = 10.0 \text{ g/cc}$

a. 221-micrometer particle
before cross sectioning
(70X)

b. 6-micrometer
diamond polished
cross section
(500X)

Figure 13. Photomicrographs of $^{238}\text{PuO}_2$ microsphere 1-24-5,
 $\rho = 9.1 \text{ g/cc}$

Test Conditions and Results

Shown on Figure 2 are curved lines which represent the release conditions during reentry at which the initial stagnation heat flux on a 250- μ m particle is equal to the values in the arc tunnel at test Conditions L, A, C, B, K, and M. These test conditions were selected to represent a range of reentry heating conditions that may occur in the use of isotopic power supplies in space. However, because of the varying nature of the heat pulse during reentry as contrasted to the constant heat flux in the test facility, caution must be exercised in comparing the arc tunnel condition to the reentry conditions. The parameter considered most important to simulate in these experiments is the stagnation heat flux. Other parameters (viz., enthalpy and body pressure) are constrained by the operational limits of the facility.

Since the heating during reentry varies with time and heating in the arc tunnel does not, it becomes important to consider the duration of the reentry heating and to compare it with the test times. Figure 14 shows the heat pulse trajectory for a 250- μ m particle released at 220,000 feet altitude and 24,000 fps. Shown also are the heat flux values corresponding to the test conditions. Noted on the reentry heat pulse is the calculated time for the initiation of melt during reentry. It can be seen that, for this case as in many others, particle melting occurs so rapidly that the simulation of this pulse with a constant value in the arc tunnel does not represent a serious problem. However, this difference does account for the fact that the heat flux lines for Conditions A and C lie generally outside the melt envelope on Figure 2 and yet melting occurs in the arc tunnel.

Also, although the equivalent test heat fluxes can be produced during reentry at various enthalpy-altitude combinations, this does not imply that these various combinations were reproduced in the experiments. From the enthalpy values given in Table VI, one can calculate an equivalent reentry velocity and for each test condition determine the altitude of reentry at which the heat fluxes are equal. Since the greatest portion of the K and M test condition heat flux lines lie well within the melt region, it seems satisfactory for these conditions to reproduce the heat flux to the particle with little regard given to the particular altitude-velocity combinations. However, as mentioned earlier, for releases near the melt zone border, these considerations become important.

Figure 14. Reentry heat flux for release of 250-micrometer particle at 220,000 feet altitude and 24,000 fps

Successful data were obtained for twenty-nine $^{238}\text{PuO}_2$ microsphere experiments and five $^{239}\text{PuO}_2$ experiments, along with numerous other exposures of nonradioactive materials (silver, iron oxide, alumina, and glass). The experiments using nonradioactive material provided data on the responses of catalytic and noncatalytic surfaces in the flow and were valuable in developing a technique for successfully holding the plutonium particles. However, because the data for plutonium dioxide microspheres are of most interest, no nonradioactive material tests are reported. Several tests were performed at each test condition, and the data from all successful experiments are given in Table VIII. In general, the repeatability of the particle response as indicated by the movie film and the debris samples was very good. The variation in the observed melt times is most likely explained by the inherent variation in the test conditions in an arc tunnel facility. However, taken as a group of data for each test condition, the repeatability appears satisfactory. In discussing the results of the experiments at each test condition, the discussion begins with the condition of minimum heat flux (Condition L) and proceeds through increasing severity to the maximum condition (Condition M).

TABLE VIII

Data on $^{238}\text{PuO}_2$ Experiments in Hyperthermal Arc Tunnel

Run No.	Particle Number	Diameter (μm)	Density (g/cc^{-1})	Test Condition	Stagnation Heat Flux ($\text{Btu/ft}^{-2}\text{-sec}^{-1}$)	Gas Enthalpy (Btu/lb^{-1})	Total Model Pressure (atms)	Observed Melt Time (sec)	Calculated Melt Time (sec)	Sampler Type	Debris Data from Sampler*
269	1-24-6	227	10.3	B	950	10,000	0.0737	0.138	0.128	A	No data available.
270	1-24-15	254	8.8	B	950	10,000	0.0737	0.144	0.135	None	No sample taken.
276	1-24-17	236	10.2	B	950	10,000	0.0737	0.146	0.130	A	No data available.
324	1-25-20	253	10.3	B	950	10,000	0.0737	0.140	0.135	Bc	975 particles by Tracerlab; range 0.31 to 0.91 μm ; peaks at 0.30 and 0.71 μm .
332	1-25-16	254	8.6	B	950	10,000	0.0737	0.136	0.135	B	No discrete particles found by LASL; background range 0.0014 to 0.005 $\mu\text{g/cm}^2$.
271	1-24-7	225	10.8	A	650	11,000	0.0485	0.193	>0.20	None	No sample taken.
285	1-24-19	221	10.4	A	650	11,000	0.0485	0.188	>0.20	None	No sample taken.
333	M-6	224	9.6	A	650	11,000	0.0485	0.198	>0.20	C	18 particles by LASL; range 0.34 to 1.91 μm ; average range 0.69 μm ; background 0.00012 to 0.009 $\mu\text{g/cm}^2$. (1)
335	M-3	218	9.4	A	650	11,000	0.0485	--	>0.20	Cc	316 particles by Tracerlab; range 0.2 to 0.9 μm ; peak 0.2 μm .
274	1-24-8	234	10.2	C	800	9,500	0.0593	0.185	0.175	None	No sample taken.
277	1-24-14	235	10.4	C	800	9,500	0.0593	0.190	0.175	Ac	No data available.
287	1-24-12	221	10.1	C	800	9,500	0.0593	0.200	0.16	None	No sample taken.
292	1-25-1	216	11.3	K	1800	13,000	0.0987	0.046	0.040	A	No data available.
293	1-25-5	230	10.2	K	1800	13,000	0.0987	0.040	0.042	A	No data available.
309	1-25-2	248	9.5	K	1800	13,000	0.0987	0.042	0.046	B	8 particles by LASL; range 0.5 to 1.0 μm ; background 0.003 to 0.023 $\mu\text{g/cm}^2$.
318	1-25-9	225	8.9	K	1800	13,000	0.0987	0.044	0.042	B	27 particles by LASL; range 0.3 to 3.4 μm ; average range 1.7 μm ; background 0.004 to 0.022 $\mu\text{g/cm}^2$.
321	1-25-3	234	10.2	K	1800	13,000	0.0987	0.041	0.043	Bc	227 particles by Tracerlab; range 0.56 to 3.4 μm ; no peak determined.
336	M-2	231	9.8	K	1800	13,000	0.0987	0.066	0.043	C	No discrete particles found by LASL; background range 0.00025 to 0.004 $\mu\text{g/cm}^2$. (1)

TABLE VIII
(continued)

Run No.	Particle Number	Diameter (μm)	Density (g/cc^{-1})	Test Condition	Stagnation Heat Flux ($\text{Btu/ft}^{-2}\text{-sec}^{-1}$)	Gas Enthalpy (Btu/lb^{-1})	Total Model Pressure (atms)	Observed Melt Time (sec)	Calculated Melt Time (sec)	Sampler Type	Debris Data from Sampler*
337	M-15	235	11.3	K	1800	13,000	0.0987	0.072	0.044	Cc	1954 particles by Tracerlab; range 0.2 to 1.0 μm ; peak 0.4 to 0.5 μm .
347	M-35	223	10.9	K	1800	13,000	0.0987	0.055	0.040	Cc	125 particles by Tracerlab; range 0.2 to 2.0 μm ; peak at 0.6 μm .
358	M-31	218	10.4	K	1800	13,000	0.0987	0.056	0.040	A	Data not available, Tracerlab.
354	M-29	235	9.8	M	2800	12,000	0.276	0.020	0.028	Cc	Preliminary size range 0.1 to >10 μm by TL.
355	M-37	227	10.6	M	2800	12,000	0.276	0.019	0.028	Cc	Preliminary size range 0.1 to >10 μm by TL.
356	M-33	220	10.3	M	2800	12,000	0.276	0.021	0.027	C	53 particles by LASL; range 0.59 to 14.7 μm ; peak 1 to 2 μm ; background --
369	M-10	225	9.0	L	100	3,600	0.0237	No melt			Particles from 0.17 to 1.8 μm by LASL; most between 0.2 and 1.0 μm .
371	M-22	219	11.2	L	100	3,600	0.0237	No melt			977 particles between 0.1 and 2.5 μm ; peak ~0.2 μm , Tracerlab.
373	M-16	221	10.6	L	100	3,600	0.0237	No melt			
376	M-32	227	7.4	L	100	3,600	0.0237	No melt			4691 particles between 0.1 and 2.5 μm ; peak 0.2 μm , Tracerlab.
378	M-30	222	9.1	L	100	3,600	0.0237	No melt			

*Analysis techniques by LASL and Tracerlab are sufficiently different to invalidate any comparison of the debris data for a given arc tunnel condition.

- (1) There is a high probability that impactors M-2 and M-6 were switched prior to debris analysis. The debris data shown for these two impactors are presented as reported by LASL. The arc tunnel conditions were listed prior to the sample switches.

Data were desired on how a microsphere responds when released well above the theoretical melt zone. To accomplish this, a test condition was determined which represents microsphere release at approximately 290,000 feet altitude and 26,000 fps. This condition (Condition L) produced a heat flux near 100 Btu/ft²-sec and according to calculations would not cause a significant rise in particle temperature. Five experiments are reported in Table VIII, and none of these particles melted. The particles were exposed for times as long as 5 seconds, and movies of the exposures revealed no detectable glow in the microspheres. For all exposures, debris samples were mounted downstream of the particles. In general, debris was collected; however, no repeatable pattern could be determined. In an effort to isolate the source of the collected debris, certain of these particles were mounted on the sting and exposed to the gas flow rate corresponding to Condition L but without the arc in operation. These "cold-gas" tests produced debris on the samplers, but subsequent exposures of that particular microsphere to the "hot" flow produced no debris. The conclusion from all data taken at Condition L tests was that microspheres released at this heat flux condition would not melt and the debris generated from this type of release is a function of the integrity of the microsphere surface (i.e., fines, cracks, voids, and clinkers located on the surface).

Experiments conducted at Condition A, which represents a release near the edge of the melt zone at velocities near 25,000 fps, revealed several interesting features. The exposures, which lasted for approximately 0.30 second, caused very gradual heating of the particle until at times typically like 0.20 second each particle began to gradually flatten. On the high speed movie film one can observe the response of the surface after melting began. In most cases ripples, oscillations, and other motions can be seen. In addition, small eruptions and protuberances were seen to randomly form during the heating. This phenomenon occurred for all test conditions with the exception of Condition L where, due to insufficient light on the particle, it likely could not be detected. Debris samplers mounted downstream of the microspheres collected debris. However, no debris which could be attributed to stripping or liquid layer breakup was seen to leave any particle. The melting was very gradual and no significant piling up of the melt was observed. Also, no large droplets were seen to form on the particles. Figure 15, which is a series of frames taken from the high speed

a. 0 second

b. 0.15 second

c. 0.188 second (melt)

d. 0.35 second

Figure 15. Ablation sequence of 221- μm diameter $^{238}\text{PuO}_2$,
 $\rho = 10.4 \text{ g/cc}$, Condition A, $H_s = 11,000 \text{ Btu/lb}$,
 $q_s = 650 \text{ Btu/ft}^2\text{-sec}$, Run 285

movies, shows a typical microsphere response at Condition A. This very slow response suggests that this heat flux is very near the thermal equilibrium value for the particle, and calculations support this observation. This fact and the transient decay of the reentry heat pulse explain why releases during reentry at this heat flux condition do not fall within the melt zone.

Three experiments using plutonium dioxide microspheres were conducted at test Condition C. The response of the particles was not significantly different from Condition A. From Table VI and Figure 14, one can see that the heat flux values for these two conditions are not drastically different, and as a result one would expect similar responses. Figure 16, also taken from the movie film, depicts the very gradual heating and subsequent flattening of the microsphere at Condition C. A debris sampler was used for only one of the particles exposed at this condition, and no data were obtained from that particular sampler. Therefore, no debris data are available for Condition C, but because of its similarity to Conditions A and B and because of the similarity of the response, as evidenced by the movie film, it is expected that the debris collected would also be similar.

Five plutonium dioxide experiments were successfully completed at test Condition B. The heat flux at this condition is somewhat more severe than either A or C but still represents a reentry release near the edge of the melt envelope. As expected, the thermal and melt responses of the particles were slightly more rapid than for Conditions A or C, with melting beginning at near 0.14 second. Ripples, waves, and oscillations were observed in the melt layer, but no large droplets were seen to form. The film suggests a thin melt layer on each particle. No debris which can be attributed to stripping of a liquid layer is seen to leave the microspheres even though the debris samplers collected a significant amount of micrometer sized debris. Figure 17 shows the typical thermal response of a microsphere at this condition. The last frame of this sequence shows a large central void in a microsphere which is not unexpected based on the results of the metallographic studies discussed previously.

a. 0 second

b. 0.130 second

c. 0.190 second (melt)

d. 0.22 second

Figure 16. Ablation sequence of 235- μm diameter $^{238}\text{PuO}_2$,
 $\rho = 10.4 \text{ g/cc}$, Condition C, $H = 9,500 \text{ Btu/lb}$,
 $q_s = 800 \text{ Btu/ft}^2\text{-sec}$, Run 277

a. 0 second

b. 0.144 second (melt)

c. 0.22 second

d. 0.24 second

Figure 17. Ablation sequence of 254- μm diameter $^{238}\text{PuO}_2$,
 $\rho = 8.8 \text{ g/cc}$, Condition B, $H_s = 10,000 \text{ Btu/lb}$,
 $q_s = 950 \text{ Btu/ft}^2\text{-sec}$, Run 270

Nine successful experiments using plutonium dioxide were conducted at test Condition K. As seen in Figure 2; this condition represents a more severe release condition and is well within the melt envelope. As expected, melting was initiated at near 0.04 second as contrasted to the 0.14 to 0.20 second for Conditions A, C, and B. The particles flattened; however, in most cases no large droplets were seen to form or leave. Figure 18 shows the typical thermal response of a microsphere at this condition. In one case, a partially ablated particle remained on the sting after the experiment and was reclaimed. This particle was weighed and found to have lost from 5 to 10 percent of its mass. The posttest specimen was examined and cross sectioned for metallographic study. Examination of the stagnation region of the flattened particle revealed several interesting features. Areas were visible which appeared to be either voids in the material or which were formed as "ripples" during the test and subsequently solidified. This type of formation possibly can be attributed to phenomena such as outgassing and flow forces acting on the surface. Figure 19 is a photomicrograph of the specimen cross section. The flattened stagnation area is clearly visible, as are voids which were likely present in the particle prior to the test. Debris samplers again showed micrometer and submicrometer size debris; there were no impact areas to indicate breakup of larger droplets. The data definitely support the thesis that microspheres released well within the melt envelope will undergo extensive melting.

