176
2/17/69

662

MASTER

SC-CR-68-3470

January 1969

AEROSPACE
NUCLEAR
SAFETY

REPORT OF STUDIES WITH PARTICLES IMPACTED ON
COPPER PLATES FROM THE ABLATION OF SIX $^{238}\text{PuO}_2$
FUEL PARTICLES UNDER CONDITIONS SIMULATING
ATMOSPHERIC RE-ENTRY

PREPARED BY

W. D. HOLLAND
TRACERLAB

A Division of Laboratory for Electronics, Inc.

SANDIA LABORATORIES

OPERATED FOR THE U. S. 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

SC-CR-68-3470

REPORT OF STUDIES WITH PARTICLES IMPACTED ON COPPER PLATES FROM THE
ABLATION OF SIX $^{238}\text{PuO}_2$ FUEL PARTICLES UNDER CONDITIONS SIMULATING
ATMOSPHERIC RE-ENTRY

January 1969

First unrestricted general distribution, October 1968

Prepared by

W. D. Holland

TRACERLAB

A Division of Laboratory for Electronics, Inc.

2030 Wright Avenue

Richmond, California 94804

for

Sandia Corporation

under

Contract Number ASB 48-6048

ABSTRACT

Size distribution analyses were performed on six samples of $^{238}\text{PuO}_2$ particles collected during the ablation of individual SNAP fuel microspheres in a hyperthermal arc-tunnel under conditions simulating atmospheric re-entry.

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.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

SUMMARY

With six samples composed of particles that contained ^{238}Pu , particle studies were performed by autoradiography and electronic alpha counting. The particles, which were collected on polyvinylchloride (PVC) coated copper plates, were collected during the ablation of individual SNAP fuel particles in a 40 kw hyperthermal arc tunnel under conditions simulating atmospheric re-entry. Three re-entry conditions were simulated. Size distribution histograms of equivalent $^{238}\text{PuO}_2$ particle diameters greater than $0.2\ \mu$ were obtained for each sample. These equivalent $^{238}\text{PuO}_2$ particle diameters range from 0.2 to $13.5\ \mu$. Typical radioactive particles were taken from each sample. Results of radiochemical analysis of all particles collected on five of the impactors and of individual particle measurements obtained by the hollow-star and electronic-counting techniques were used to tabulate the mass distributions from zero to the size of the largest particle found. The results of radiochemical analysis indicate that about 95% of the mass of PuO_2 collected is in particles with equivalent $^{238}\text{PuO}_2$ diameters $< \sim 0.1\ \mu$. Duplicate tests performed with samples collected under two re-entry conditions showed outstanding precision in mass distributions, indicating that there were no experimental errors.

TABLE OF CONTENTS

	Page
1.0 TEST DESCRIPTION	1
2.0 SAMPLE DESCRIPTION	5
3.0 DESCRIPTION OF WORK	7
3.1 GENERAL PROCEDURE FOR PARTICLE ANALYSIS	7
4.0 RESULTS	11
4.1 EXPOSURES TO X-RAY FILM	11
4.2 SIZE DISTRIBUTIONS OF $^{238}\text{PuO}_2$ PARTICLES GREATER THAN 0.2 μ IN DIAMETER	11
4.3 RADIOCHEMICAL ANALYSES	11
5.0 CONCLUSIONS	17
6.0 RECOMMENDATIONS	19
REFERENCES	20
APPENDIX A - RADIOAUTOGRAPHS OF IMPACTORS BEFORE AND AFTER WASHING	21
APPENDIX B - HOLLOW-STAR TECHNIQUE	28
APPENDIX C - TYPICAL $^{238}\text{PuO}_2$ PARTICLES	34
APPENDIX D - NUMERICAL SIZE DISTRIBUTION HISTOGRAMS OF $^{238}\text{PuO}_2$ PARTICLES DETERMINED BY THE HOLLOW STAR TECHNIQUE	111
APPENDIX E - CALCULATION OF THE CUMULATIVE MASS DISTRIBUTION VALUES FOR EQUIVALENT $^{238}\text{PuO}_2$ PARTICLES COLLECTED ON IMPACTORS	123
APPENDIX F - STANDARD DEVIATIONS OF ALPHA ACTIVITIES FOR PARTICLES AFTER ISOLATION, AND PERCENTAGES OF INITIAL ALPHA ACTIVITIES RECOVERED	131
APPENDIX G - SPECIAL PROCEDURE FOR RADIOCHEMICAL ANALYSIS OF PLUTONIUM COLLECTED ON IMPACTOR PLATES	133
APPENDIX H - PRELIMINARY REPORT OF STUDIES WITH PARTICLES COLLECTED ON COPPER PLATES FROM ABLATION OF $^{238}\text{PuO}_2$ FUEL PARTICLES NOS. 1-25-3 AND 1-25-20 UNDER CONDITIONS SIMULATING ATMOSPHERIC RE-ENTRY	135
APPENDIX I - PRELIMINARY REPORT OF STUDIES WITH PLUTONIUM PARTICLES ON COPPER COLLECTION OF PLATES FROM ABLATION OF A $^{238}\text{PuO}_2$ FUEL PARTICLE UNDER CONDITIONS SIMULATING ATMOSPHERIC RE-ENTRY	177

LIST OF FIGURES

Figure		Page
1	$^{238}\text{PuO}_2$ Microsphere M-29 Before Testing-Diameter 235.4 Microns	2
2	Diagram Showing Arrangement of the Hyperthermal Nozzle, the Holder for the SNAP Fuel Particle, and the Collection Plate	3
3	Special Copper Collection Plates in Lucite Holders for Shipment	6
4	Diagram Illustrating how Radioactive Particles from a Washed Impactor are Exposed to Nuclear Emulsion Plates	9

LIST OF TABLES

Table		Page
I	Description of Collection Plates Analyzed and $^{238}\text{PuO}_2$ SNAP Fuel Particles Ablated	5
II	Results of Radiochemical Analyses Performed to Determine the Equivalent $^{238}\text{PuO}_2$ Masses Collected on Impactors	12
III	Percentages of Total Mass for Two Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-3	13
IV	Percentages of Total Equivalent Mass for Two Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-15	13
V	Percentages of Total Mass for Three Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-35	13
VI	Percentages of Total Mass for Four Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-29	13
VII	Percentages of Total Equivalent Mass for Four Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-37	13
VIII	Percentages of Total Mass of $^{238}\text{PuO}_2$ Ablated from Microspheres	16

REPORT OF STUDIES WITH PARTICLES IMPACTED ON COPPER PLATES FROM THE
ABLATION OF SIX $^{238}\text{PuO}_2$ FUEL PARTICLES UNDER CONDITIONS SIMULATING
ATMOSPHERIC RE-ENTRY

1.0 TEST DESCRIPTION

The response of radioactive fuel materials upon re-entry into the earth's atmosphere due to orbital decay or an abort situation is an important factor in the safety analysis of SNAP power supplies. Division 9511, Sandia Corporation, has completed a series of experiments to determine the amount and size distribution of debris ablated from $^{238}\text{PuO}_2$ microspheres when exposed to a simulated re-entry environment. These experiments are described in this report, the preliminary tests being included as Appendices H and I.

The $^{238}\text{PuO}_2$ microspheres tested ranged in size from 217.6 to 235.4 microns diameter. Figure 1 shows a typical microsphere prior to testing. The microspheres were mounted on small needle type sting supports and exposed to various different plasma stream conditions simulating re-entry in the Sandia 40 kw hyperthermal arc tunnel. (Figure 2). A polyvinylchloride coated copper impact collection plate was mounted 4 inches downstream from the microspheres to collect the debris ablated during the tests.