To represent the release of microspheres in a relatively low altitude abort environment, three experiments were performed at test Condition M. The melting at this condition was very rapid, and large droplets were seen to form. These droplets in some cases were stripped from the microsphere and in other cases ran back onto the sting where they either resolidified or were stripped from the sting as smaller debris. The debris samplers mounted downstream of the particle collected debris which proved to be of larger size than for any of the other conditions. Experiments at this condition provide support for calculations of extensive melting of microspheres released at velocities in excess of 15,000 fps at altitudes corresponding to the heat flux at this test condition. However, for releases below near 15,000 fps, calculations show that the rapid particle deceleration will prevent total melting (see Figure 2). Figure 20 presents a sequence of the movie film which depicts the melting at this heat flux condition.

a. 0.041 second

b. 0.046 second (melt)

c. 0.063 second

d. 0.08 second

Figure 18. Ablation sequence of 216- μm diameter $^{238}\text{PuO}_2$,
 $\rho = 11.3 \text{ g/cc}$, Condition K, $H_s = 13,000 \text{ Btu/lb}$,
 $q_s = 1800 \text{ Btu/ft}^2\text{-sec}$, Run 292

Figure 19. Photomicrograph of microsphere M-35 (1000X)

a. 0.015 second

b. 0.020 second (melt)

c. 0.032 second

d. 0.037 second

Figure 20. Ablation sequence of 235- μ m diameter $^{238}\text{PuO}_2$,
 $\rho = 9.8 \text{ g/cc}$, Condition M, $H_s = 12,000 \text{ Btu/lb}$,
 $q_s = 2800 \text{ Btu/ft}^2\text{-sec}$, Run 354

Ablation Debris

As previously stated, debris samplers were mounted downstream of the microspheres during ablation. These samplers, which are basically thin copper plates of various sizes and shapes, operate on an impaction principal and therefore are associated with some unknown collection efficiency. This question of efficiency is discussed later in this report. After each ablation experiment, the test chamber was cleaned with a special solution for cleaning radioactive material from surfaces. This cleaning prevented cross contamination of the clean samplers which were mounted on the holder immediately prior to each exposure.

After each successful exposure, the impact sampler was removed from the test chamber and placed in a container designed especially for a sampler of that particular size and shape. A 0.25-mil thick piece of mylar mounted on a ring was glued into place over the sampler and provided a seal to prevent the removal of any debris from the sampler. A lucite cover was placed over the mylar covering for ease of handling, and the sampler was removed to the Aerospace Nuclear Safety Department Radiochemistry Laboratory for initial examination.

An autoradiograph of each debris sampler was made as the initial step in the debris analyses. The autoradiographs, which were made by placing nuclear emulsion film over the thin mylar covering on the sampler, provided qualitative information as to the presence and general pattern of the debris. One can distinguish between areas of uniform deposition of smaller, numerous debris and areas of less-numerous, discrete larger pieces of debris. The samplers were then gamma counted in an effort to determine the quantity of material present. Because of the effects of size and shape of the material and because of the lack of any standards of the proper shape to calibrate the instrument, these data are not conclusive.

The impact samplers were analyzed for size distribution and total quantity of material present by Tracerlab, Inc., under Sandia contract. Los Alamos Scientific Laboratory also provided assistance in analyzing the debris on certain impactors and established the feasibility of analyzing the debris. Only a general outline of analytical technique used by Tracerlab is given here. Prior to each experiment, the samplers were coated with a vinyl material. This material provided a soft

surface to assist in entrapping the debris and also allowed Tracerlab to remove the ablation debris from the sampler by dissolving the vinyl. The solution containing the collected particulate was centrifuged, and a statistical portion of the centrifuged residue was placed on microscope slides. Nuclear emulsion film was exposed to the radioactive particles on the slides. These films were then used to obtain the size distribution of the particulate by the hollow star technique. Because of the limited exposure times of the nuclear emulsion film, the lower limit of resolution for these analyses by the hollow star method was 0.05 micrometer. Therefore, the size distribution is truncated at this diameter as a lower limit, with the upper limit being the largest size present on the slide. In order to account on a mass basis for any debris smaller than 0.05 micrometer, a radiochemical analysis of the entire inventory was performed for selected samplers. Included in this analysis was the supernate solution, the remaining centrifuged residue, and the particles previously counted. By subtracting the mass equivalent to all material found larger than 0.05 micrometer, the mass present in particulate smaller than this was determined.

Included in Table VIII are data on the debris collected on each sampler. When grouped by test conditions and considered in light of the severity of each test condition, definite trends are suggested. Of the test conditions which cause melting, Condition A is the least severe. Debris collected and analyzed by Tracerlab for this condition is smaller in quantity of particles, size of debris, and total mass than for the other conditions. Figure 21 presents the radiograph of a sampler for Condition A together with the debris data. As stated previously, no samplers were analyzed for Condition C. Examination of the debris data determined by Tracerlab for Condition B shows more particles than at Condition A, with the debris size smaller than K or M but equivalent to that at Condition A. It is significant to recall the very slow rates of melt for these two test conditions. It should also be remembered that test times were 0.3 second for these conditions, whereas they were only 0.1 second for Conditions K and M. A typical autoradiograph together with the debris data for Condition B is shown in Figure 22. The debris reported for test Condition K reveals larger size particulate than for either Conditions A or B. As will be discussed later, the total quantity of material present is also greater than for

Condition A. Figure 23 presents a typical autoradiograph for Condition K together with the data on the debris analyzed by Tracerlab. Finally, the debris collected at Condition M, the most severe of the test conditions, was larger than any other collected. The quantity of debris collected at this condition was larger than that for either A or B and was roughly equivalent to that collected at Condition K. Because of the large quantity of melt at this severe heating, molten material ran back onto the sting and it is thought that this affected the debris data at this condition. However, the larger size debris is consistent with the trend established by the remaining test conditions. Figure 24 presents a typical autoradiograph for Condition M together with the debris data determined by LASL.

36 hours

Arc tunnel condition - A

Number of particles sized - 316

Diameter range - 0.2 to 0.9 μm

Peak - 0.2 μm

Lab - TL

Sampler type - Cc

Figure 21. Autoradiograph of Impactor M-3

47 hours

Arc tunnel condition - B

Number of particles sized - 975

Diameter range - 0.91 to 0.31 μm

Peaks - 0.3 μm , 0.71 μm

Lab - TL

Sampler type - Bc

Figure 22. Autoradiograph of Impactor 1-25-20

3-3/4 hours

Arc tunnel condition - K

Number of particles sized - 125

Diameter range - 0.4 to 1.2 μm

Peak - 0.6 μm

Lab - TL

Figure 23. Autoradiograph of Impactor M-35

3 hours

Arc tunnel condition - M

Number of particles sized - 53

Diameter range - 0.59 to 14.7 μm

Peak - 1 to 2 μm

Lab - LASL

Sampler type - C

Figure 24. Autoradiograph of Impactor M-33

The tests and debris collected at Condition L warrant comment primarily because the particle did not melt in 5 seconds of test and yet debris was collected on the samplers. The initial experiment at Condition L was done with particle M-10 and Figure 25 presents the autoradiograph of this sampler. The sampler was analyzed by LASL and a complete particle distribution was not attempted. However, LASL did measure debris from 0.17 to 1.8 micrometers in diameter. Since melting did not occur, it was suspected that the debris collected represented loose material or fines on the surface. Additional tests were undertaken to confirm this by passing cold gas over the microsphere prior to the hot gas test and by using a clean sampling surface for each of the cold and hot gas runs. Table IX summarizes the results of these experiments and appears to confirm that the debris generated is a function of the quality of the microsphere surface.

Figure 25. Autoradiograph of $^{238}\text{PuO}_2$ particle M-10 debris from Condition L test

TABLE IX
Condition L Test Results

Particle Number	Debris Collected		Remarks
	Cold Gas	Condition L	
M-10	--	Yes	No cold gas test
M-22	No	Yes	Amount collected slightly less than M-10
M-16	No	No	Microsphere fell off early in test
M-32	Yes	No	Amount collected slightly less than M-10
M-30	No	No	Valid test

The sampler from the hot gas run for particle M-22 and that from the cold gas run for particle M-32 were sent to Tracerlab for analysis. The results of these analyses are given in Table X. It can be seen that the total debris on sampler M-32, a cold gas run, is greater than that for M-22, a normal Condition L run. This, in addition to the randomness of debris collected, as shown in Table IX, appears to confirm that the debris generated is a direct function of the original state of the microsphere surfaces.

Because of the technique used, the size range of the debris counted by Tracerlab using the hollow star method extended from 0.05 μm to the largest particle found on each of the slides. However, since the mass of $^{238}\text{PuO}_2$ represented by this size range accounted for only a small percent of the total mass collected on the samplers, a radiochemical analysis of the entire inventory on selected samplers was performed. Samplers selected were M-3, representing test Condition A; M-35, representing Condition K; and M-29, representing Condition M. This analysis, included the supernate solution, the remaining centrifuged residue, and particles previously counted. Since the particles previously sized and the centrifuged residue accounted for all the debris larger than the lower limit of resolution, the remaining mass was contained in particles less than the smallest size previously counted. Table XI shows the mass and percent of total mass for the size ranges counted by Tracerlab.

TABLE X

Percentages of Total Equivalent Mass
for Various Ranges of Equivalent $^{238}\text{PuO}_2$
Particles Collected on Impactors

Size Range of Particle Diameters (μm)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	Percent	Cumulative Percent
<u>Four Size Ranges - Impactor M-32</u>			
0 - 0.1	3.97×10^{-3}	30.46	30.46
0.1 - 0.6	9.39×10^{-4}	7.21	37.67
0.6 - 1.0	5.88×10^{-4}	4.52	42.19
1.0 - 10.2	7.52×10^{-3}	57.80	99.99
Total	1.30×10^{-2}		
<u>Three Size Ranges - Impactor M-22</u>			
0 - 0.1	1.1×10^{-3}	70.56	70.56
0.1 - 0.6	1.8×10^{-4}	10.84	81.40
0.6 - 2.5	3.0×10^{-4}	18.60	100.00
Total	1.62×10^{-3}		

TABLE XI

Percentages of Total Mass
of $^{238}\text{PuO}_2$ Collected on Samplers

Size Range of Particle Diameters (μm)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	Percent of Collected Mass	Cumulative Percent
<u>Condition A - Sampler M-3</u>			
0 - 0.3	0.071	99.86	99.86
0.3 - 0.9	0.098×10^{-3}	0.14	100.00
<u>Condition K - Sampler M-35</u>			
0 - 0.05	0.3390	95.79	95.79
0.05 - 0.10	0.0122	3.45	99.24
0.10 - 1.6	0.0027	0.76	100.00
<u>Condition M - Sampler M-29</u>			
0 - 0.2	1.649	95.1	95.1
0.2 - 0.6	0.0012	0.07	95.17
0.6 - 1.0	0.0022	0.12	99.53
1.0 - 5.0	0.0231	1.3	96.60
5.0 - 14.0	0.0586	3.4	100.00

Sandia attempted a mass balance of the material ablated from the $^{238}\text{PuO}_2$ microspheres. Where possible, a microsphere was weighed before and after testing, but in most cases the percent of the mass ablated was determined by estimating the volume of the microsphere before, during, and after testing from the movie films of the tests. This analysis revealed that less than 100 percent of the mass ablated from the microsphere was actually collected on the sampler. A collection efficiency analysis of the samplers was performed 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 particulate between 0.01 and 10 micrometers in diameter would be collected with approximately 100-percent efficiency. Particulate less than 0.01 micrometer in diameter would be entrained in the flow and diverted around the sampler. Unless it was originally projected toward the sampler, particulate greater than 10 micrometers in diameter would not be affected by the flow and also would miss the sampler. The results of the collection efficiency analysis indicated that the size distribution of the debris collected was not exactly representative of the ablated size distribution in all cases. Therefore, it was necessary to modify the size distributions given in Table XI so as to reflect the total ablated mass and the debris not collected during the test.

For the Condition A test, the largest particle size found on sampler M-3 was 0.9 micrometer in diameter. Since the collection efficiency of the sampler was approximately 100 percent up to 10 micrometers, it is assumed that the entire ablated inventory was less than 0.9 micrometer in diameter. The 100 percent collected mass from 0.01 to 0.9 micrometer represents 52.20 percent of the total ablated mass. The uncollected remainder must then be less than 0.01 micrometer in diameter and represents 47.73 percent of the ablated mass. This size distribution does not differ appreciably from that given in Table XI.

The analysis of sampler M-35 (Condition K) resulted in conclusions similar to those reached for the Condition A test. Since the largest particle collected was 1.6 micrometers in diameter, the entire ablated inventory was assumed to be less than 1.6 micrometers. The 100 percent collected mass from 0.01 to 1.6 micrometers represented 9.29 percent of the ablated mass. The remaining 90.71 percent was contained in particles

less than 0.01 micrometer in diameter. This size distribution is also not much different from the values given in Table XI.

The analysis of sampler M-29 (Condition M) resulted in a significant change from the size distribution shown in Table XI. A review of the movie films showed that all the Condition M exposures resulted in rapid melt and runback of the molten $^{238}\text{PuO}_2$ onto the sting. Several droplets from 25 to 100 micrometers in diameter were stripped from the molten runback during the test; however, none of these droplets was collected on the impact sampler. Most of the runback material remained on the sting after the test and accounted for the great majority of the "ablated" mass. The collected mass between 0.01 and 14 micrometers in diameter represented 6.94 percent of the total ablated mass. It was estimated that the runback material accounted for almost all of the remaining 93.06 percent. Therefore, the percent of ablated debris less than 0.01 micrometer in diameter was very small.

Table XII shows the final size distribution of the ablated debris, based on the Tracerlab analysis and modified by the theoretical sampler efficiency analysis. Although the data presented here are based on one test for each reentry condition, it is believed 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. It is apparent that the debris size distribution is dependent on the heat input and the melt rate of each microsphere.

Discussion of Arc Tunnel Results

In discussing the results of the arc tunnel experiments, consideration will be given first to the thermal and general melting responses of the microspheres and then to the data on debris generated and collected. The experimental data on thermal response consist of reducing the timing on the high speed film to determine the time required to initiate stagnation point melting. The timing marks placed on the film are used, and this results in timing accurate to within one millisecond.

TABLE XII
Percentages of Total Mass
of $^{238}\text{PuO}_2$ Ablated from Microspheres

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

Table VIII gives the time required in each experiment for initiation of microsphere melting at the stagnation point. The repeatability of the times is reasonably good, particularly when one considers that the repeatability of the arc tunnel flow conditions is not precise. To compare the measured thermal response of the microspheres with the calculated values, a specialized version of the SPHERE 2 program⁶ was used. This version, called ARCJET, computes the temperature response of a microsphere for the constant heat flux conditions of the arc tunnel.

Since the diameters of the microspheres varied from test to test, a parametric study in heat flux and particle diameter was performed using this version of the program. The results of this study are given in Figure 26 for microsphere diameters of 190, 210, and 230 micrometers.

Shown on the figure are the heat flux values for the conditions of the experiments. Data interpolated from these curves for the various size particles tested at each condition are shown in Table VIII.

Figure 26. Time for melt initiation versus heat flux

The calculated time required to initiate melting is compared with the observed time for each experiment at a particular test condition, and these ratios, with tolerances, can be grouped for each condition. Table XIII presents these data and shows the relatively good agreement between the calculated and observed times.

TABLE XIII
Comparison of Calculated
and Observed Melt Times

Condition	Ratio of Observed Melt Time to Calculated Melt Time
C	1.12 ± 0.09
B	1.06 ± 0.05
K	1.16 ± 0.2
M	0.72 ± 0.1

Several interesting features can be observed from these data. Although agreement between observed and calculated melt times for Condition M is not as good as for some of the other conditions, the observed melt time data for Condition M represent the most repeatable data. For the other conditions, some very noticeable variation in observed melt times was found. At these lower heat flux values, the variation in test conditions becomes critical, particularly at the lowest heat flux conditions, since the heat flux is approaching a value which will not cause melting but will only result in a state of equilibrium temperature due to the radiation heat loss. The repeatability of the arc tunnel (generally regarded to be about ± 10 percent of the nominal value) can be critical at conditions such as A and C near the equilibrium point, but it is not nearly as noticeable at the high heat flux conditions. Comparison of the possible variation at these conditions and the observed slow melt rate with Figure 26 offers further support that the calculated heat flux values are experimentally correct. For equivalent heat flux conditions, calculations indicate that the time required for particles to begin melting is some direct function of the particle diameter. Because of the size limit on the particles that could be tested, and the variation of the test conditions from run to run, this function cannot be demonstrated directly from these data.

The general melting characteristics of the microspheres also bear out the relative positions of the calculated heat flux values.

Conditions B, C, and A (see Table VI) are not extremely different in heat flux. Consequently, the time and event differences from the films are not extreme. However, even with the limited number of tests conducted, there is some variation in the ablation phenomena for different conditions.

No particle mass loss is apparent from the films of the Condition A tests, which is the least extreme of all the melt conditions tested. In fact, during the majority of the tests at this condition, the $^{238}\text{PuO}_2$ microspheres did not change shape appreciably or develop a flat front face. Small "bubbles" or protuberances did form on the surface of the particles (usually on or near the front face). It is important to note that even with this relatively mild exposure of the microsphere and no apparent mass loss, radioactive debris was collected on the sampler in every test.

The microspheres exposed to Condition C exhibited essentially the same response as those exposed to Condition A except that there was some distortion of the particle and the surface was somewhat more active. As in the Condition A exposures, the film record indicated that the temperature gradient was small. One particle showed a large void in the center front face during the latter stages of exposure.

Particles exposed to Condition B, which is slightly more extreme than Conditions A and C, exhibited progressively more distortion and surface activity. Also, the time was less to first glow, beginning of melt, and a one-third flat face. The flat face appeared to undulate or "ripple" with a definite movement. The temperature gradient appeared to be somewhat larger than at Conditions A and C. Again, no mass loss (except a change in the size of the particle) or ablation was obvious, but radioactive debris was collected on the sampler. Two of the particles showed voids in the center of the front face.

Condition K is a much more extreme reentry simulation than Conditions B, C, or A. The majority of the microspheres exposed to this condition showed extreme activity on the films. They began melt much sooner and formed flat front faces until they were hemispherical in shape. The front surface was very active, and in addition to "bubbles" on the surface, several small eruptions were obvious. These eruptions appeared to yield debris; however, no individual particles could be distinguished. One frame of one film showed a streak of debris which could be traced back to a surface eruption. On several of the microspheres the melted surface ran back along the sides of the particle. This runback was not observed to shear or fall away from the main microsphere during a test. More substantial temperature gradients were from the front face to the back of each microsphere than for the previous conditions.

Condition M is the most extreme condition tested, and the exposure events reflected this extreme. First glow and melt occurred soon after exposure. During the early stages of exposure there was a definite line between the heated front and the unheated back of each microsphere. This indicated a very large temperature gradient. This gradient later disappeared almost completely, and the entire particle was white hot. The flat face configuration occurred early and progressed until the

particle was two-thirds flat and almost disc shaped. Some melt runback formed into individual droplets approximately 50 micrometers in diameter. These droplets were subsequently sheared away from the main microspheres by the stream or were forced back and remained on the sting. It can be said that the $^{238}\text{PuO}_2$ microspheres exposed to the M condition melted completely.

The debris data presented must be interpreted with consideration given to the type of facility used in the experiments and to how the parameters which may cause mass loss in reentry are reproduced in the facility. The most important factor not reproduced in the arc tunnel experiments was the body force resulting from the deceleration of a particle during reentry. It must be recognized that the addition of this parameter may significantly alter the results of the debris generation. It appears that the consensus of the literature and the analyses is that the body force tends to increase the instability of a liquid in the stagnation region of a body. This increase may be caused by the waves generated on the liquid surface "cresting" or "piling up" more rapidly, and thereby becoming unstable. If this increase in instability with body force does in fact occur, then the mass loss from the experiments may be reasonable. The high deceleration levels which accompany the reentry of small particles likely causes molten material to form flanges on the particles; the effect which these flanges have on the mass loss of the particles is not known. The phenomenon is not reproduced in the experiments discussed in this report.

It was pointed out previously that a Weber number of about one was the lowest value reported experimentally. The arc tunnel results do not prove that this is the proper value to use even though debris was collected at a Weber number near 1.0. Indeed, some other phenomenon may be causing the mass loss. Moreover, since debris was collected at Condition L, a no-melt condition, one can assume that a portion of the debris generated at all conditions was the result of a similar type response. The equivalent $^{238}\text{PuO}_2$ mass determined to be on sampler M-22 represented 3×10^{-3} percent of the original microsphere mass, and that on sampler M-32 represented 3×10^{-2} percent. Since sampler M-32 was from a cold gas run and M-22 was from a no-melt Condition L run, it is apparent that hydrodynamic instabilities are not responsible for this debris. The conclusion is that debris, which is a function of the

surface condition of the microsphere, will be generated when these particles are released in reentry. Since it is believed that the sampler efficiency is less than 100 percent, the total mass of debris generated cannot be determined. Based on this limited data, it appears that as much as 3×10^{-2} percent debris can be generated for microspheres released at conditions which do not cause melting.

The debris data resulting from tests at Conditions A, B, and C are grouped together and are considered to represent the response of microspheres released at heat flux values near the edge of the melt zone. From Table XI it can be seen that the overwhelming proportion (99.86 percent) of the debris was determined by Tracerlab to be less than 0.3 micrometer in diameter and that only 0.14 percent was larger than 0.3 micrometer. In addition, the largest particle found was 0.9 micrometer; this is a smaller maximum than that found for any of the other conditions, including Condition L. The total debris collected represented 0.41 percent of the original microsphere mass, but because of sampler inefficiencies it is expected that more debris was ablated than was collected on the samplers. Because of the very large percentage of debris which was smaller than the minimum sized, vaporization mass loss rates were estimated to determine if an amount of material near that collected could be vaporized. The calculational relationships are shown in Appendix A. For the exposures typical of Conditions A, B, and C, the results show that more mass than needed to account for the mass on the samplers could be vaporized. Consequently, vaporization must be considered as one source of much of the debris found on the samplers at these conditions.

The debris data resulting from exposures at Condition K show that more mass was collected on the samplers than for Condition A although the test duration was only one-third as long. The maximum size debris found was larger than for Conditions A and B; however, as before the greater percentage of the debris was determined to be less than 0.05 micrometer in size. Approximate calculations indicate it may be possible to vaporize the quantity of material present on the sampler, but more definite calculations cannot be performed because of lack of knowledge of the surface temperature.

The total mass present on sampler M-35 (Condition K) represented 0.56 percent of the original microsphere mass. This particle was reclaimed after the test and weighed. It had lost from 5 to 10 percent of its original weight. Figure 19 is a photomicrograph of the particle cross section. No large droplets were seen to leave the microsphere during the test. One can then assume that the entire 5 to 10 percent mass loss can be represented by the type of debris collected by the sampler. Since the collected debris was 0.5 percent of the microsphere mass, the sampler appears to be collecting only about 10 percent of the ablated mass. If this is true, vaporization does not account for the total debris ablated, and other mechanisms must be participating in the ablation.

As reported earlier in this report, the melting at Condition M was very rapid, and large molten droplets were sheared from the microsphere onto the sting where they either resolidified or were shattered into smaller debris. The debris data from the samplers confirm that larger size debris was present and also that a larger total mass than at other conditions was ablated in less than 100 milliseconds. A mass representing about 2.6 percent of the original microsphere mass was collected, and approximately 95.1 percent of this was smaller than 0.2 micrometer. There appears to be no doubt regarding substantial melt at this condition. However, size distribution data are less reliable since the effects of the sting were very apparent in the movie film.

In summary, the following conclusions result from the arc tunnel experiments.

1. Based on the agreement between the observed and the calculated melt times shown in Table VIII, it is concluded that the thermal response of the particles can be satisfactorily predicted using the Sandia heat transfer program. If the particles are stable during reentry, they will melt in a liquid layer fashion.
2. The formation of protuberances on the particles should be expected during reentry. This formation appears to be a random and unknown function of the particle density and possibly the test condition. However, as a mechanism of mass loss, this phenomenon appears to be secondary to the melting ablation, particularly in the region of the melt envelope.

3. Based on the limited number of experiments performed at a "no-melt condition," debris should be expected when microspheres are released at significant velocities even though melting does not occur. The formation and quantity of debris appears to be random with the condition of the surface.
4. Vaporization may be responsible for a substantial portion of the debris formed at conditions of release near the edge of the melt zone. However, the percentage mass loss for a particular particle exposed to Condition K indicates that other mechanisms of mass loss are in effect.
5. The debris collected on the arc tunnel samplers represents the size of some, but not necessarily all, of the debris generated by the ablation. Unknowns in sampler efficiency versus particle size preclude stating that the size distribution determined from the samplers is an exact reproduction of the size distribution in the arc tunnel or during reentry.
6. The oscillating motions of the surface melt layer and the general ablation appearance suggest that the forces on the melt layer could well be causing instabilities and as a result generating the ablation debris.
7. Since particles as small as a few micrometers in diameter would leave evidence on the film of their being ablated, it is concluded that the size of the debris on the samplers, which in general is smaller than a few micrometers, is typical of the ablated debris and does not represent larger particles being further broken up by the sampler.
8. Based on successful experiments in which the partially ablated particle remained on the needle after the test, it is concluded that the debris on the samplers was generated during the ablation and not by the partially ablated particle striking the needle.

Study of Breakup of Partially Molten Spheres in a Hypersonic Shock Tunnel

As a part of the overall theoretical and experimental program presented earlier, a program was initiated to study the breakup of particles whose melt percentage varied from 0 to 100 percent of the particle. The experiment program was implemented at Cornell Aeronautical Laboratory (CAL) with Dr. John L. Beal as Principal Investigator. The

purpose of this program was to determine a critical Weber number for the breakup of particles as a function of the liquid layer thickness on the particle. To accomplish this, a program was conducted in the 8-foot leg of the CAL Hypersonic Shock Tunnel. Because of the relatively short run times in this facility (~5 milliseconds), it was not possible to heat the test model to an ablating condition. Therefore, particles were preheated to a preselected partially molten state by passing them through a high temperature furnace. Upon leaving the furnace, the particles entered directly into the hypervelocity flow. Details and results of the program are given in the following sections which are taken predominately from CAL progress reports and the final report on the study.

Test Equipment and Instrumentation

The test program was conducted in the 8-foot leg of the CAL Hypersonic Shock Tunnel. A contoured nozzle with a 4-foot-diameter exit was used to expand the nitrogen test gas to the desired test section conditions. The test section is equipped with two pairs of 16-inch-diameter observation windows mounted a short distance aft of the nozzle exit.

A 33-inch-long carbon tube furnace was employed to radiatively heat the FeO spheres. The furnace was mounted in the test section of the tunnel and was protected from the tunnel flow by a shroud arrangement. This shroud had vertical side plates in a double-wedged array and a flat floor plate, with the latter being located 2 inches above the horizontal centerplane of the tunnel. The center of the holes in the floor plate directly beneath the furnace tube was 20.5 inches downstream from the leading edge of the plate.

It was determined that spheres could be most reliably released from a holder in which each of the approximately 200 particles required for a test was contained in an individual compartment. The device retained 225 spheres in a 0.25-inch-square array of 0.014-inch-diameter holes. When the array was pulled past the oblique end of the bottom cover slide, the spheres fell at fairly regular time intervals, as measured by high-speed motion pictures. For the tests, a motor was arranged to drop the 225 specimens over a period of 1 second.