The three test conditions, A, K, and M, utilized for this series of experiments are described in Table I. The time of exposure of the microspheres was 0.3 second for condition A and 0.1 second for conditions K and M. A water-cooled, solenoid actuated shield, located between the plasma nozzle and the microsphere was raised and lowered to control the time of exposure.

A comprehensive description of these experiments, re-entry conditions and theoretical analysis have been documented in References 1 and 2.

Figure 1. $^{238}\text{PuO}_2$ Microsphere M-29 Before
Testing-Diameter 235.4 Microns

Figure 2. Diagram Showing Arrangement of the Hyperthermal Nozzle, the Holder for the SNAP Fuel Particle, and the Collection Plate

BLANK

2.0 SAMPLE DESCRIPTION

The plutonium particles were collected on two types of copper impactor plates (shown in Figure 3). The Type A plates, coated with polyvinylchloride (PVC), were used for all samples described in this report, except impactor M-31, which was used to show the effects of using an uncoated Type B plate. Table I lists the collection plates investigated, with a description of the plutonium dioxide SNAP fuel particles that were ablated under conditions simulating atmospheric re-entry.

The particles were placed individually in a hyperthermal arc tunnel filled with a gas of a composition and pressure appropriate for the simulation of atmospheric re-entry. As shown in Figure 2, an impactor plate was placed downstream of each particle to collect a representative portion of the ablation fragments.

The effect on the surfaces of the collection plates being heated by the plasma in the arc tunnel varied from practically no effect on those subjected to slight heating (Condition A) to severe charring and polymerization of the coatings subjected to extreme heating (Condition M).

Table I. Description of Collection Plates Analyzed and $^{238}\text{PuO}_2$ SNAP Fuel Particles Ablated*					
Impactor Number	Original Particle Diameter (μ)	Particle Weight (μg)	Density (gm/cc)	Plate Coating	Ablation Condition**
M-3	217.9	51.1	9.4	PVC	A
M-15	235.3	77.0	11.3	PVC	K
M-35	222.7	63.3	10.9	PVC	K
M-29	235.4	66.8	9.8	PVC	M
M-37	227.2	65.3	10.6	PVC	M
M-31	217.6	56.4	10.4	None	K
<p>*The density of PuO_2 is 11.46, according to the 48th edition of the <u>Handbook of Chemistry and Physics</u>, published by the Chemical Rubber Company. The values of density listed in this table were calculated from the values determined experimentally for particle diameter and weight. The difference may be due to voids in the particles.</p> <p>**Ablation conditions were as follows:</p> <p>Condition A - Enthalpy, 11,000 BTU/lb.; model pressure, 0.0485 atm.; stagnation heat flux, 650 BTU/ft.²sec.</p> <p>Condition K - Enthalpy, 13,000 BTU/lb.; model pressure, 0.0987 atm.; stagnation heat flux, 1800 BTU/ft.²sec.</p> <p>Condition M - Enthalpy, 12,000 BTU/lb.; model pressure, 0.276 atm.; stagnation heat flux, 2800 BTU/ft.²sec.</p>

Type A

Type B

Figure 3. Special Copper Collection Plates in Lucite Holders for Shipment.
The Diameters are 3-1/2" and 3/4" for Types A and B, respectively.

3.0 DESCRIPTION OF WORK

3.1 GENERAL PROCEDURE FOR PARTICLE ANALYSIS

The following procedure was used to analyze the plutonium particles collected on the impactor plates:

The collection plate is removed from the shipping container, and its mylar covering is tested for alpha contamination. If the test proves negative, a piece of x-ray film is placed in contact with the plate. A 30-minute exposure is usually appropriate. The resulting radioautographs are shown in Appendix A. The impactor is then exposed to Kodak nuclear emulsion slides placed in contact with it. An exposure of one to 15 minutes is selected to record "countable" hollow stars produced by the most radioactive particles on the plates.

The hollow stars, described and illustrated in Appendix B, are characteristic patterns of alpha tracks produced by a point source of alpha activity on a microscope slide that is shimmed with mylar film to a known distance from a nuclear emulsion plate. The hollow stars can be used to locate, as well as to determine the activity of, individual alpha-emitting particles. Alpha activities for $^{238}\text{PuO}_2$ particles of various diameters are presented in Appendix B (Table B-II).*

After processing, all the nuclear emulsion plates are scanned with an optical microscope at a magnification of 150X. The number of alpha tracks in each hollow star is estimated by comparison with a set of 20 standard hollow stars which exhibit from 100 to 20,000 tracks each. From these values, the appropriate shim thicknesses and exposure times, the equivalent $^{238}\text{PuO}_2$ diameters are calculated with a computer program. The diameters are plotted as numerical size-distribution histograms.

The mylar film is removed from the plate, placed in a clean 400-ml beaker that contains 60 ml of methyl ethyl ketone (MEK), scrubbed with a policeman (rubber-tipped glass rod), and removed from the beaker. The collection plate is then placed in the beaker with the same 60-ml portion of solvent and also scrubbed with the policeman. After the plate is removed, it is re-exposed to x-ray film for from one to three hours.

The 60 ml of MEK with the plutonium-dioxide particles removed from the plate is ultra-centrifuged at 8000 rpm for 10 minutes. This is sufficient to collect all PuO_2 particles with diameters $> 0.09 \mu$ in the residue. All but ~ 3 ml of the supernate is removed. The residue is suspended

* In this report, plutonium measurements are calculated as equivalent $^{238}\text{PuO}_2$ spherical diameters or masses. Thus, the small amount of $^{239}\text{PuO}_2$ present with the $^{238}\text{PuO}_2$ is neglected. An equivalent $^{238}\text{PuO}_2$ mass value should be multiplied by 1.25 to obtain the corresponding $^{238,9}\text{Pu}$ value for a given number of alpha disintegrations per minute (dpm). An equivalent $^{238}\text{PuO}_2$ diameter should be multiplied by 1.08 to obtain the corresponding equivalent $^{238,9}\text{PuO}_2$ diameter for a given dpm.

in the 3 ml of solvent by sonic agitation and transferred evenly to ten clean, glass microscope slides. These slides are placed in a Tracerlab LTA-600 Low-Temperature Asher for ~ 20 minutes to remove the PVC which was dissolved in the solvent or charred and polymerized in the arc tunnel.

The ten slides with the PuO_2 particles from each sample are placed in special holders and exposed to nuclear emulsion plates. The exposure times vary from 0.25 to 5 minutes, depending on the time required for the most active particles to produce hollow stars. The hollow stars, formed by alpha tracks, are recorded in the nuclear emulsion plates, which are prevented by shims from contacting the slides with the radioactive particles. The number of tracks in the hollow stars are determined and these values recorded.

All the large radioactive particles thus measured are removed from one of the ten slides from each sample. A particle is isolated by adding 50% collodion to the slide, locating the particle under the optical microscope by its hollow star, and removing it with a sharp-pointed metal rod. Before and after removal, two of these particles are photomicrographed to indicate the general appearance of particles from each sample for this exposure and to see if the particles were broken by manipulation. Isolation also prevents hollow stars produced by large radioactive particles from obscuring those produced by smaller radioactive particles during subsequent exposures.

To verify hollow-star alpha-counting of radioactive particles with equivalent $^{238}\text{PuO}_2$ diameters $> 1 \mu$, these particles were counted electronically after isolation. The alpha-counting system consists of a Nuclear Enterprises plastic scintillator, type NE102, with a 1-1/2" diameter, coupled to a Dumont 6292 photomultiplier tube attached to a Tracerlab Compu/Matic scaler, model SX-4. The background for this system is about 0.6 cpm.