Free-stream pitot pressures were measured with three pitot probes with the planes of the probe tips located 47.125 inches downstream from the leading edge of the furnace floor plate. Furnace temperature was determined from the output of a Radiamatic optical pyrometer mounted on the furnace in such a manner as to view a local area of the outer surface of the heated carbon tube.

Four motion picture and two still cameras were used in the initial runs; two of the motion picture cameras were considered "primary" and were relatively unchanged in position or settings during the tests, with the exception that color film was substituted for black and white in one camera after the first run. Two of the motion picture cameras provided useful images. They were located with their optical axes nearly perpendicular to the tunnel axis, but on opposite sides of the tunnel. Camera I was a Photosonics 16-mm running at 1000 frames per second, with its lens at f2 and loaded with Tri-X (black and white) film. Its field size was approximately 15 inches by 10 inches. Camera II was identical, but was loaded with Ektrachrome EF film and was equipped with a lens yielding a field size near 9 inches by 6.5 inches. Film development was "pushed" by using a first development time of 20 minutes.

Test Material

Since the intent of this study was to provide information which could be used in estimating the breakup of plutonium dioxide microspheres, an analog test material was selected for plutonium dioxide which could not be tested in the facility because of its radioactive nature. Since the phenomena to be studied were those of mass loss and breakup, parameters which affected this response were used to determine the simulant. Primary emphasis was given to liquid surface tension and viscosity, with thermal diffusivity of secondary importance. Initially, aluminum oxide was selected, but because of its high melting temperature and poor radiant heating characteristics, it was concluded that this material could not be preheated to a partially molten state in the furnace. An additional search led to the selection of ferrous oxide (FeO) as the analog material.

The surface tension and viscosity of the fuel material at its melting point are:

viscosity in centipoise	$32 \pm 25\%$
surface tension in dyne/cm	$525 \pm 15\%$

The viscosity of ferrous oxide is as follows:¹⁹

<u>Temperature (K)</u>	<u>Viscosity in Centipoise</u>
1650 (m.p.)	50.4
1700	39.5
1750	31.5
1800	25.3

The surface tension at 1843 K is listed as 585 dyne/cm (0.040 lb/ft), and values of Weber number for molten ferrous oxide were computed using this surface tension value.

No numerical use of the values of viscosity was required in the program other than noting, as mentioned earlier, that they are near those for the fuel material.

Preparation of suitable spheres presented some problems, and several different methods were attempted. The initial approach called for a dense mass of ferrous oxide (wustite) which could be broken up, sized, and melted in a plasma gun to produce spheres. An effort was made to produce the wustite by melting Fe and Fe_2O_3 together under an inert gas purge. However, these attempts were unsuccessful because the mixture dissolved the crucible material, and crucibles which would not dissolve could not be obtained in a satisfactory time period.

Efforts then turned to the direct oxidation of iron spheres. Iron spheres were purchased and exposed to a $\text{CO}_2\text{-H}_2$ mixture at about 1500°F. Although ferrous oxide spheres were produced, they were unsatisfactory in that they showed excessive porosity which was traceable in part to the porosity of the commercial iron spheres. Finally, low porosity iron spheres were produced by a wire melting process, but these

fully dense iron spheres also developed some porosity during oxidation. Figure 27 shows typical spheres before and after oxidation. Composition was determined solely by weight gain during the preparation experiments and was later checked by wet chemical analysis. Data from both of these procedures agree that a wustite composition was obtained. Sphere sizing was by wire sieves, and the spheres were inspected and culled under a microscope prior to loading into the dropping mechanism.

To calibrate the degree of sphere melting in the preheat furnace, spheres were subjected to metallographic analyses after being dropped through the furnace. It appeared reasonable that a partially molten condition would show a difference in grain size or some other detail which would differentiate the molten and nonmolten portions, but such was not the case. Calibration tests indicated that complete melting occurred when the furnace temperature was 4380°R or higher. This was determined by examining the sectioned spheres and observing their smoothing and rounding, including that of their porosity.

Test Procedure and Conditions

The main objective of each test run was to allow a series of some 200 FeO spheres to fall freely through the furnace tube, thus incurring some degree of fusion, and to continue falling into the tunnel test section where a midsample or test group of the set of 200 would be subjected to a short-duration, steady-state hypervelocity gas flow. The resulting dynamic behavior of the test group of spheres in the "drop zone" of the gas stream was photographically recorded. Those spheres falling through the test section (essentially in vacuo) prior to the arrival of the tunnel flow were retrieved in a tray of silicone oil positioned on the test section floor beneath the furnace tube. The recovered spheres were subsequently sectioned in an unsuccessful effort to determine the actual molten radius fraction of the test droplets.

a. 450- μ m iron sphere made by melting wire; not oxidized (150X) (midsection, etched in 5% nitric acid + 95% methanol)

b. Typical FeO sphere yielded by dense iron spheres (150X) (midsection)

c. FeO specimen as oxidized; etched to reveal grain boundaries (150X) (midsection)

Figure 27.

In general, the test section conditions were determined from measured shock tube conditions and test section pitot pressures; it was further assumed that the test gas processing imposed by the shock tunnel nozzle was an equilibrium real nitrogen isentropic expansion. Table XIV presents the test conditions for each of the successful runs.

A heated sphere falling in a vacuum leaves the furnace at a velocity which requires about 3 milliseconds for 1 inch of fall. In the first inch of fall, it will pass through the region where a boundary layer will form on the underneath side of the furnace plate and will then be in the region where good flow occurs. In this first 3 milliseconds, a sphere cools by radiation. If all of the radiation cooling results in solidification, a solid crust approximately 0.5-micrometer thick will form over the surface of the molten material. When the stream envelops the sphere, aerodynamic heating occurs and this tends to remelt the crust. At low Weber number conditions, the aerodynamic heating is estimated to remelt the crust on the frontal hemisphere in slightly less than 0.5 millisecond.

One of the reasons why the test spheres were released at a rapid rate, more than 200 per second, was to have them falling with only short distances between them. This would tend to ensure the test of a sphere which was still very close to the furnace so that any crust would be quickly remelted. In practice, the spheres were not always uniformly spaced. In the test results, no correlation was observed between tendency toward breakup and closeness to the furnace, which suggests that any such crust was without influence under the test conditions.

Hypersonic Shock Tunnel Test Conditions*

Run Number	1	4	6	7	8	9	11	12	13	15
Reservoir Conditions										
P_O , psia	2,971	2,894	6,304	7,392	6,602	6,253	6,603	2,802	2,843	1,747
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	7,076	9,067	10,352	9,648	10,514	10,500	10,534	9,131	9,185	8,443
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	0.01242	0.01859	0.03693	0.06111	0.03534	0.03626	0.03688	0.01702	0.01779	0.01411
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	0.1066	0.1927	0.3026	0.1888	0.1887	0.1928	0.1135	0.1154	0.07737
Sphere Conditions**										
Re_∞ , D	149	101	183	288	180	179	183	108	109	73
We	4.07	5.24	10.4	15.3	9.82	10.0	10.25	4.83	5.00	3.65
Kn_∞ ***	0.118	0.162	0.088	0.0536	0.093	0.090	0.088	0.152	0.149	0.213

* Based on the properties of real nitrogen.

** Based on a sphere diameter of 283 μ m. The variation in diameter was ± 5 percent.

*** Computed by $Kn_\infty = 1.486 M_\infty / Re_{\infty D}$.

Test Results

Motion pictures provided the primary means of observing sphere behavior and of relating instantaneous stream conditions to sphere behavior. Preliminary trials with melting iron wires indicated that radiation from the iron oxide spheres would permit framing rates up to about 1000 per second. More information on stream flow time would have been obtained at higher framing rates, but at 1000 frames per second the cameras employed in these tests had a "blind" time of only 0.8 millisecond between frames and events could be immediately related to stream flow starting time to within some fraction of 1 millisecond. An even closer relation was established through knowledge of sphere velocity and acceleration yielded by the photographs.

The manner in which sphere behavior was related to stream flow timing in each run may be learned by studying Figures 28 and 29 which apply to Test 6. The first flow-related occurrence visible in Figure 28 is the so-called preglow seen in Frame (a). This preglow is of unknown origin, and thus is not helpful in establishing timing. In the upper left portion of Frame (c) there are visible the reflected images of glowing gas caps at the blunt noses of two pitot probes. These images are a positive indication of the establishment of flow at the downstream location of the pitot probes, and by counting back 0.4 millisecond on the basis of stream velocity and the distance between the probes and the test station, 1.6 milliseconds may be set as the latest time at which flow could have been established at the test station. The earliest possible time of flow establishment is 1.2 milliseconds. This is determined from the fact that no wake from the furnace bottom corner plate is seen in the upper right corner of Frame (b); this wake is visible in Frames (c) and (d).

The end of the contamination-free flow time is determined by "counting back" from the time at which the glowing gas caps on the pitot probes are quenched by H_2 driver gas. Supplementary timing information is obtained by calculation and from the starting time of the postglow which occurs when the starting shock arrives at the end wall of the tunnel. The motion downstream of the test spheres also serves to confirm timing, e.g., the slight movement visible in Figure 28 confirms that flow started between Frames (b) and (c).

- (a) FOUR SPHERES IN
VERTICAL FALL
JUST PRIOR TO
STREAM INCIDENCE

- (b) ONE MILLISECOND
LATER "PRE-GLOW"
MOMENTARILY OB-
SCURES SPHERES

- (c) FLOW ESTABLISHED -
SPHERES SHOW
SLIGHT MOVEMENT
TO RIGHT

- (d) FOUR SPHERES PLUS
TWO ADDITIONAL
WHICH WERE
ORIGINALLY LEFT
OF FIELD
MOVE TO RIGHT

ALL FRAMES ARE
ADJACENT AND ONE
MILLISECOND APART
IN TIME

FIELD SIZE $\approx$
9 INCHES X 6.5 INCHES

- (e) SPHERES CLEARLY
STABLE - BACK-
GROUND GLOW
INCREASES TO
OBSCURE NEXT
FRAME

→ STREAM FLOW

Figure 28. Test Run Number 6

OBSERVATIONS

INFERENCES

NOTE: EACH TIME DIVISION = 0.2 MSEC; FILLED INTERVAL INDICATES SHUTTER OPEN TIME.

Figure 29. Technique of correlating motion pictures with events

Ten of the fifteen tunnel starts in which stream flow was achieved resulted in coincidence of the stream and the ferrous oxide spheres. All of these ten runs are reported and discussed. Reproductions of motion picture frames from nine of the ten runs are presented herein; Run 4 motion pictures could not be reproduced satisfactorily.

Table XV summarizes the test results, and Figures 30 through 37 present the photographs of each run. The table and the figures are arranged in the order of decreasing estimated molten fraction. Photographs of Run 6 were presented in Figure 28. (The original test run numbers are employed herein for convenience in reporting.) Descriptive information on the figures makes them largely self-explanatory, but the following additional comment may be in order.

Note that in several runs fewer spheres are shown in the photographs than are listed in Table XV in the column headed "Approximate Number of Spheres Visible in Proper Flow." This is because some spheres visible in film from cameras other than the one used for these reproductions are included in Table XV. The spheres so included were in a region which did not extend more than about 3 inches below the camera field. As noted in a previous section, no relationship between falling distance and sphere behavior was detected in the 10-inch-high field considered.

The photographs of Runs 12 and 9 (Figures 31 and 34) appear to show no disintegrations, while disintegrations are reported for these runs in Table XV. In Run 12, the sphere at the top of Frame (c) clearly is seen to disintegrate when projected images of either the color or black and white film are examined. The disintegration is surely normal, rather than being due to improper flow associated with the bottom cover plate, inasmuch as a sphere that does not disintegrate can be seen above the one in question in other films (not shown). In Run 9, none of the spheres shown in Figure 34 disintegrate, but one just below the viewing field does and two or more above the field appear to disintegrate.

A small percentage of the spheres falling through the furnace appeared to hit the edges of the hole in the bottom cover plate and were deflected to the right of the vertical path. These deflected spheres were ignored in assessing results. Disintegration is visible, when it occurs, as a wake extending downstream from the sphere.

TABLE XV
Sphere Behavior Recorded on Photographs

Test Number	Figure Number	Estimated Molten Fraction of Radius	Weber Number	Knudsen Number	Time Visible in Stream (ms)	Approximate Number Spheres Visible in Proper Flow	Number of Spheres Which Disintegrate	Remarks
15	30	1.0	3.65	0.213	4 to 5	7	0	One appears to enlarge but does not disintegrate.
12	31	1.0	4.83	0.152	4 to 5	10+	1	
8	32	1.0	9.82	0.093	2 to 3	8	2+	Two clearly disintegrate; several others probably do.
7	33	0.45	15.3	0.0536	1 to 2	10+	Several	Superimposition hampers film study; possibly all disintegrate.
9	34	0.37	10.0	0.090	2 to 3	9	1+	
6	28	0.30	10.4	0.088	3 to 4	7	0	
4	--	0.30	5.24	0.162	4 to 5	1	0	
13	35	0.19	5.00	0.149	3 to 4	7	0	
11	36	0.19	10.25	0.088	2 to 3	15	5 to 15	Five clearly disintegrate; others possibly do.
1	37	0 to 0.04	4.07	0.118	2 to 3	10	0

(a) **FOUR SPHERES
FALLING VERTICALLY**

(b) **FLOW INDICATED
BY SPHERE MOVEMENT**

(c) **NO
DISINTEGRATION**

(d)

(e)

**ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME**

**FIELD SIZE $\approx$
9 INCHES X 6.5 INCHES**

→ STREAM FLOW

Figure 30. Test Run No. 15

(a) ELEVEN
SPHERES
FALLING
VERTICALLY

(b) FLOW

(c) SPHERE AT
EXTREME
TOP OF FRAME
DISINTEGRATES
(SEE TEXT)

(d) NO
FURTHER
DISINTEGRATIONS

(e)

ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME

FIELD SIZE $\approx$
9 INCHES X 6.5 INCHES

→ STREAM FLOW

Figure 31. Test Run No. 12

- (a) FIVE SPHERES
IN FRAME - (LATER
FRAMES SHOW BY
ACCELERATION
THAT HOT OBJECT
AT BOTTOM IS NOT
A SPHERE)

- (b) FLOW
BEGINS

- (c) DISINTEGRATION
EVIDENT

- (d) FURTHER
DISINTEGRATION -
GLOW NEAR
OBSCURATION

ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME

FIELD SIZE $\approx$
9 INCHES X 6.5 INCHES

→ STREAM FLOW

Figure 32. Test Run No. 8

(a) TEN SPHERES
VISIBLE IN
FRAME

(b) FLOW BEGINS -
SHOCK CAP
IMAGES IN
UPPER LEFT

(c) DISINTEGRATION
EVIDENT

(d)

ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME

FIELD SIZE $\approx$
9 INCHES X 6.5 INCHES

→ STREAM FLOW

Figure 33. Test Run No. 7

(a) SIX SPHERES
VISIBLE IN
VERTICAL
FALL

(b) FLOW
ON

(c) NO
DISINTEGRATION
IN FRAME, BUT
SEE TEXT

(d)

ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME

FIELD SIZE $\approx$
9 INCHES X 6.5 INCHES

→ STREAM FLOW

Figure 34. Test Run No. 9

(a) **FOUR
SPHERES
VISIBLE
IN FRAME**

(b) **FLOW
ON**

(c) **NO
DISINTEGRATION**

(d)

(e)

→ **STREAM FLOW**

**ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME**

**FIELD SIZE
9 INCHES X 6.5 INCHES**

Figure 35. Test Run No. 13

(a) TEN SPHERES
IN FRAME

(b) FLOW
BEGINS

(c) DISINTEGRATION
BEGINNING IN
ONE SPHERE

(d) SEVERAL
DISINTEGRATIONS
EVIDENT

(e)

ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME

FIELD SIZE $\approx$
9 INCHES X 6.5 INCHES

→ STREAM FLOW

Figure 36. Test Run No. 11

(a) ABOUT TEN
SPHERES
VISIBLE

(b) FLOW
ON

(c) NO
DISINTEGRATIONS

(d)

ALL FRAMES
ARE ADJACENT
AND ONE
MILLISECOND
APART IN TIME

FIELD SIZE $\approx$
15 INCHES X 10 INCHES

→ STREAM FLOW

Figure 37. Test Run No. 1

Discussion of Results--Fully Molten Spheres

During each test, some spheres which fell through the furnace also fell to the floor of the test section before the test stream was started. These were collected in silicone oil and were later examined microscopically after cross sectioning, polishing, and etching. It was expected that the etching would reveal crystallographic differences which would indicate the extent to which melting occurred. This was not found to be the case, and melting was estimated by heat transfer calculation and the smoothness condition of the sphere surface.

Photomicrographs of sectioned spheres from representative tests are presented in this report; the spheres from Test 7 were accidentally destroyed.

Figures 38, 39, and 40 show initial specimens from Tests 8, 12, and 15; the specimens were collected after they fell through the furnace too early to encounter the test stream. The indicated furnace temperature was either 4380° or 4390° R. The specimens can be compared with a similar specimen from Test 1 (Figure 41) in which the indicated furnace temperature was 3800° R, and with a specimen not subjected to the furnace heating (Figure 27c).

Figure 38. Specimen from Test 8-- 4380° R (etched, 150X)
(material lost from left side in sectioning)

Figure 39.
Specimen from Test 12--
4380°R (etched, 150X)

Figure 40.
Specimen from Test 15--
4390°R (etched, 150X)

Figure 41.
Specimen from Test 1--
3800°R (etched, 150X)

The high furnace temperature used in Tests 8, 12, and 15 greatly changed the appearance of the specimens. The external surfaces became much smoother, more reflective, and more circular. The porosity (compare Figures 40 and 41) changed from irregular shapes to more spherical shapes which have a smoother surface. This change in appearance is taken as good evidence that an indicated furnace temperature of 4380°R is sufficient to cause complete melting.

Figure 42 shows the results of the tests as functions of Weber number and furnace temperature. On the basis of the appearance of the specimens shown in Figures 38 through 43, specimen 1 on the chart 3800°R is labeled not melted, and specimens 8, 12, and 15 at 4380°R or above are labeled fully melted. The fully melted specimens did not break up at a Weber number calculated as 3.85, slight breakup was observed at 4.83, and much breakup occurred at 9.82. This is in general agreement with the work of previous programs in continuum flow in which the critical value of Weber number was found to be near 6. The fact that breakup was observed at a Weber number of 4.83 suggests that, in these tests in transition flow, the critical value may be slightly less than 6.

In attempting to assign an uncertainty to the calculated value of 4.83, ± 5 percent is associated with knowledge of specimen diameter and ± 3.5 percent is associated with the product of stream density and the square of velocity. There is little basis for associating an accuracy figure with the surface tension. The determinations described in Reference 20 show a very high degree of repeatability (± 0.77 percent) with no consistent variation over the small temperature range (12°C) of the five tests. Ignoring uncertainty in the value of surface tension, and there appears to be no basis for doing otherwise, the calculated value of Weber number is 4.83 ± 8.5 percent, that is, in the range from 4.4 to 5.2.

For comparison, in the experimental work in continuum flow at low velocity (reported in Reference 21) breakup was observed in one case at a Weber number of 5.1 and was not observed in another case at 5.2.

Figure 42.

Breakup phenomena as a function of furnace temperature

Figure 43.
Exterior of FeO specimens after being fully molten; Test 12 (50X)

While Test 12 suggests that in transition flow the critical value of Weber number may be slightly lower than it is in continuum flow, it is conceded that this result is not definitive, and the critical value may well be considered to be near its customary value, that is, near 6.

Discussion of Results--Partially Molten Spheres

Evidence for the association of partial melting at intermediate furnace temperatures in the temperature range of 4150° to 4300°R takes three forms. Gross evidence that spheres exposed to this temperature range were neither fully molten nor fully solid is provided by the test results. For example, much breakup was observed in Test 11 at 4150°R (Figure 42), suggesting that the spheres were at least partially molten. In tests at a Weber number near 5, breakup did not occur at 4150°R (Test 13) but did occur at 4390°R (Test 12). A logical explanation of this difference is that a furnace temperature of 4150°R produces partially rather than fully molten spheres. Additionally, this result suggests the overall conclusion that partially molten spheres are more stable than fully molten ones.

Figures 44 through 48 show specimens which were exposed to intermediate furnace temperatures. Their appearance is consistent with a belief that partial melting occurred. For example, those heated to 4150°F show loss of detailed roughness of the external surface but little change toward the more spherical shapes of Figures 38 through 40. Some progressive changes toward the fully molten state seem evident on the specimens at progressively higher temperatures. It is disappointing that the photographic evidence of the partially molten state is not more clearly defined.

Figure 44.

Specimen from Test 6--4230°R
(etched, 150X)

Figure 45.

Specimen from Test 4--4230°R
(etched, 150X) (some decomposition believed evident)

Figure 46.

Specimen from Test 13--4150°R
(etched, 150X)

Figure 47.

Specimen from Test 11--4150°R
(etched, 150X) (centers of available specimens lost in sectioning)

Figure 48. Specimen from Test 9--4270°R
(etched, 150X) (some decomposition believed evident)

A third type of evidence of the extent of melting derives from the heating which occurred as the specimens fell through the furnace. During the preliminary heating of the specimens in the hot tubular furnace, heating by radiation greatly predominated over convective heating inasmuch as the pressure was low, typically only 10 microns of mercury. The total heating time may be thought of as divided into two parts. The first part is the time required to raise a sphere from its initial temperature, T_1 , to its melting point, T_m . The second part is the time for supplying latent heat of fusion. T_1 and T_m are expressed by the first and second terms, respectively, on the right-hand side of the following equation:

$$t = \int_{T_1}^{T_m} \frac{CU}{\sigma \epsilon A} \frac{dt_s}{(T_F^4 - T_s^4)} + \frac{(\text{wt fraction melted}) \rho U F}{\sigma \epsilon A (T_F^4 - T_m^4)}. \quad (7)$$

For the situation in which only the furnace temperature is varied and T_{F1} is the furnace temperature required to raise a sphere to the melting point without causing any melting and T_{F2} is a higher furnace temperature which causes some melting, the equation can be simplified to

$$\int_{T_1}^{T_m} \frac{dt_s}{T_{F1}^4 - T_s^4} = \int_{T_1}^{T_m} \frac{dt_s}{T_{F2}^4 - T_s^4} + \frac{(\text{wt fraction melted})F/C}{T_{F2}^4 - T_m^4} \quad (8)$$

from which the fraction melted may be evaluated as a function of T_{F2} if appropriate values are known for the other terms.

Only the values of latent heat and specific heat are needed when using Eq. (8) because emissivity, volume, area, and time cancel out as long as they remain constant. Using the latent heat value from Reference 22 and the specific heat value from Reference 19, F/C is $980^\circ R$. Using this value and taking T_1 as $1000^\circ R$, the equation was solved. If one accepts the photographic evidence of complete melting with the furnace temperature at $4380^\circ R$ (i.e., $T_{F2} = 4380^\circ R$) and the weight fraction melted as unity, use of Eq. (8) to calculate the furnace temperature, T_{F1} , required to raise a specimen to the melting point but with no melting results in a value of $3940^\circ R$. Therefore, one can conclude that some melting occurred in Tests 11 and 13 in which the furnace temperature was $4150^\circ R$.

Numerical values were assigned to the extent of melting by using Eq. (7) and estimating the time of fall through the hot zone of the furnace. The electrodes at each end of the furnace were water cooled, and in preliminary tests their temperatures were near $1000^\circ R$. Because of the cool electrodes, the end zones of the graphite tube were cooler than the central region. To allow for this, the effective hot zone at the measured furnace temperature was taken as 22 inches long beginning 4 inches below the point of release of the spheres; that is, the effective hot zone was assumed to begin 1 inch below the bottom of the upper electrode and to extend to 1 inch above the top of the bottom electrode. These figures were set primarily on an arbitrary basis. It is possible that a traverse of temperature through the length of the furnace could be made, although instrumenting for this would be difficult in this

temperature region; it is questionable whether greater accuracy on this point would be a dominant factor in justifying the method of estimation as one of high precision. These values yield a fall time through the hot gas of 0.223 second.

In the calculation, the specific heat C was taken from Reference 19 as 0.20 Btu/lb $^{\circ}$ R; density, ρ , was taken from Reference 19 as 332 lb/ft 3 ; emissivity, ϵ , was taken from Reference 19 as 0.90; latent heat, F , was taken as 196 Btu/lb. Solving the above equation, using these values, yields the plots of Figure 49. The plots agree with the previously mentioned conclusion, based on observation of spheres collected, that complete melting occurred at a furnace temperature between 4300 $^{\circ}$ and 4380 $^{\circ}$ R. The calculation merely provides a method of extrapolating from such a point to lower furnace temperatures. A different arbitrary assumption about the effective length of the hot zone would shift the calculated values.

Figure 49. Estimated fraction melted

Based on Figure 49, the estimated extent of melting in the tests is given in Table XVI.

TABLE XVI
Estimated Extent of Melting

<u>Test</u>	<u>Furnace Temperature (°R)</u>	<u>Weight Fraction Molten</u>	<u>Radius Fraction Molten</u>
15 and 12	4390	1.0	1.0
8	4380	1.0	1.0
7	4300	0.83	0.45
9	4270	0.75	0.37
4 and 6	4230	0.65	0.30
11 and 13	4150	0.46	0.19
1	3800	0	0

Conclusions

For the porous specimens tested and for the transition flow region used in the tests, it is concluded that: (1) the critical Weber number for fully molten spheres is near, but perhaps slightly lower than, the value of 6 customarily accepted for continuum flow; (2) the specimens which can be reasonably believed to have been heated to the partially molten state exhibited less tendency for breakup than did those which were fully molten; and (3) on a chart such as that of Figure 50, the breakup region occurs at the upper right and the stable region at the lower left, with the boundary between these regions being near a Weber number of 5 or 6 for fully molten spheres and near a Weber number of 10 for the extent of melting in Tests 11, 6, and 9.

Figure 50. Breakup phenomena as a function of furnace temperature

Small Particle Ablation Experiments in a Free-Flight Gun Range

A free-flight gun range provides the best ground simulation of the complete reentry environment. Since the particle is in free flight into a preselected density at a predetermined velocity, all forces that exist in an actual reentry at the same density and velocity combination also exist in the gun range. Therefore, experiments to investigate the heating and ablation of small spherical particles were undertaken.

Under Sandia contract, the initial program was undertaken in the General Motors Defense Research Laboratories (now ACDRL) Hypervelocity Range C which was constructed for approximately 60 feet of free flight. The program was undertaken as a feasibility study with the stated purpose of determining the feasibility of making quantitative observations on small (200 to 800 micrometers diameter) ablating spherical particles traveling in air at velocities in the range of 20,000 to 25,000 fps. To accomplish this, the gun range was set up in the configuration shown in Figure 51. Three Q-switched laser stations, together with an open shutter streak camera and four spark shadowgraph stations, provided data on ablation and velocity of the particles.

Figure 51. Free-flight gun range configuration

Crystalline aluminum oxide (Al_2O_3) spheres were used in the experiments as a simulant for the plutonium dioxide fuel material. The conditions and results of the program are presented in Table XVII. Details of the program are presented in Reference 23.