The particles on each slide from which the largest particles have been removed are transferred to four clean slides so that the hollow stars produced during the next exposure period can be separated. These four slides are exposed to nuclear emulsion plates for a period of from 15 to 30 minutes. (See Figure 4). The exact time again depends on the time appropriate to the maximum particle size. Again, all these slides are scanned for hollow stars, the number of alpha tracks in each star counted, and equivalent $^{238}\text{PuO}_2$ diameters determined. From one of these slides, representing 1/40 of the actual sample, all the particles associated with the hollow stars that were just estimated are isolated. As a control, two of the isolated particles are photomicrographed before and after isolation. Examples are shown in Appendix C. The slide is exposed to nuclear emulsion plates for 45 to 60 minutes. The slide is then scanned, the equivalent $^{238}\text{PuO}_2$ diameters are determined, and photomicrographs of two typical particles are prepared. Particles with equivalent $^{238}\text{PuO}_2$ diameters as small as 0.1μ can be determined. The numbers of the particles with the various equivalent $^{238}\text{PuO}_2$ diameters for each exposure are normalized for the sample, and the resulting values are used to plot the numerical size distribution histogram and the mass distribution of the particles. Examples are shown in Appendix D.

For each impactor, the equivalent $^{238}\text{PuO}_2$ mass of particles with equivalent spherical diameters $< \sim 0.1 \mu$ is estimated as follows:*

* Examples of these calculations are presented in Appendix E.

Exposure 1 -- Ten slides exposed
for $\frac{1}{4}$ - 5 minutes

Exposure 2 -- Four slides exposed
for 15 - 30 minutes

Exposure 3 -- Four slides exposed
for 45 - 60 minutes

Figure 4. Diagram illustrating how radioactive particles from a washed impactor are exposed to nuclear emulsion plates. Material on one slide is dispersed on four other slides, which are then exposed to nuclear emulsion plates. This step is repeated so that a total of three exposures is obtained.

- Radiochemical analyses are performed to determine the equivalent $^{238}\text{PuO}_2$ masses of (1) the material transferred as the centrifuged residue and subjected to particle analysis, (2) the supernate from the centrifugation, and (3) the residue on the washed impactor.
- The total equivalent mass of $^{238}\text{PuO}_2$ found by particle analysis is subtracted from the total mass of $^{238}\text{PuO}_2$ on the slides. This difference is the equivalent $^{238}\text{PuO}_2$ mass of particles with equivalent diameters $< \sim 0.1 \mu$ which were on the microscope slides.
- The equivalent $^{238}\text{PuO}_2$ mass found for the supernate, which represents a size range of $0 - 0.09 \mu$ and the equivalent masses found for particles on the slides with size ranges such as $0 - 0.3$ and $0.3 - 0.9 \mu$ are tabulated. The percentages of the total equivalent mass washed from the impactor are calculated for each of these ranges.
- Assuming that the ^{238}Pu remaining on the washed impactor was in the form of particles with the same equivalent $^{238}\text{PuO}_2$ size distribution as those washed from the impactor, the percent mass values for the particles washed from the impactor will be the same as the percent mass values for the particles collected initially on the impactor. (The percentages for the $0 - 0.09 \mu$ range for the supernate and the $0-0.3 \mu$ range for the slides are added.)
- To calculate the equivalent mass of each size range collected on the impactor, the equivalent mass of the particles removed by washing is added to the mass of those which remained on the impactor. The total is multiplied by each of the above mass percentages to obtain the desired values.

3.2 EXCEPTIONS

- During the analysis of the radioactive particles collected on impactor plate M-3, the MEK washing was centrifuged at ~ 2000 rpm for 30 minutes, rather than at 8000 rpm for 10 minutes. This is considered satisfactory for removing particles with equivalent $^{238}\text{PuO}_2$ spherical diameters $> 0.1 \mu$.
- The MEK was replaced by about 50 ml of isopropyl alcohol to remove the particles from impactor M-31, which was uncoated when the radioactive fragments were collected. The alcohol was evaporated to 3 ml, stippled onto 10 clean microscope slides, and then analyzed according to the general procedure.
- A total of six, not three, exposures was made of the particles transferred from impactor M-35 to microscope slides. After the third exposure, the material on one of the slides was dispersed onto four slides. These four slides were exposed a fourth time (one hour) to nuclear emulsion plates. In the same way, fifth and sixth exposures were made for 8 and 72 hours, respectively.
- Radiochemical analyses on material from impactor M-31 were not performed, because this impactor was used to show the effects of not coating the impactor with PVC.

4.0 RESULTS

In the following paragraphs, the results of the analyses of all samples are grouped by analytical phases. This procedure was adopted because of the similarity of results.

4.1 EXPOSURES TO X-RAY FILM

Photographs of the x-ray films that were exposed to the collection plates before and after the plates were washed are shown in Appendix A (Figures A-1 through A-6). The difficulty of removing the PVC from the collection plates, directly related to the amount of heating to which the plates were subjected during the tests, is illustrated by Figures A-4 and A-6, which show that much of the radioactivity on plates M-29 and M-37 was not removed by washing. That PVC is useful in removing at least part of the plutonium for analysis is shown by Figure A-6, which reveals that even less plutonium was removed from the uncoated impactor M-31.

4.2 SIZE DISTRIBUTIONS OF $^{238}\text{PuO}_2$ PARTICLES GREATER THAN $0.2\ \mu$ IN DIAMETER

The equivalent $^{238}\text{PuO}_2$ diameters obtained by the hollow-star technique and by electronic counting were used to plot the size distribution of each sample.

The numerical size distribution histograms of the radioactive particles found on each collection plate before and after washing are shown in Appendix D (Figures D-1 through D-10). No histograms are presented for radioactive particles detected on plates M-15 and M-31 before washing, because, even though exposures to nuclear emulsion slides for one and 15 minutes were made, only one hollow star was associated with each plate; these two hollow stars indicated equivalent $^{238}\text{PuO}_2$ diameters of $0.7\ \mu$ and $0.9\ \mu$, respectively.

Comparison of the numbers of particles in the four largest size ranges on the impactors before washing (Figures D-1 through D-4) with their corresponding values determined after washing (Figures D-5, D-7, D-8, and D-9) reveals that the two sets of values are essentially identical. Hence, the larger particles on each impactor were probably not broken in the washing. Further, no agglomeration of these particles occurred during their removal from the impactor.

Three categories of particles were not used in plotting the histograms: (1) those so small that they were not recorded, even by the longest exposure; (2) particles which remained on the plates after washing; and (3) particles which fell to the bottom of the arc tunnel before reaching the plates. Typical radioactive particles are described in Appendix C. Appendix F illustrates the calculation of standard deviations of alpha activities for particles after isolation and percentages of initial alpha activities recovered.

4.3 RADIOCHEMICAL ANALYSES*

Results of the radiochemical determination of the total amounts of PuO_2 collected on impactors M-3, M-35, and M-29, which were subjected to ablation conditions A, K, and M, respectively, are presented in Table II. These totals are compared with the masses of the respective microspheres before ablation (see Table I) to calculate the percentages of $^{238}\text{PuO}_2$ collected on each impactor, and the percentages are also listed in Table II. Photomicrograph movies of the microspheres were

* The procedure for radiochemical analysis is described in Appendix G.

taken during the experiment. The masses of the microspheres removed by ablation during each run were estimated from the 100X projections of the frames that showed the particles before and after ablation. These estimates were $2 \times 10^{-1} \mu\text{g}$ for M-3, $4 \mu\text{g}$ for M-15, $5.6 \mu\text{g}$ for M-35, $26 \mu\text{g}$ for M-29, and $32 \mu\text{g}$ for M-37. On the basis of these estimated values, the mass percentages of the ablated PuO_2 that was collected on each impactor were calculated and are listed in Table II.

As described in Section 3.0, the results of radiochemical analysis were used with the data from individual particle analysis to calculate the equivalent $^{238}\text{PuO}_2$ mass distribution for diameters from zero to the largest collected on each impactor. The mass distributions for each impactor were calculated as shown in Appendix E. The results shown in Tables III and VII indicate that at least 95% of the mass of PuO_2 collected is in particles with equivalent $^{238}\text{PuO}_2$ diameters $< \sim 0.1 \mu$. Numerically, most of these are Type I particles. It appears that each of these particles is formed by many very small PuO_2 particles encapsulated in a plastic matrix. Thus these Type I particles may have equivalent diameters much larger than those of particles obtained under actual condensation conditions. Therefore, the values (Tables III through VII) should be considered as upper limits for the ranges $> \sim 1.0 \mu$.

Table II. Results of Radiochemical Analyses Performed to Determine the Equivalent $^{238}\text{PuO}_2$ Masses Collected on Impactors					
Portion Analyzed	M-3 (μg)	M-15 (μg)	M-35 (μg)	M-29 (μg)	M-37 (μg)
Centrifuged Residue	2.9×10^{-2}	2.0×10^{-1}	2.9×10^{-1}	3.5×10^{-1}	2.3×10^{-1}
Washed Impactor	4.0×10^{-3}	5.6×10^{-1}	3.0×10^{-2}	3.5×10^{-1}	2.5×10^{-1}
Supernate	3.8×10^{-2}	4.1×10^{-2}	3.0×10^{-2}	1.0	3.2×10^{-2}
Total Collected	7.1×10^{-2}	8.0×10^{-1}	3.5×10^{-1}	1.7	5.1×10^{-1}
Fraction of Initial Particle Collected	0.14%	1%	0.55%	2.6%	0.8%
Portion of Ablated Mass Collected	35%	20%	6.2%	6.5%	1.6%

Table III Percentages of Total Mass for Two Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-3*			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0. - 0.3	6.99×10^{-2}	99.9	99.9
0.3 - 0.9	1.0×10^{-4}	1.4×10^{-1}	100

Table IV. Percentages of Total Equivalent Mass for Two Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-15*			
Size Ranges of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0. - 0.2	8.0×10^{-1}	99.5	99.5
0.2 - 1.4	4.0×10^{-3}	5.0×10^{-1}	100

Table V. Percentages of Total Mass for Three Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-35*			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0. - 0.05	3.4×10^{-1}	95.8	95.8
0.05 - 0.10	1.2×10^{-2}	3.39	99.2
0.10 - 1.6	2.7×10^{-3}	7.6×10^{-1}	100

Table VI. Percentages of Total Mass for Four Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-29*			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0. - 0.2	1.62	95.1	95.1
0.2 - 0.6	1.02×10^{-3}	6×10^{-2}	95.2
0.6 - 1.0	1.7×10^{-3}	1×10^{-1}	95.3
1.0 - 14.0	8.16×10^{-2}	4.8	100

Table VII. Percentages of Total Equivalent Mass for Four Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-37*			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0. - 0.2	4.8×10^{-1}	92.9	92.9
0.2 - 0.6	2.9×10^{-3}	5.7×10^{-1}	93.5
0.6 - 1.0	1.6×10^{-3}	3.1×10^{-1}	93.8
1.0 - 8.0	3.2×10^{-2}	6.18	100

*As shown by the above tables, the cutoff for particles measured by the hollow-star technique varied from 0.1 to 0.2 μ . Since it was desired to have a limit of three hollow-star exposures per collection, the last exposures were for varying periods, depending on the sizes and numbers of larger particles present.

As shown in Table I, impactors M-15 and M-35 were exposed under condition K, and impactors M-29 and M-37 under condition M. Comparing the values in Table IV for M-15 with those in Table V for M-35 and the values in Table VI for M-29 with those in Table VII for M-37 reveals a striking agreement and indicates unusual precision for sets of experimental results. Similar agreement of tabulated mass distributions was reported for the particles collected on impactors M-22 and M-32,³ which were exposed under ablation condition L.*

Thus it was established that reproducible mass distributions of plutonium particles were collected on the impactors after runs under three different ablation conditions. However, it cannot be established whether a mass distribution of particles collected on an impactor is characteristic of the mass distribution of the particles produced by ablation (or, in other words, whether the mass distribution of the particles not collected on the impactors is the same as the mass distribution of the particles which were collected on the impactors). This can be determined only by developing a method to collect all particles produced by ablation during the experiments.

A mass balance of the material ablated from the $^{238}\text{PuO}_2$ microspheres was conducted by Sandia. Where possible, the 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 35 percent of the mass ablated from the microsphere was actually collected on the sampler. A collection efficiency analysis of the samplers was performed by Sandia to determine the size range of the particles which could be expected to be collected. This analysis included flow patterns, effect of shock waves, and sampler configuration. In general, the analysis indicated that particles between 0.01 and 10 μ in diameter would be collected with approximately 100 percent efficiency. Particles less than 0.01 μ in diameter would be trapped in the flow and diverted around the sampler. Unless it was originally projected toward the sampler, particles greater than 10 μ in diameter would not be affected by the flow and also would miss the sampler. The results of the collection efficiency analysis indicate that the size distribution of the debris collected is not exactly representative of the ablated size distribution in all cases. Therefore, it was necessary to modify the size distributions given in Tables III - VII 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 sample M-3 was 0.9 μ in diameter. Since the collection efficiency of the sampler was approximately 100 percent up to 10 μ , it is safe to assume that the entire ablated inventory was less than 0.9 μ in diameter. The collected and uncollected (due to the holes in the impactors) mass from 0.01 to 0.9 μ represents 52.20 percent of the total ablated mass. The uncollected remainder must then be less than 0.01 μ in diameter and represents 47.73 percent of the ablated mass. This size distribution does not differ appreciably from that shown in Table III.

* Condition L -- enthalpy, 3600 BTU/lb.; model pressure, 0.0237 atm.; stagnation heat flux, 100 BTU ft² sec.

The analysis of sample M-35 (Condition K) resulted in conclusions similar to those reached for the Condition A test. Since the largest particle collected was 1.6μ in diameter, the entire ablated inventory was less than 1.6μ . The collected and uncollected mass from 0.01 to 1.6μ represented 9.29 percent of the total ablated mass. The remaining 90.71 percent was contained in particles less than 0.01μ in diameter. This size distribution is also not much different from the values given in Tables IV and V.

The analysis of sample M-29 (Condition M) resulted in a significant change from the size distribution shown in Tables VI and VII. 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 100μ 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 and uncollected mass between 0.01 and 14μ in diameter represented 6.94 percent of the total ablated mass (which included the melted runback material). It was estimated that the runback material accounts for almost all of the remaining 93.06 percent. Therefore, the percent of ablated debris less than 0.01μ in diameter was very small.

Table VIII shows the final size distribution of the ablated debris, based on the Tracerlab size distribution analysis and modified by the Sandia 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. The final fate and debris size of this runback material in an actual reentry cannot be determined in the arc tunnel facility. However, it is apparent that the debris size distribution is dependent on the heat input and melt rate of the microsphere.