TABLE XVII

Summary Table of Tests Conducted in Gas Gun Feasibility Study

Round No.	Model mass (mg)	Model diam. (μ m)	Chamber pressure (mm Hg)	Launch velocity (m/msec)	Launch weight (g)	Impact comments
1185	0.72	700	25	7.65	0.44122	
1186	0.72	700	25	7.83	0.43742	1 large crater, 5 small
1190	0.72	700	25		0.43812	1 large crater, 5 small
1191	0.72	700	25	7.80	0.44032	Foil, 1 large crater, 2 small
1192	0.72	700	25		0.44602	Foil, 1 large crater, 2 small
1193	0.72	700	25		0.44672	
1194	0.72	700	25		0.45612	
1195	0.72	700	25	7.21	0.45332	Foil destroyed, 1 large crater, 1 small
1196	0.72	700	25	7.28	0.45602	Foil, 1 large crater, many small
1197	0.72	700	50	7.16	0.47802	Block, 1 large crater, clean
1200	0.72	700	50	7.27	0.45102	Block, 1 large crater, 1 small
1201	0.72	700	40		0.42142	Block, 1 large crater, 1 medium and 1 small
1202	0.26	500	25	7.77	0.45036	Few small craters
1203	0.26	500	25		0.44116	
1204	0.26	500	15		0.45526	1 large crater
1205	0.13	400	20		0.44683	
1207	0.13	400	20		0.44203	
1209	0.26	500	25	7.77	0.44266	Good data, 1 crater
1210	0.13	400	25	7.91	0.44133	
1211	0.26	500	35	(7.73)	0.4499	

From Table XVII it can be seen that for several runs a single large impact crater and several smaller ones were found on the impact plates or sheets of foil placed at the end of the range. The laser photographs for certain of these runs clearly showed irregular front surfaces on the spheres from which some mass had been removed. Although melting was calculated by ACDRL to occur at certain of these conditions, the conclusion reached by ACDRL was that excessive thermal stresses in the crystalline alumina resulted in pieces of solid material breaking from the sphere. The low thermal diffusivity of alumina results in a very thin layer of temperature penetration on the surface which might be expected to result in layers of the particle flaking off prior to formation of a melt layer. ACDRL concluded that it is possible to launch and measure the velocity of individual spheres from 400 to 700 micrometers in diameter traveling at velocities near 25,000 fps. Also, definitive laser shadowgraphs of these high velocity spheres could be obtained. It was also concluded that mechanical breakup or disintegration rather than liquid layer ablation occurred at the flight conditions used in these experiments.

Based on the results of this initial feasibility study, a program was outlined to further study liquid layer ablation in the free-flight environment. A contract was again placed with ACDRL for launching small spherical particles and photographing their response in the 120-foot gun range.

Extensive thermal calculations were performed by Sandia using the SPHERE 2 program to determine what conditions would be most appropriate for ablation studies. As previously pointed out, the Weber number is considered an important parameter in a mass loss study. The stability analyses performed by Aerotherm Corporation and GE/VFSTC suggested that Weber numbers on the order of 1 to 6 are necessary to investigate the region dividing the stable-unstable condition.

Since Weber numbers and stagnation heating are both functions of the flight velocity and gas density, the requirement for a relatively low Weber number is not totally consistent with having melting occur early in the flight along a limited length of test chamber. It was not feasible to test materials with high melting temperatures at low Weber numbers in the available 120-foot long range. Attention was turned to

materials with lower melting temperatures which were also inert to chemical reactions. In particular, gold microspheres were thought to offer the most suitable compromise for the ACDRL 120-foot range.

At the request of AEC/SNS and the Martin Marietta Corporation, 8 shots of the total of 30 required in the contract with ACDRL were to be conducted using Al_2O_3 . The test objectives stated by the Martin Company were to investigate: (1) mass loss due to liquid layer stripping, and (2) possible Weber number effects. Suggested flight conditions are given in Table XVIII together with the results of thermal analyses performed by Sandia using the SPHERE 2 program.

TABLE XVIII
Theoretical Predictions for Alumina Spheres

Pressure (mm)	Velocity (fps)	Diameter (μm)	Calculated Weber Number at Melt	Calculated Distance Downrange at Melt Initiation (ft)	Percent Melt at End of Range
3	20,000	400	47.4	83	2.93
5	20,000	400	74.5	41	4.63
10	20,000	400	149.0	22	7.23

The Weber number for these conditions is well above the range of 1 to 10 where the critical value is expected to be. Also, the very small amount of material that is molten prior to reaching the end of the range makes detection of melting, mass loss, and shape change extremely difficult. Polycrystalline alumina spheres formed by melting presized alumina powder were used for these experiments rather than the single crystal material used in the feasibility study. The spheres were sized and weighed by Sandia. The gross density of the particles varied from 3.1 to 3.5 g/cc compared with a theoretical value of 4.01 g/cc.

The results of these eight shots plus two additional shots performed at no cost are shown in Table XIX. For these experiments, laser shadowgraph stations were located downrange at 33, 91, and 108 feet. Martin furnished soft catchers which were located downrange at 118 feet

in an attempt to obtain some information on total mass loss and debris by recovering the particles from the catchers. For some of the shots, aluminum impactors furnished by ACDRL were used in place of the Martin catchers. The experiments were not completely successful because no laser shadowgraphs were obtained due to failure of the triggering systems.

TABLE XIX
Summary of Test Results
of Polycrystalline Alumina

Test No.	Pressure (mm)	Velocity (fps)	Laser data	Impact data	Streak data	Remarks
1	--	--	--	--	--	Model did not fly.
2	--	--	--	--	--	Model did not fly.
3	3	20,000	None	None	None	Voided due to launch cloud and debris.
4	3	20,000	None	Single crater	None	--
5	3	20,000	None	Martin catcher	None	Switched to light-interrupt trigger.
6	3	20,000	First laser station shadowgraph of intact particle	Single crater	None	--
7	5	20,000	None	Two craters	None	--
8	5	20,000	None	Single crater	None	Open shutter camera focused on impact plate photographed impact of the particle but recorded no track of incoming particle.
9	--	--	--	--	--	Model did not fly.
10	10	20,000	None	Single crater	None	Open shutter camera focused on impact plate photographed impact of the particle but recorded no track of incoming particle.
Note: All particles were nominal 400-micrometer diameter. Tests 1 through 4 utilized a light emission trigger system for the laser stations. Failure of the lasers to trigger prompted ACDRL to alter the trigger to a light interruption system for Tests 5 through 8. Only one successful trigger was obtained (Test No. 6, Station No. 1).						

Following these tests an extensive analysis which compared the thermal response of alumina, gold, and plutonium dioxide microspheres was performed. The results of this are shown in Figure 52. From these curves it can be seen that in the 120-foot range alumina does not provide satisfactory responses; for example, the calculation for 25 millimeters of pressure and 20,000 fps velocity indicates that the percentage of melt decreases toward the end of the range as molten material resolidifies. The explanation for this is found in the ballistic

parameter of the fairly low density alumina (compared to PuO_2 and gold). The curves also show that extensive melt can be generated on gold microspheres for pressures as low as 3 millimeters. Therefore, gold was chosen as the material for the remaining experiments in this program.

Figure 52. Comparison of melt response for free-flight gun range

A second series of 7 shots using 400 to 600 micrometers diameter particles was conducted. Because of the unsuccessful results of the first test series, the laser shadowgraph trigger systems were modified by ACDRL in efforts to obtain more reliability. The results of these shots are presented in Table XX as Runs G-1 through G-7. Laser

photographs and good impact data were obtained for most of the shots. The impact surfaces confirmed that debris was generated, and the results will be discussed later. In this series, a microwave trigger system for one laser shadowgraph station was used with excellent success. For the remaining 15 shots, ACDRL agreed to provide this type of trigger system for all laser stations in an effort to improve the reliability of all lasers.

In an attempt to sample the debris ablated from the particle along the flight path, the Aerospace Sciences Division and the Analytical Chemistry Division at Sandia assisted in the study by designing samplers to be placed along the flight path and in analyzing the data obtained. Essentially, the samplers consisted of 4-inch diameter, 12-inch-long cylinders lined with IPC filter paper; 1.5-inch-diameter holes were cut into the end caps on the center line of the cylinder. The intent was to allow the parent particle to pass through the sampler, with some of the debris that was generated spreading out and striking either the front or back end plates. The data from these samplers are presently being analyzed.

Since the goal of this program was to reduce the flight Weber number as much as possible while still retaining liquid layer melting, a high pressure preheat section was incorporated into the facility to heat the particle to near melting. Since the particle was preheated, melting could then be accomplished with a lower density (and/or velocity) and a resultant lower Weber number in the remaining portion of the flight chamber. For these conditions, a fast-acting valve was placed either 23 or 27 feet downrange from the gun muzzle. This initial section was maintained at a preselected pressure such that the particle entered the remaining section of lower pressure air at a temperature near melting. Final heating, melting, and ablation then occurred in the low pressure, low Weber number range.

Table XX presents the data for all experiments conducted with gold. Included are the flight conditions, calculations of melt initiation, Weber number, and general comments regarding the success of the tests.

TABLE XX

Test Conditions for Gold Microspheres Fired in ACDRL Gun Range

Run	Diameter (μ m)	Velocity (fps)	P ₁ [*] (mm)	P ₂ ^{**} (mm)	Weber Number	Calculated Melt Initiation Distance (ft)	Calculated Total Melt Distance (ft)	Comments
G-1	620	21,600	8	8	150	~ 70	>120	Laser photographs show melt and ablation; one large and numerous smaller craters.
G-2	620	21,800	8	8	150	~ 70	>120	Laser photographs show melt and ablation; one large and numerous smaller craters.
G-3	520	21,800	7	7	110	~ 60	~120	Laser photographs show melt and ablation; one large and numerous smaller craters.
G-4	520	21,800	7	7	110	~ 60	~120	Laser photographs show melt and ablation; one large and numerous smaller craters.
G-5	520	21,900	2	2	31.5	>120	>120	Single impact crater, no melt apparent.
G-6	380	21,500	7	7	80.5	~ 40	~ 72	No laser photographs; impact surface shows fine debris with no large crater.
G-7	380	22,000	2	2	23	~ 90	>120	Photographs show melt; large impact and several smaller ones on plate.
FG-1	380	22,500	12	1	12.5	~ 84	>120	No samplers; single impact crater on impact plate.
FG-2	380	22,150	12	4	47.7	~ 27	~ 72.5	Flew entire range; small craters on sampler 4 with no larger crater.
FG-3	380	22,700	15	1	12.4	~ 65	>120	Struck sampler 2, one large and several small craters.
FG-4	380	18,500	8	1	8.2	--	--	Voided due to pressure leak in flight chamber.
FG-5	380	16,300	8	1	6.4	~120	>120	One larger crater and several smaller ones at sampler 4.
FG-6	380	22,200	12	1	11.8			Struck sampler 2.
FG-7	250	22,400	8	1	6.7	~ 20	~ 73	Few small debris craters; no evidence of primary particle.

TABLE XX
(continued)

Run	Diameter (μ m)	Velocity (fps)	P ₁ [*] (mm)	P ₂ ^{**} (mm)	Weber Number	Calculated Melt Initiation Distance (ft)	Calculated Total Melt Distance (ft)	Comments
FG-8	380	22,150	12	1	11.8	~84	>120	Numerous small craters and single large crater on sampler 2.
FG-9	380	22,200	12	4	47.7	~27	~ 72.5	Numerous small impact craters on all sampler surfaces.
FG-10	380	21,600	15	1	11.2	~27	>120	Single large crater and numerous smaller ones at sampler 2.
FG-11	380	21,800	15	1	11.3	~27	>120	No evidence of projectile flight.
FG-12	250	23,000	7	1	7.05	~27	~ 48	Two large craters and numerous smaller ones at sampler 2.
FG-12	250	22,900	7	1	7.0	~27	~ 48	Projectile and piston hit sampler 1.
FG-13	250	23,000	7	1	7.01	~27	~ 48	Struck sampler one; single crater.
FG-14	250	22,850	7	1	7.0	~27	~ 48	Flew entire range; one large crater and several smaller ones.
FG-15	380	22,900	15	1	12.5	~27	~102	Single large crater and numerous smaller craters on sampler 4; small craters on others.
FG-16	250	18,200	12	1	5.5	~70	~100	Struck sampler 2, single large crater.
[*] P ₁ - Pressure in preheat section. ^{**} P ₂ - Pressure in flight section.								

Discussion of Gas Gun Experiments

Complete results from these experiments are not available at this time. The impact surfaces are being analyzed to correlate the quantity and size of the debris which was ablated from the microsphere and impacted the surfaces at hypervelocity or was collected in the filter paper. Thermal neutron activation techniques are being employed to analyze the debris contained within the filter paper which was placed within the samplers. All data from those analyses will be reported in separate reports in the future, and only the thermal data are reported here.

The laser photograph system functioned with only moderate success during the experiments. Several changes were made to the system during the experiments, but the combination of small microsphere size, large degree of ablation for some shots, and degree of scatter in the flight path made for less than total success, particularly with the lower Weber number runs.

Runs G-1 through G-7 (see Table XX) were conducted as a series with no preflight chamber and at rather high Weber numbers. Weber numbers varied from 23 to 150, and laser photographs and impact data were obtained. When one compares the calculated melt distances with the laser photographs and impact plates, certain agreement is apparent. Figure 53 is a laser photograph of the particle in flight in test Run G-2. The impact plate for this run revealed a large crater and several smaller ones, indicating liquid layer stripping but not total particle breakup. The thermal calculations agree with this in that they show that total melting would not occur. Figure 54 shows an ablating particle for test Run G-3, and Figure 55 shows the same for test Run G-4. Again the impact plates show liquid layer stripping from a less-than-totally molten particle. Thermal calculations agree that less than total melt will occur in 120 feet. Figure 56 shows the particle in flight in Run G-7, and ablation is not clearly obvious. However, again the impact plate showed partial melt and ablation. No laser photographs were obtained for Runs G-5 or G-6; however, comparison of the impact plate data and the thermal calculations show agreement for the state of particle melt at the position of the impact plate. In general, this segment of the tests presents very conclusive data that the thermal response of the particles is satisfactorily predicted and that liquid layer stripping for Weber numbers from 23 to 150

will occur for partially molten particles within a range of this length. The rate and size of the mass removed is not available. Figures 57 and 58 show impact surfaces from partially ablated particles; Figure 59 presents the single impact for a nonablating condition, and Figure 60 presents the impactor photograph for a totally molten condition.

In the next series of tests, Runs FG-1 through FG-8, attempts were made to obtain ablation data at lower Weber numbers (5 to 45) and to obtain information on the quantity and size of the resulting debris. The laser shadowgraph systems were completely unsuccessful in this series in providing any meaningful data. However, the samplers provided more data confirming the thermal response calculations. With the exception of Runs FG-1 and FG-5, the results of the thermal calculations agree with the type of craters (i.e., one large crater, single large crater and several small ones, or all small craters) found on the impact surfaces. In summary, liquid layer stripping from partially molten particles was demonstrated for Weber numbers as low as near 12.0, and total breakup was seen for a Weber number near 48 in the 120-foot flight.