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

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

5.0 CONCLUSIONS

- Radiochemical determinations of inventories of particles collected on impactors subjected to ablation conditions A, K, or M were used with individual particle measurements determined by the hollow-star and electronic-counting techniques to tabulate the percentage of the total PuO_2 mass in several size ranges from zero to the largest found on each of these impactors. In each instance, approximately 95% of the mass of PuO_2 collected on the impactor was in particles with equivalent $^{238}\text{PuO}_2$ diameters $< \sim 0.1 \mu$.
- From two of the six collection plates analyzed (M-29 and M-37), particles composed mainly of $^{238}\text{PuO}_2$ were obtained which had equivalent $^{238}\text{PuO}_2$ diameters from 1.3 to 13.5 μ , as determined by the hollow-star and electronic-counting techniques.
- The radioactive particles from M-3, M-15, and M-35 were found, by the hollow-star technique, to have equivalent $^{238}\text{PuO}_2$ diameters ranging from 0.1 to 1.6 μ . These particles have a transparent matrix believed to be mainly charred or polymerized PVC, collodion, and/or silicon. The actual sizes of these particles are much larger than their equivalent $^{238}\text{PuO}_2$ diameters. Electron microprobe analysis revealed that these particles contain silicon, iron, copper, and chromium.
- Except for a few particles of almost pure $^{238}\text{PuO}_2$, all the radioactive particles from M-29 and M-37 were composed of the transparent material.
- The radioactive particles washed from the uncoated copper plate M-31 were composed of a transparent matrix with opaque spots. The actual sizes of these particles were at least one order of magnitude larger than the equivalent $^{238}\text{PuO}_2$ diameters.
- Coating the collection plates with a material such as PVC permits a more complete removal of the radioactive particles collected, as shown by comparing the autoradiographs of the coated and uncoated plates after washing.

BLANK

6.0 RECOMMENDATIONS

A system should be developed to collect all the particles produced from a SNAP particle ablated in the arc tunnel. (Table II shows that a maximum of 35% of the ablated mass of each microsphere was collected.)

A better coating than PVC for the collection surface of the impactors is desirable.

REFERENCES

1. SC-TM-68-15, "Experiments on Small Particle Ablation in Simulated Reentry-Interim Report," B. W. Marshall and I. B. White, April 1968.
2. SC-RR-68-547, "Thermal and Ablation Response of Small Spherical Particles in Reentry-Summary Report," B. W. Marshall, October 1968.
3. SC-CR-68-3673, "Radiochemical Analyses of $^{238}\text{PuO}_2$ Collected on Impactors M-15, M-37, and M-32," W. Holland, Tracerlab for Sandia Corporation, June 21, 1968.

APPENDIX A

RADIOAUTOGRAPHS OF IMPACTORS BEFORE AND AFTER WASHING

M-3 before washing
30-minute exposure

M-3 after washing
2-hour exposure

Figure A-1. X-ray Film Exposures of M-3 Collection Plate Before and After Washing.

M-15 before washing
30-minute exposure

M-15 after washing
1-hour exposure

Figure A-2. X-ray Film Exposures of M-15 Collection Plate
Before and After Washing

M-35 before washing
30-minute exposure

M-35 after washing
1-hour exposure

Figure A-3. X-ray film exposures of M-35 Collection Plate
Before and After Washing

M-29 before washing
30-minute exposure

M-29 after washing
3-hour exposure

Figure A-4. X-ray Film Exposures of M-29 Collection Plate
Before and After Washing

M-37 before washing
30-minute exposure

M-37 after washing
3-hour exposure

Figure A-5. X-ray Film Exposures of M-37 Collection Plate
Before and After Washing

M-31 before washing
30-minute exposure

M-31 after washing
1-hour exposure

Figure A-6. X-ray Film Exposures of M-31 Collection Plate
Before and After Washing

APPENDIX B

HOLLOW-STAR TECHNIQUE (B 1)

The hollow-star technique is used to measure the alpha activity to permit calculation of the equivalent $^{238}\text{PuO}_2$ diameters of particles. The exposure arrangement uses shims to maintain a known distance between the particles and the slide of thick-nuclear emulsion.

Figures B-1 and B-2 show how this arrangement is used to make the desired calculations. The shim distance AA' and the exposure time t (in minutes) are known. By optical microscopy, the center A' of the hollow star is located. Also, an alpha track such as BE, at a convenient distance from the center (about 400 μ), is selected, and the distance A'B is measured with a filar microscope. By counting the single alpha tracks in the circle of radius A'B (including those intersecting this circle), N is determined. A photomicrograph of a hollow star produced by this arrangement is shown in Figure B-3. Table B-II presents the alpha activities for $^{238}\text{PuO}_2$ particles of various diameters.

To calculate the solid angle Ω in which these tracks were produced, the following relationship is used (see Figure B-1):

$$\tan \theta = \frac{A'B}{A'A} \quad \theta = \tan^{-1} \frac{A'B}{A'A}$$

The solid angle Ω in which N tracks are produced is given by:

$$\Omega = \int_0^\theta 2\pi \sin \theta d\theta = 2\pi(1 - \cos \theta)$$

The number of alpha disintegrations D_t from the particle during time (t) is calculated by:

$$D_t = \frac{4\pi}{\Omega} \times N = \frac{2N}{1 - \cos \theta}$$

The alpha activity D_m (in dpm) of the particle is calculated by:

$$D_m = \frac{D_t}{t}$$

Likewise, the equivalent diameter d(in μ) of a $^{238}\text{PuO}_2$ particle is calculated by:

$$d = \left(\frac{10^{-2} D_t}{2t} \right)^{1/3}$$

Table B-1 shows the results of using this method of calculation for a SNAP-9A abort particle of unusually high activity,* which was isolated from a high altitude filter collection. That there were no large experimental errors in these results is shown by comparing the 2.9% standard error for the average dpm based on Poisson statistics for the actual tracks counted with the 5.1% normal standard error for this average.

(B 1) Third Quarterly Report of SNAP-9A Particle Studies During 15 April 1966 - 15 July 1966, Tracerlab, A Division of Laboratory for Electronics, HASL Contract No. AT(30-1)-3547 Mod. 3, September 14, 1966.

* A computer program was used to obtain these data.

Figure B-1. Side View of the Geometric Configuration Used for Calculation of Total Alpha Disintegrations from a Particle Being Exposed by the Hollow-Star Technique

Figure B-2. Top View of the Geometric Configuration for Calculation of the Total Alpha Disintegrations from a Particle Being Exposed by the Hollow-Star Technique

Figure B-3. Photomicrograph of a Hollow Star Produced by Alpha Radiation from Particle A-2-C-3 of Sample M-3. The Particle was Exposed to the Nuclear Emulsion Plate for 2 Minutes. The Shim Thickness was 76μ . Approximately 400 Alpha Tracks are Shown in this Photomicrograph

Table B-I. Results of Hollow-Star Exposures of a SNAP-9A Particle

Exposure No.	Exposure Time (hrs.)	Shim Thickness AA' (μ)	A'B (μ)	θ^* ($^\circ$)	Actual Track Count in Circle Radius A'B**	Alpha Activity (dpm)**	Particle Diameter (μ)	Particle Volume ($10^{-5} \mu$)	Particle Mass (10^{-17} gm)
3	91	76	351	77.8	$65 \pm 12\%$	$0.030 \pm 12\%$	0.053	7.80	89.4
4	96	76	439	80.1	$63 \pm 13\%$	$0.026 \pm 13\%$	0.051	6.95	79.6
5	77	76	439	80.1	$52 \pm 14\%$	$0.027 \pm 14\%$	0.051	6.95	79.6
6	96	76	439	80.1	$73 \pm 12\%$	$0.031 \pm 12\%$	0.054	8.25	94.6
7	72	76	439	80.1	$45 \pm 15\%$	$0.025 \pm 15\%$	0.050	6.55	75.1
8	91	127	439	73.8	$55 \pm 14\%$	$0.028 \pm 14\%$	0.052	7.36	84.4
9	73	127	548	76.9	$46 \pm 15\%$	$0.027 \pm 15\%$	0.051	6.95	79.6
10	73	127	548	76.9	$46 \pm 15\%$	$0.027 \pm 15\%$	0.051	6.94	79.5
11	145	483	548	48.6	$53 \pm 14\%$	$0.036 \pm 14\%$	0.056	9.20	105.4
12	168	483	548	48.6	$65 \pm 12\%$	$0.038 \pm 12\%$	0.057	9.70	111.2
13	170	483	548	48.6	$72 \pm 12\%$	$0.042 \pm 12\%$	0.059	10.80	123.8
14	170.5	483	548	48.6	$48 \pm 14\%$	$0.028 \pm 14\%$	0.052	7.36	84.3
15	96	51	548	84.6	$69 \pm 12\%$	$0.029 \pm 12\%$	0.051	6.94	79.5
Average						$0.030 \pm 2.9\%$	0.053		

* θ is the window angle A'AB

** The variations are standard deviations based on the Poisson statistics of the actual track counts. The standard error for the average alpha activity is listed at the bottom of the column.

Table B-II Alpha Activities for $^{238}\text{PuO}_2$ Particles of Various Diameters*

Diameter (μ)	DPM
100	2.0×10^8
50	2.5×10^7
20	1.6×10^6
10	2.0×10^5
5	2.5×10^4
2	1.6×10^3
1	200
0.5	25
0.2	1.6
0.1	0.2

*These values are not corrected for self-absorption, which is significant for particles $> 15 \mu$ in diameter.

Figure B-4 shows the special holder prepared for the microscope slides during the hollow-star exposure.

Figure B-4. Photographs of Special Holder Used for Securing Glass Microscope Slides During Exposure for the Hollow-Star Technique. The Holder is Shown About One-Half Actual Size

APPENDIX C

TYPICAL $^{238}\text{PuO}_2$ PARTICLES

To obtain a general idea of the appearance of the radioactive particles collected on each plate, photomicrographs were taken of typical radioactive particles before and after isolation (Figures C-1 through C-44). In a few cases, the photomicrographs of radioactive particles before isolation only remotely resemble those taken after isolation. This is attributed to both breakage and changes in particle orientation.

As shown in Figures C-1 through C-18 and C-35 through C-45, the plutonium is associated with a transparent matrix, but, in some cases, of pure $^{238}\text{PuO}_2$. The latter category of particles (Type II) is illustrated in Figures C-20, C-21, C-22, C-23, C-26, C-27, C-29, and C-30. The pure $^{238}\text{PuO}_2$ particles have diameters that vary from $1.3\ \mu$ to $13.5\ \mu$. In addition, another type of particle (Type III) was found that combined the characteristics of the other two (Figures C-19 and C-28). These particles appear to consist of fragments of plutonium dioxide attached to clear matrices.

Several particles representative of each of the three categories were analyzed with an electron microprobe by personnel at Los Alamos. Particle A6-11 from impactor M-29 (Figure C-21) was found to consist of two phases: its perimeter containing iron, chromium, nickel, and possibly manganese, and its center consisting of a heterogeneous mixture of plutonium, chromium, silicon, calcium, and manganese with some iron, nickel, and copper. This analysis is reported in Appendix C. This analysis also contains the reports of the electron microprobe analyses on eleven other radioactive particles as well as oscillographs obtained by backscattered electrons. The size and shape of each radioactive particle as shown in these oscillographs agrees well with that shown in the corresponding photomicrograph.

The particles were prepared for analysis by attaching them with 50% collodion to bakelite discs 1" in diameter and $1/4$ " thick. A circle about 1 mm in diameter and two scratches, forming an "eye" about $100\ \mu$ wide, are scribed around each particle, and three photomicrographs at magnifications of 100X, 200X, and 500X for each particle were prepared to enable the electron microprobe analyst to locate the particles of interest (see Figures C-4 and C-45).

The results of electron microprobe analysis are summarized in Table C-I. The Type I particles appear to be predominantly silicon, with some iron, and/or copper with traces of chromium or iron. (A high concentration of polymerized PVC must be assumed because of the relatively small concentration of silicon and other elements found in particles A9-C8 from M-15 and A7-6 from M-31.) Even though the equivalent $^{238}\text{PuO}_2$ diameter for each of these particles was found by the hollow-star technique to be about $0.5\ \mu$, the electron microprobe analyzer did not detect plutonium in these particles. Either the quantity of plutonium was too small to detect, or the particles were embedded at depths greater than $2\ \mu$, so that the plutonium X-rays were weakened by the intervening material.

The Type II particles are mainly plutonium with other major elements, such as iron, chromium, nickel, manganese, silicon, calcium, and copper.

The Type III particles appear to be mainly plutonium, but are attached to a matrix of elements lighter than sodium. This matrix is most probably polymerized PVC or collodion.

The high concentrations of such elements as iron, chromium, and nickel in the Type II particles indicates volatilization of significant portions of these elements in the Sandia arc tunnel. The specifications for $^{238}\text{PuO}_2$ microspheres permit no more than 2% impurities by weight.^{C1} Electron microprobe analysis of plutonium dioxide particles formed by SNAP microspheres subjected to conditions simulating intact reentry indicated extremely low concentrations of these impurities.^{C2} Thus it is assumed that the impurities found in the Type II particles are added when the particles are formed in the tunnel. Although these contaminants could be eliminated, their concentrations are not great enough to affect the sizes of the Type II particles.

Except for particle A6-11, which was undergoing electron microprobe analysis, all particles with equivalent $^{238}\text{PuO}_2$ diameters greater than $1.0\ \mu$, as determined by the hollow-star technique, were verified by electronic counting. All results of electronic counting agreed within 10% of the hollow-star values, except for particles which differed by 14% (A6-6 from M-29), 26% (A-2-3 from M-29), and 42% (A2-7 from M-37).

The equivalent $^{238}\text{PuO}_2$ diameter for particle A6-11 from M-29, as determined by the hollow-star technique, was used in plotting the size distribution histogram for M-29. Self-absorption correction factors were not applied to these results because they are less than 5% for $^{238}\text{PuO}_2$ particles with actual diameters smaller than $15\ \mu$.^{C3} It is planned to apply the self-absorption correction factor to the results obtained by electronically counting A6-11 from M-29.

For A2-3 from M-29 and A2-7 and A2-C3 from M-37, the electronic counting results were used in preference to the hollow-star results. For particles with equivalent $^{238}\text{PuO}_2$ diameters greater than $5\ \mu$ (an activity greater than 2.5×10^4 dpm), the electronic counting technique is considered more accurate because of the difficulty of obtaining accurate exposures of less than one minute for the hollow-star technique. Thus the electronic-counting results for these three particles were used in the plots of the results from M-29 and M-37.

Tables C-2 through C-7 compare the ^{238}Pu activities for particles before and after isolation for each sample. The method used to calculate the standard deviations of the alpha activities of particles after isolation and the percentages of initial alpha activities recovered is described in Appendix F. Generally, the radioactive particles were isolated intact and without loss of alpha activity.

It is assumed that during the formation and analysis of the three types of radioactive particles the plutonium dioxide fragments of the SNAP fuel particles were agglomerated rather than fractured.