Runs FG-9 through FG-16 of Table XX were the final tests conducted under this contract. The purpose of this sequence was to determine the ablation characteristics of a partially molten microsphere at lower Weber numbers. As before, the calculated thermal response is generally in agreement with the reported impact surface data. It was shown that debris is stripped from a partially ablated particle at Weber numbers as low as 7.0 (see Runs FG-2 and FG-14). The final shot of the series was conducted at a Weber number of 5.5. The particle struck sampler 2 at 97 feet from the gun muzzle with only a single crater. For this condition melting was calculated to begin at about 70 feet. Whether a Weber number of 5.5 is stable or whether the slight melt and/or short distance traveled after melting began resulted in the lack of ablated debris cannot be definitely stated from the data available.

Figure 53. Laser shadowgraph of 620- μ m gold particle--Run G-2

Figure 54. Laser shadowgraph of ablating 520- μ m gold particle--Run G-3

Figure 55. Laser shadowgraph of ablating 520- μ m gold particle--Run G-4

Figure 56. Laser shadowgraph of 380- μ m gold particle--Run G-7

Figure 57. Impact surface for partially ablated
620- μ m gold particle--Run G-2

Figure 58. Impact surface for partially ablated
380- μ m gold particle--Run G-7

Figure 59. Impact surface for nonablating
520- μ m gold particle--Run G-5

Figure 60. Impact surface for totally ablated
380- μ m gold particle--Run G-6

DISCUSSION OF RESULTS

In considering the results of these studies, emphasis will be placed on pointing out the areas where the data from all sources appear consistent and also where particular areas need further study before conclusive results can be stated.

The thermal response of the microspheres appears to be satisfactorily described with the Sandia SPHERE 2 computer program. As stated earlier, independent studies were performed under contract to provide spot checks on the Sandia results, and the independent and the Sandia results were in general agreement. A modification of the SPHERE 2 program, called ARCJET, was used to calculate the thermal response of the microspheres in the arc tunnel experiments. Considering the inherent variation from run to run in the arc tunnel conditions, good agreement between the observed and calculated times to initiate melting resulted.

The analytical treatments of liquid layer stripping and/or droplet breakup are less conclusive. Whereas the results of the General Electric studies showed the liquid layer to be more stable than the totally molten droplet, the results of the Aerotherm analyses showed precisely the opposite. Since the analyses were formulated in completely different manners, it is not too surprising that the results were contradictory. Since neither of these analyses was verified experimentally and since they both suffer certain restrictive assumptions in their formulations, it cannot be stated on an unqualified basis which, if either, of the analyses is applicable to the reentry mass loss from small spherical particles.

The results of Reynolds⁸ reveal several interesting features. Possibly the most important is that thin liquid layers are more unstable than totally molten droplets. Reynolds concluded that instabilities at large Weber numbers (~ 6) are due to Taylor instabilities, while those at lower Weber numbers (~ 1) result from the Craik-Benjamin instabilities. Reynolds states that the Craik-Benjamin instability is caused by the disturbance shear stress acting on the liquid. He reports that the fluid tends to be piled up at the wave crests when the shear

stress is in phase with the wave slope and that this mechanism is most effective when the layer is very thin. Since this type of instability plays an important role in free molecular flow, Reynolds concludes that the shear stress must be included for a stability analyses to be meaningful.

It is appropriate to suggest that basic experimental studies to verify the validity of these results should be performed. Several methods to check these analyses might be suggested. In an attempt to obtain data which might be used to verify the stability theories, Sandia retained CAL to construct and demonstrate a vertical low-density test facility. In this facility spheres of materials with known physical properties would be suspended in the hypersonic low-density test stream at conditions where the drag force would balance the force of gravity. Details of the melting and mass loss would be studied by photographic means. At this time the facility has been constructed and certain modifications are being made prior to efforts to demonstrate its feasibility. Because of budget limitations any experiments which might have been performed following the feasibility study were canceled. However, the need for these data remains.

The data from the arc tunnel experiments provide evidence of the thermal response and also information on the ablative response of the actual fuel material. Comparison of observed melt initiation times in the experiments with calculated values provides generally good agreement.

The debris data obtained from the impact samplers in the arc tunnel present unqualified evidence that debris was lost from the microspheres at heat flux levels and in periods of time that are representative of an actual reentry environment. The mechanisms that are responsible for this mass loss are not known, but several sources will be discussed.

Data were obtained for heat flux values representing release from well above the melt zone to values representing lower altitude releases well within the melt zone. For the condition above the melt zone, no melt was expected and none was observed. However, as reported in a previous section, debris was collected on the impactors; its source was attributed to the quality of the microsphere surface.

For the test conditions representing release near the edge of the melt zone, a much larger quantity of debris was found on the samplers. During the exposure, the very gradual flattening of the particles confirmed the slow melting rate. However, melting was evident from the movements of the surface. Ripples, wave motions, eruptions, and protuberances were noted. No large flanges or large droplets were noted. Calculations to estimate the mass loss by vaporization indicated that this phenomenon could be a major source of the debris. However, the micrometer size debris found on the samplers is not attributed to vaporization. It is interesting to note that all three test conditions representing release near the melt zone edge plot in the unstable region according to the results of Reynolds' analysis.

Melting at a heat flux value associated with release within the melt zone was more evident. The motion and movement of the liquid layer were apparent, and in some cases surface eruptions were noted. Much of the debris at this condition was too large to be attributed to vaporization. In addition, based on the criteria of Reynolds as applied to these particles, this condition is unstable.

The most severe test condition, representing release well within the melt zone, caused very rapid melt and generally large droplet runback. Much large debris was noted for this condition, but its source is questionable because of the breakup of larger droplets on the sting.

Generally the melting of the microspheres did not result in large droplets or runback (except for Condition M). Therefore, it can be stated that the source of the debris was not the breakup of large droplets. Although no mass was seen to leave the melt surface, with the exception of a limited number of eruptions, the films suggest that the source of the debris is some mechanism associated with the liquid layer, but not breakup of runback material. Finally, it is of interest to note that the data from these experiments are in total agreement with the theory of Reynolds when the theory is modified to eliminate any deceleration effects. However, the data are opposite in trend to the results of GE, although the results of that analysis were not modified to eliminate deceleration.

On the other hand, the experiments performed at CAL are interpreted by CAL to indicate that the critical Weber number for breakup increases with decreasing melt thickness. This agrees with the qualitative results of the GE analyses and appears contrary to the Aerotherm results. While this interpretation of the data may be reasonable in determining total breakup of a particle, there are several features of the experiments which in this authors opinion prevent complete conclusions from being drawn from the CAL experiments. First, since the degree of melting could not be measured and the spheres contained voids of unknown sizes and locations, the degree of melting was subject to some uncertainty. In addition, the approximately 2 to 5 milliseconds run time for the facility may be sufficient to allow total breakup of a liquid or liquid/solid body, but there is no assurance that mass loss cannot occur by liquid layer stripping in some manner such that the experiment run times were too short. Moreover, it is extremely unlikely that mass loss by any means other than chaotic breakup would be apparent on the film. Since the actual problem is one of reentry in which the times of interest can be as long as from 0.5 to 1.0 second, additional data are necessary regarding the possibility of mass loss by means other than total breakup (i.e., liquid layer stripping).

The experiments conducted in the ACDRL free-flight gas gun provided data from a facility where the complete reentry environment is simulated. However, because of the finite length of the range (120 ft), certain compromises were necessary. Gold microspheres were fired at conditions resulting in Weber numbers from near 5.5 to approximately 150. The debris samplers provided conclusive evidence that molten material is stripped from the partially molten particles for Weber numbers as low as 7.5. Total melt and breakup was observed for Weber numbers near 45.

The impact data shown in Table XX provide data regarding whether the microsphere was totally or partially molten. Comparison of these data with the calculated melt initiation distances provides confirmation of the calculated thermal response.

Because of the limited length of the gas gun range, conditions resulting in Weber numbers of primary interest, from 1 to 6, were not attained and as a result no experimental confirmation of either the GE

or Aerotherm stability analyses was provided. However, the experiments illustrate that liquid layer stripping is a real mechanism of mass loss, and further experiments in this type of facility should be pointed toward obtaining mass loss data at lower Weber numbers (from 1 to 6).

In summary, it might be well to recall the overall objectives of the program--that is, to develop an analytical technique, verified by experimental data, which could be used to predict the melting and mass loss resulting from the reentry of small spheres. A portion of these objectives have been accomplished. The calculation technique used to calculate the thermal response of the spheres appears sound. Although no technique to calculate the mass loss has been developed, much data on the mass loss response of actual fuel material have been generated. In analyzing the potential safety hazard resulting from the exposure of plutonium dioxide microspheres in reentry, one should expect that some mass loss, in the form of micrometer and millimicrometer size debris, will occur. However, no general agreement on the conditions that will result in an unstable liquid layer and cause mass loss has been found. Consequently, the complete unqualified answer to the question of conditions of mass loss, together with the rate of loss and the size of the debris as a function of release conditions, must await further study.

APPENDIX A

Sample Calculation for Heat Flux to a
Microsphere in the Hyperthermal Arc Tunnel

Blank Page

APPENDIX A

Sample Calculation for Heat Flux to a Microsphere in the Hyperthermal Arc Tunnel

Because of the inability to obtain a calorimeter as small as the microspheres tested in the arc tunnel, calculations must be relied upon to determine the stagnation heat flux. Table III presents the pertinent flow parameters for the test conditions used. Essentially, a relatively large blunt nosed calorimeter was used to determine the gas enthalpy, and the necessary pressures were measured. Calculations are made from these data for the heat flux to a microsphere, and a sample calculation is given below.

For Condition B, the following parameters were measured

Plenum pressure, $P_0 = 0.40$ atm

Chamber pressure, $P_1 = 0.00619$ atm

Model pressure, $P_{t2} = 0.0737$ atm

$q\sqrt{R}$ (on 0.125-inch diameter calorimeter) = 106

From Equation (4):

$$H_s = 106/0.039 (0.0737)^{1/2} = 10,000 \text{ Btu/lb.}$$

With this enthalpy and plenum pressure, the local properties of the gas are calculated using the computer program developed by Farnsworth.¹² For frozen flow through the nozzle and an area ratio 9 expansion, one gets

$$V_\infty = 12,800 \text{ fps}$$

$$\rho_\infty = 2.69 \times 10^{-5} \text{ lb/ft}^3$$

$$T_\infty = 2156^\circ \text{R}$$

$$M_\infty = 4.37$$

$$C_O = 0.259 \text{ mole/mole}$$

$$C_N = 0.442 \text{ mole/mole}$$

$$C_{N2} = 0.298 \text{ mole/mole}$$

From the above composition

$$\Delta H_c = \frac{(0.259)(16)6600 + (0.442)(14)14,400}{(0.259)16 + (0.442)14 + 0.298(28)} = 6250 \text{ Btu/lb}$$

where

$$H_O = \text{dissociation energy of oxygen} = 6600 \text{ Btu/lb}$$

$$H_N = \text{dissociation energy of nitrogen} = 14,400 \text{ Btu/lb.}$$

From normal shock tables for $M_\infty = 4.37$,

$$\rho_2/\rho_1 = 4.8.$$

$$\text{Then } \rho_e = 4.8 (2.69 \times 10^{-5}) = 1.29 \times 10^{-4} \text{ lb/ft}^3.$$

From Reference 24 at the total temperature,

$$\mu_e = 7.5 \times 10^{-5} \text{ lb/ft-sec.}$$

Assuming that the surface of the plutonium dioxide is noncatalytic to surface recombination of the oxygen and nitrogen atoms in the gas, $\phi = 0$, as used in Equation (6).

Using the above properties and taking the Prandtl number as a constant equal to 0.71, we have from Equation (6),

$$q_{BL} = 0.763 \sqrt{\frac{1.28 \times 10^4}{4.1 \times 10^{-4}}} (1.29 \times 10^{-4} \times 7.5 \times 10^{-5})^{0.5}$$

$$\frac{1}{(0.71)^{0.6}} [0.208(1.792)]^{0.25} (10,000 - 6,250)$$

$$q_{BL} = 1550 \text{ Btu/ft}^2\text{-sec.}$$

For a 250-micrometer diameter particle at these test conditions, the boundary layer heat flux value must be corrected for noncontinuum flow. To do this, we will make use of Eq. (5) where the Reynolds number function is given by Figure A-1 which is taken from Reference 7. The shock Reynolds number is calculated from

$$Re = \frac{V_{\infty} \rho_{\infty} R}{\mu_e} = \frac{(1.28 \times 10^4)(2.69 \times 10^{-5})(4.1 \times 10^{-4})}{7.5 \times 10^{-5}} = 1.9.$$

For $\epsilon = \rho_1/\rho_2 = 0.208$;

then $\epsilon^2 Re = 0.208^2(1.9) = 0.0815$.

From Figure A-1

$$q/q_{BL} = 0.6$$

$$q = 0.6(1550) = 950 \text{ Btu/ft}^2\text{-sec.}$$

This is the calculated stagnation point heat flux on a 250-micrometer diameter microsphere, based on frozen flow through the nozzle and a noncatalytic surface.