Notes to Appendix C:

C. F. Metz, Group Leader CMB-1, is with Los Alamos Scientific Laboratory,
Los Alamos, New Mexico.

- C1 Plutonium-238 and Polonium-210 Data Sheets, Mound Laboratory for Monsanto Research Corporation, Report No. MLM-144, page 27, November 9, 1967.
- C2 TLW-6004, "Particle Analysis of Radioactive Fragments with Diameters $< 10 \mu$ from Impacted Samples 18 and 20," W. Holland, Tracerlab for Mound Laboratory, May 28, 1968.
- C3 "Special Particulate Characteristics," C. C. Dunn and R. Bersin, Operation Rollercoaster, FTR/WT Project 2.6C, Tracerlab, A Division of Laboratory for Electronics, November 4, 1964.
- For the July 28, 1967, report located on page 86, the specimen number is A6-11.
- For the December 7, 1967, report located on page 87, the first specimen number should be M-3-A5-D-1.

Table C-I. Summary of Results of Electron Microprobe Analyses					
Particle Designation		Type	Predominant Element	Other Major Elements	Trace Elements
M-29	A6-11	II	Pu	Fe, Cr, Ni, Mn, Si, Ca, Cu	--
M-3	A5-D1	I	Si	Fe	--
M-15	A9-C8	I	Si	Fe, Cu	Cr
M-31	A7-6	I	Si	Cu	Fe
M-29	A2-3	III	Pu	Si	Fe, Cr
M-29	A6-B3	II	Pu	Si, Cr	Fe, Cu, Mn
M-29	A6-6	II	Pu	Si, Cr	Fe, Mn
M-29	A6-B6	II	Pu	--	Si, Fe
M-37	A2-7	III	Pu	--	Fe
M-37	A2-1	II	Pu	Si, Cr	Fe, Mn, Cu
M-37	A2-3	II	Pu	Si, Cr, Fe, Mn	--
M-37	A2-C3	II	Pu	Cr, Fe	Si

Table C-II. Comparison of ^{238}Pu Activities for Particles before and after Isolation from Plate M-3						
Particle Designation	Exposure Time (min.)	Initial Equivalent Diameter (μ)	Pu Activities (dpm)		Number of Radioactive Fragments after Isolation	% of ^{238}Pu Recovered after Isolation
			Before Isolation	After Isolation		
A5-5	5	0.55	$33.9 \pm 12\%$	$31.3 \pm 5\%$	1	92 ± 6
A5-C1	30	0.41	$13.9 \pm 8\%$	$13.3 \pm 8\%$	2	96 ± 11
A5-C2	30	0.39	$12.1 \pm 8\%$	$12.1 \pm 8\%$	1	100 ± 11
A5-C3	30	0.54	$31.3 \pm 5\%$	$29.6 \pm 5\%$	1	95 ± 5
A5-D1	60	0.25	$3.1 \pm 12\%$	$0.7 \pm 25\%$	1	23 ± 8
A5-D5	60	0.28	$4.4 \pm 10\%$	$4.3 \pm 10\%$	1	98 ± 14

Table C-III. Comparison of ^{238}Pu Activities for Particles before and after Isolation from Plate M-15						
Particle Designation	Exposure Time (min.)	Initial Equivalent Diameter (μ)	Pu Activities (dpm)		Number of Radioactive Fragments after Isolation	% of ^{238}Pu Recovered after Isolation
			Before Isolation	After Isolation		
A9-1	5	0.86	$125 \pm 6\%$	$75 \pm 4\%$	8	60 ± 4
A9-2	5	0.72	$73.1 \pm 8\%$	$28.3 \pm 6\%$	7	39 ± 4
A9-C2	30	0.43	$15.8 \pm 7\%$	$21.2 \pm 6\%$	1	134 ± 12
A9-C7	30	0.46	$19.8 \pm 6\%$	$22.2 \pm 6\%$	1	112 ± 9
A9-C8	30	0.46	$19.8 \pm 6\%$	$31.7 \pm 5\%$	1	160 ± 13
A9-C9	30	0.58	$39.6 \pm 4\%$	$42.0 \pm 4\%$	1	106 ± 6
A9-C10	30	0.39	$11.8 \pm 8\%$	$5.9 \pm 12\%$	1	50 ± 7
A9-C61	60	0.34	$7.9 \pm 7\%$	$9.75 \pm 6\%$	1	123 ± 11
A9-C62	60	0.41	$13.8 \pm 5\%$	$14.0 \pm 5\%$	1	101 ± 7

Table C-IV. Comparison of ^{238}Pu Activities for Particles before and after Isolation from Plate M-35						
Particle Designation	Exposure Time (min.)	Initial Equivalent Diameter (μ)	Pu Activities (dpm)		Number of Radioactive Fragments after Isolation	% of ^{238}Pu Recovered after Isolation
			Before Isolation	After Isolation		
A7-1	5	0.94	$170 \pm 5\%$	$190 \pm 4\%$	1	112 ± 7
A7-2	5	0.84	$120 \pm 6\%$	$33 \pm 8\%$	1	28 ± 3
A7-3	5	0.72	$76 \pm 8\%$	$96 \pm 5\%$	1	126 ± 12
A7-B3	30	0.43	$16 \pm 7\%$	$3.9 \pm 14\%$	1	24 ± 4
A7-B5	30	0.40	$13 \pm 8\%$	$5.9 \pm 12\%$	1	45 ± 6
A7-B14	30	0.49	$24 \pm 6\%$	$6.7 \pm 11\%$	4	28 ± 11
A7-BB54	60	0.40	$13 \pm 6\%$	$11 \pm 6\%$	1	85 ± 7
C7-C-38	120	0.28	$4.4 \pm 7\%$	$1.1 \pm 21\%$	2	25 ± 6
C7-CB-84	480	0.18	$1.2 \pm 6\%$	$1.8 \pm 5\%$	1	150 ± 12
C7-CE-48	4320	0.09	$0.14 \pm 6\%$	$0.14 \pm 6\%$	1	100 ± 8

Table C-V. Comparison of ^{238}Pu Activities for Particles before and after Isolation from Plate M-29						
Particle Designation	Exposure Time (min.)	Initial Equivalent Diameter (μ)	^{238}Pu Activities (dpm)		Number of Radioactive Fragments after Isolation	% of ^{238}Pu Recovered after Isolation
			Before Isolation	After Isolation		
A2-1	$\frac{1}{4}$	7.8	$9.5 \times 10^4 \pm 1\%$	$11.4 \times 10^4 \pm 3\%$	1	120 ± 4
A2-3	$\frac{1}{4}$	10.4	*	$2.9 \times 10^5 \pm 2\%$	1	--
A6-6	$\frac{1}{4}$	2.7	$3.8 \times 10^3 \pm 5\%$	$4.5 \times 10^3 \pm 5\%$	1	118 ± 8
A6-8	$\frac{1}{4}$	2.6	$3.4 \times 10^3 \pm 5\%$	$4.8 \times 10^3 \pm 20\%$	2	139 ± 30
A6-11	$\frac{1}{4}$	7.8	$9.5 \times 10^4 \pm 1\%$	$22.8 \times 10^4 \pm 2\%$	1	230 ± 5
A6-12	$\frac{1}{4}$	1.9	$1.3 \times 10^3 \pm 8\%$	$1.5 \times 10^3 \pm 10\%$	1	114 ± 15
A6-B3	2	1.3	$4.8 \times 10^2 \pm 5\%$	$5.0 \times 10^2 \pm 3\%$	1	106 ± 6
A6-B6	2	1.7	$9.5 \times 10^2 \pm 4\%$	$9.5 \times 10^2 \pm 2\%$	1	100 ± 4
A6-B9	2	1.8	$1.2 \times 10^3 \pm 3\%$	$1.4 \times 10^3 \pm 4\%$	1	117 ± 6
A6-B10	2	0.89	$1.4 \times 10^2 \pm 9\%$	$1.2 \times 10^2 \pm 6\%$	1	87 ± 9
A6-BB2	30	0.93	$1.6 \times 10^2 \pm 2\%$	$2.7 \times 10^2 \pm 9\%$	1	160 ± 15
*The activity of the particle before isolation was not considered because it varied too greatly from the count determined electronically after isolation.						