Figure A-1. Normalized gas phase heating rate

Similar calculations can be performed assuming equilibrium flow through the nozzle. For this assumption, the following properties are calculated:

$$V_{\infty} = 14,100 \text{ fps}$$

$$\rho_{\infty} = 2.43 \times 10^{-5} \text{ lb/ft}^3$$

$$T_{\infty} = 875^{\circ}\text{R}$$

$$M_{\infty} = 3$$

$$C_O = 0.3 \text{ mole/mole}$$

$$C_N = 0.204 \text{ mole/mole}$$

$$C_{N2} = 0.497 \text{ mole/mole}$$

As before

$$\Delta H_c = \frac{(0.3)(16)(6600) + 0.204(14)(14,400)}{0.3(16) + 0.204(14) + 0.497(28)} = 3400 \text{ Btu/lb.}$$

From Equation (6)

$$q_{BL} = (0.763) \left(\frac{14,100}{4.1 \times 10^{-4}} \right)^{0.5} (3.86 \times 2.43 \times 10^{-5} \times 8 \times 10^{-5})^{0.5}$$

$$\frac{1}{(0.71)^{0.6}} [0.259(1.41)]^{0.25} (10,000 - 3,400)$$

$$q_{BL} = 2900 \text{ Btu/ft}^2\text{-sec.}$$

Correcting as before for nonequilibrium

$$q = 0.42(2900) = 1200 \text{ Btu/ft}^2\text{-sec.}$$

For equilibrium flow and a noncatalytic surface, this represents the stagnation heat flux to the particle. One would expect that the actual heat flux to the particle must lie within these limits. Based on information in published literature, the frozen flow calculations are assumed to represent the more realistic calculation at the relatively low pressures in those experiments.

Calculations for the other conditions were made in a similar fashion.

Blank Page

APPENDIX B

Estimate of Mass Loss by
Vaporization in Arc Tunnel Experiments

Blank Page

APPENDIX B

Estimate of Mass Loss by Vaporization in Arc Tunnel Experiments

Because of the large proportion of debris present on the impactors which was smaller than 0.05 micrometer in diameter, it is of interest to examine the possibility that vaporization may account for this debris. Aerotherm Corporation, in their study for Sandia, calculated the thermochemical mass loss from $^{238}\text{PuO}_2$ microspheres; the results are presented in Reference 4. To estimate vaporization mass loss in the arc tunnel, we will use their results together with certain modifications.

Assuming free molecular flow, an estimate of the rate of surface vaporization is provided by the Langmuir equation

$$(\rho V)\omega = \frac{P_V}{\sqrt{2\pi\bar{M}RT}} \quad (\text{B-1})$$

where

$(\rho V)\omega$ = surface mass loss rate

T = surface temperature

P_V = free molecular vaporization pressure

M = molecular weight

$\bar{R}$ = gas constant.

The free molecular vaporization pressure is defined as

$$P_V = \alpha_L P_{eq} \quad (\text{B-2})$$

where

α_L = Langmuir coefficient

P_{eq} = equilibrium vapor pressure.

Then

$$(\rho V)\omega = \frac{\alpha_L P_{eq}}{\sqrt{2\pi\bar{M}RT}} \quad (\text{B-3})$$

A relationship for the equilibrium vapor pressure is

$$\log_{10} P_{eq} = 8.072 - \frac{29,240}{T} \quad (B-4)$$

where P_{eq} is in atmospheres and T is in degrees Rankine. A convenient form for the vaporization rate is then

$$(\rho V)\omega = 4.7 \times 10^4 P_{eq} \sqrt{M/T} \quad (B-5)$$

where $(\rho V)\omega$ is in $\mu\text{g}/\text{cm}^2\text{-sec}$, P_{eq} is in mm of Hg, and T is in degrees Rankine.

For the problem at hand, we assume that the surface temperature is 4600°R . From Eq. (B-4)

$$\log_{10} P_{eq} = 8.072 - \frac{29,240}{2560} = -3.368$$

$$P_{eq} = 0.00233 \text{ atm} = 1.77 \text{ mm.}$$

Substituting into Eq. (B-5)

$$(\rho V)\omega = (4.7 \times 10^4) (1.77) \sqrt{\frac{0.2422}{270/4600}} = 0.644 \times 10^4 \mu\text{g}/\text{cm}^2\text{-sec.}$$

For a 250-micrometer diameter particle and assuming 70 percent of the surface area is free to vaporize (the remaining surface area is covered by the mounting to the sting),

$$A = 0.7(4\pi)(0.0125^2) = 13.75 \times 10^{-4} \text{ cm}^2.$$

For a reasonable exposure time of 0.1 second, the total mass lost by vaporization is

$$\omega = (0.644 \times 10^4) (13.75 \times 10^{-4}) (0.1) = 0.885 \mu\text{g.}$$

Since the rate of vaporization is extremely sensitive to the surface temperature, assumptions of higher temperatures for the molten surface will result in greater vaporization rates.

REFERENCES

1. Stephenson, A. E., Reentry Thermal Response Analyses of a Plutonium Fuel Form, SC-RR-65-342, Sandia Laboratories, Albuquerque, New Mexico, October 1965.
2. Stephenson, A. E., Reentry Thermal Response Analyses of a Plutonium Fuel Form, Stable Reentry, SC-RR-66-342, Sandia Laboratories, Albuquerque, New Mexico, August 1966.
3. Matting, F. W., and Chapman, D. R., Analysis of Surface Ablation of Non-Charring Materials with Description of Associated Computing Program, NASA-TN-0-3758, National Aeronautics and Space Administration, Washington, D.C., December 1966.
4. Lundberg, R. E., Reynolds, W. C., and Rindal, R. A., A Study of the Behavior of Small Spheres During Reentry, SC-CR-67-2635, Sandia Laboratories, Albuquerque, New Mexico, June 1967.
5. Lees, L., "Laminar Heat Transfer over Blunt-Nosed Bodies at Hypersonic Flight Speeds," Jet Propulsion, 26, 4, April 1956.
6. Watkins, C. B., Sphere 2 - A Two Dimensional Heat Conduction and Trajectory Computer Program for Reentry Heating of Small Spherical Bodies, SC-RR-67-2996, Sandia Laboratories, Albuquerque, New Mexico, December 1967.
7. Gilbert, L. M., Liner, R., Scala, S. M., and Schuyler, F. L., Theoretical Studies of the Reentry of Plutonium Fuel, SC-CR-67-2573, General Electric for Sandia Laboratories, Albuquerque, New Mexico, June 1967.
8. Reynolds, W. C., Hydrodynamic Stability of the Liquid Layer on a Small Object in High Altitude Re-entry, SC-CR-67-2636, Sandia Laboratories, Albuquerque, New Mexico, June 1967.
9. Luna, R. E., and Klikoff, W. A., On Aerodynamic Breakup of Liquid Drops, SC-RR-66-2716, Sandia Laboratories, Albuquerque, New Mexico, June 1967.
10. Sonnenschein, G., and Winn, R. A., High Intensity Thermal Flux Measurements in the Unsteady State, WADC-WCLT-TM-57-93, Wright Air Development Center, July 1957.
11. Rosner, D. E., "Scale Effects and Correlations in Non-Equilibrium Convection Heat Transfer," AIAA Journal, 1, 7, 1963. pp 1550-1555.
12. Farnsworth, A. V., Unpublished work, Sandia Laboratories, Albuquerque, New Mexico.
13. Carden, William H., "Heat Transfer in Non-equilibrium Dissociated Hypersonic Flow with Surface Catalysis and Second Order Effects," AIAA Journal, 4, 10, October 1966.

14. Rosner, Daniel E., "Analysis of Air Arc-Tunnel Heat-Transfer Data," AIAA Journal, 2, 5, May 1964.
15. Wethern, R. J., "Method of Analyzing Laminar Air Arc-Tunnel Heat Transfer Data," AIAA Journal, 1, 7, July 1967.
16. Winkler, Ernest L., and Griffin, Roy N., Jr., Effects of Surface Recombustion on Heat Transfer to Bodies in a High Enthalpy Stream of Partially Dissociated Nitrogen, NASA-TM-D-1146, National Aeronautics and Space Administration, Washington, D.C., December 1961.
17. Deriugin, V., Koh, J. C. Y., and Tong, H., An Investigation of Heat Transfer from Plasma Jets to Surfaces of Various Atomic Recombination Catalyticities, BOE-D2-23253, Boeing Company, June 1964.
18. SC-RR-66-674, Re-entry Studies of Plutonium Fuel-Interim Report, Sandia Laboratories, November 1966.
19. Toloukian, Y. S., Recommended Values of the Thermophysical Properties of Eight Alloys, Major Constituents and Their Oxides, Thermophysical Properties Research Center, Purdue University, Lafayette, Indiana, February 1966.
20. Kozakevitch, Paul, "Tension Superficielle et Viscosité des Scories Synthétiques," Revue de Metallurgie, 46 (8), 9, 1949, p 505.
21. Beal, J. L., et al., Dispersal of Reactor Core Materials by Re-entry Processes, CAL Report I08-100, Suborder No. 1, Cornell Aeronautical Laboratory, May 1963.
22. Grey, Dwight, E., editor, American Institute of Physics Handbook, McGraw-Hill, New York, N.Y., 1957.
23. Collins, D. J., Sangster, D. K., and Rogers, W. K., Feasibility Study on Small Particle Ablation Using a Free-Flight Range, SC-CR-66-2128, Sandia Laboratories, Albuquerque, New Mexico, November 1966.
24. Scala, S. M., and Baulknight, C. W., "Transport and Thermodynamic Properties in a Hypersonic Laminar Boundary Layer - Part 2 Applications," ARS Journal, April 1960, pp 329-336.

DISTRIBUTION:

TID-4500 (52nd Ed.), UC-36 (201 copies)

U.S. Atomic Energy Commission
Division of Space Nuclear Systems
Space Electric Power Office
Washington, D.C. 20545

Attn: G. A. Newby, Assistant
Director (1)
G. P. Dix, Chief,
Safety Branch (1)
R. S. Decker, Jr., Chief,
Safety Branch (1)
R. T. Carpenter, Chief,
Isotope Power Sys. Br. (1)
J. A. Powers, Chief, Isotopes
and Materials Branch (1)
C. Johnson, Chief, Reactor
Power Systems Branch (1)

U.S. Atomic Energy Commission
Division of Isotope Development
Washington, D.C. 20545

U.S. Atomic Energy Commission
Director of Regulation
Washington, D.C. 20545
Attn: C. K. Beck, Deputy Director
F. D. Anderson, Regulation

U.S. Atomic Energy Commission
Division of Biology and Medicine
Washington, D.C. 20545
Attn: J. Z. Holland, Chief,
Fallout Studies Branch
H. D. Bruner, Asst. Director
Medical and Health Research

U.S. Atomic Energy Commission
Space Nuclear Propulsion Office
Albuquerque Extension
Albuquerque Operations Office
P.O. Box 5400
Albuquerque, New Mexico 87115

U.S. Atomic Energy Commission
Albuquerque Operations Office
P.O. Box 5400
Albuquerque, New Mexico 87115
Attn: S. A. Upson, Director
Nonnuclear Activities Div.
J. F. Burke, Director,
Operational Safety Div.

AEC Site Representative
National Aeronautics and Space
Administration
Manned Space Center
Houston, Texas 77058
Attn: W. C. Remini
Bldg. 16, Code ZS-5

Donald W. Douglas Laboratories
P.O. Box 310
Richland, Washington 99352
Attn: S. P. Gydesen

Douglas Aircraft Company
Missile & Space Systems Division
5301 Balsa Avenue
Huntington Beach, California 92646
Attn: Sig Gronich, Program Mgr.,
Nuclear Stages

Deputy I. G. for Insp. & Safety,
USAF
Kirtland Air Force Base
New Mexico 87117
Attn: Col. D. C. Jameson (AFINSR)

General Electric Company
Valley Forge Space Technology
Center
P.O. Box 8555
Philadelphia, Pennsylvania 19101
Attn: S. M. Scala, Room M9539
Space Sciences Lab
G. W. Rivenbark, Isotope
Power Systems Operations

Los Alamos Scientific Laboratory
P.O. Box 1663
Los Alamos, New Mexico 87544
Attn: Dr. L. D. P. King
Dr. Wright Langham
C. F. Metz, CMB-1
F. W. Schonfeld, CMF-5

Isotopes
Nuclear Systems Division
P. O. Box 4937
Middle River, Maryland 21220

Monsanto Research Corporation
Mound Laboratory
P. O. Box 32
Miamisburg, Ohio 45342
Attn: G. R. Grove

Thomas B. Kerr
Code RNS
National Aeronautics and Space
Administration
Washington, D.C. 20546

National Aeronautics and Space
Administration
Ames Research Center
Moffett Field, California 94035
Attn: Glenn Goodwin

DISTRIBUTION (Cont):

National Aeronautics and Space
Administration
Goddard Space Flight Center
Glenn Dale Road
Greenbelt, Maryland 20771
Attn: A. W. Fihelly, Nimbus Project

Dr. D. W. Sherwood
National Aeronautics and Space
Administration
Manned Spacecraft Center (MSC-TD)
Houston, Texas 77058

U.S. Naval Radiological Defense
Laboratory
San Francisco, California 94135
Attn: S. Z. Mikhail, Nuclear
Systems Group

Naval Facilities Engineering Command
Department of the Navy, Code 042
Washington, D.C. 20390
Attn: Graham Hagey

Space Nuclear Propulsion Office
Lewis Research Center
21000 Brookpark Road
Cleveland, Ohio 44135
Attn: L. Nichols

TRW Systems
P.O. Box 287
Redondo Beach, California 90278
Attn: Dr. Donald Jortner

Union Carbide Corporation
Nuclear Division
P.O. Box X
Oak Ridge, Tennessee 37831
Attn: R. A. Robinson, Isotope
Development Center
B. R. Fish, Health Physics
Division

U.S. Public Health Service
Nat. CTR for Radiological Health
1901 Chapman Avenue
Rockville, Maryland 20852
Attn: Nuclear Facilities Section

Jet Propulsion Laboratory
California Institute of Technology
4800 Oak Grove Drive
Pasadena, California 91103
Attn: A. L. Klascius, Radiation
Health and Safety

D. B. Shuster, 1200
J. R. Banister, 5120
J. D. Shreve, 5271
L. E. Lamkin, 7300
G. A. Fowler, 9000
H. E. Viney, 9130
R. C. Maydew, 9320
L. A. Hopkins, Jr., 9500
V. E. Blake, 9510
H. E. Hansen, 9511
B. W. Marshall, 9511
S. L. Jeffers, 9512
S. McAlees, Jr., 9513
J. D. Appel, 9514
ARPIC, 9514 (2)
R. J. Everett, 9515
A. J. Clark, 9520
J. W. McKiernan, 9521
B. F. Hefley, 8232 (5)
B. R. Allen, 3421
L. C. Baldwin, 3412
L. L. Alpaugh, 3412
C. H. Sproul, 3428-2 (10)