Table C-VI. Comparison of ^{238}Pu Activities for Particles before and after Isolation from Plate M-37						
Particle Designation	Exposure Time (min.)	Initial Equivalent Diameter (μ)	^{238}Pu Activities (dpm)		Number of Radioactive Fragments after Isolation	% of ^{238}Pu Recovered after Isolation
			Before Isolation	After Isolation		
A-2-1	$\frac{1}{4}$	2.8	$4.56 \times 10^3 \pm 5\%$	$4.53 \times 10^3 \pm 5\%$	1	99 ± 7
A-2-3	$\frac{1}{4}$	6.2	$4.75 \times 10^4 \pm 1\%$	$7.61 \times 10^4 \pm 1\%$	1	160 ± 2
A-2-7	$\frac{1}{4}$	7.2	*	$6.6 \times 10^4 \pm 1\%$	1	--
A2-C3	2	1.2	$4.12 \times 10^2 \pm 5\%$	$4.12 \times 10^2 \pm 5\%$	1	100 ± 7
A2-C4	2	1.3	$4.28 \times 10^2 \pm 5\%$	$4.56 \times 10^2 \pm 5\%$	1	107 ± 8
A2-C9	2	1.4	$5.47 \times 10^2 \pm 5\%$	$3.95 \times 10^2 \pm 5\%$	1	72 ± 5
A2-C11	2	0.9	$1.42 \times 10^2 \pm 9\%$	$1.33 \times 10^2 \pm 9\%$	1	94 ± 12
A2-C12	2	1.3	$4.75 \times 10^2 \pm 5\%$	$5.28 \times 10^2 \pm 5\%$	1	111 ± 8
A2-CC-2	30	0.36	$9.51 \pm 9\%$	$11.8 \pm 7\%$	1	124 ± 14
A2-CC-3	30	0.43	$15.8 \pm 7\%$	$11.3 \pm 7\%$	1	72 ± 7
A2-CC-4	30	0.40	$12.6 \pm 8\%$	$14.2 \pm 5\%$	1	113 ± 10
A2-CC-6	30	0.78	$95.1 \pm 2\%$	$73.7 \pm 3\%$	1	78 ± 3
*The activity of the particle before isolation was not considered because it varied too greatly from the count determined electronically after isolation.						

Particle Designation	Exposure Time (min.)	Initial Equivalent Diameter (μ)	^{238}Pu Activities (dpm)		Number of Radioactive Fragments after Isolation	% of ^{238}Pu Recovered after Isolation
			Before Isolation	After Isolation		
A7-1	10	0.30	$5.2 \pm 21\%$	$4.8 \pm 16\%$	1	92 ± 24
A7-2	10	0.55	$33.3 \pm 8\%$	$28.5 \pm 6\%$	1	86 ± 9
A7-3	10	0.36	$9.5 \pm 16\%$	$9.5 \pm 11\%$	1	100 ± 19
A7-4	10	0.37	$10.4 \pm 15\%$	$9.0 \pm 11\%$	1	87 ± 16
A7-5	10	0.48	$28.5 \pm 9\%$	$29.9 \pm 6\%$	1	105 ± 11
A7-6	10	0.38	$11.4 \pm 14\%$	$30.3 \pm 6\%$	2	80 ± 8
A7-7	10	0.48	$21.8 \pm 10\%$	$20.9 \pm 8\%$	1	96 ± 12
A7-8	10	0.38	$11.4 \pm 14\%$	$9.1 \pm 12\%$	2	79 ± 15
A7-9	10	0.46	$19.0 \pm 11\%$	$12.3 \pm 10\%$	1	65 ± 14
A7-10	10	0.39	$11.4 \pm 14\%$	$10.7 \pm 10\%$	1	94 ± 16
A7-11	10	0.36	$9.5 \pm 16\%$	$9.5 \pm 11\%$	1	100 ± 19
A7-B1	30	0.21	$1.9 \pm 20\%$	$0.4 \pm 32\%$	1	21 ± 8
A7-B2	30	0.27	$3.8 \pm 14\%$	$1.6 \pm 16\%$	1	42 ± 9
A7-B3	30	0.22	$3.3 \pm 19\%$	$0.2 \pm 41\%$	1	9 ± 4
A7-B4	30	0.22	$2.2 \pm 19\%$	$0.8 \pm 22\%$	1	38 ± 4
A7-B5	30	0.24	$2.8 \pm 17\%$	$1.3 \pm 17\%$	1	46 ± 11
A7-B6	30	0.26	$3.5 \pm 15\%$	$2.3 \pm 13\%$	1	66 ± 13
A7-B9	30	0.21	$1.9 \pm 20\%$	$1.9 \pm 14\%$	1	100 ± 24
A7-B10	30	0.26	$3.5 \pm 15\%$	$2.2 \pm 13\%$	1	63 ± 13
A7-BB1	60	0.16	$0.8 \pm 22\%$	$0.7 \pm 17\%$	1	88 ± 25
A7-BB3	60	0.16	$0.6 \pm 25\%$	$0.6 \pm 18\%$	2	100 ± 31
A7-BB4	60	0.22	$2.3 \pm 13\%$	$1.7 \pm 11\%$	1	74 ± 13

Before isolation

After isolation

Figure C-1. Photomicrographs of Radioactive Particle A5-5, from the Washing of Collection Plate M-3, After an Exposure of 5 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.55 and 0.54 μ , Respectively. The Magnification is Such That 10 μ is Equivalent to 6 mm.

Before isolation

After isolation

Figure C-2. Photomicrographs of Radioactive Particle A5-C2, from the Washing of Collection Plate M-3, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameter for Both Before and After Isolation is 0.39μ . The Magnification is Such That 10μ are Equivalent to 6 mm.

Before isolation

After isolation

Figure C-3. Photomicrographs of Radioactive Particle A5-C3, from the Washing of Collection Plate M-3, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.54 and 0.53 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 6 mm.

Before isolation

After isolation

Figure C-4. Photomicrographs of a Radioactive Particle, A5-D1 from the Washing of Collection Plate M-3. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.25 and 0.15 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 6 mm.

Before isolation

After isolation

Figure C-5. Photomicrographs of Radioactive Particle A5-D5, from the Washing of Collection Plate M-3, After and Exposure of 60 Min. The Equivalent $^{238}\text{PuO}_2$ Diameter for Both Before and After Isolation is 0.28μ . The Magnification is Such That 10μ are Equivalent to 6 mm.

Before isolation

One of 8 fragments isolated

Figure C-6. Photomicrographs of Radioactive Particle A9-1, from the Washing of Collection Plate M-15, After an Exposure of 5 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for the Particles Designated Above are 0.86 and 0.4 μ , Respectively. The Magnification is Such That 10 μ is Equivalent to 6 mm.

Before isolation

After isolation

Figure C-7. Photomicrographs of Radioactive Particle A9-C8 from the Washing of Collection Plate M-15. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are $0.46\ \mu$ and $0.54\ \mu$, Respectively. The Magnification is Such That $10\ \mu$ are Equivalent to $6\ \text{mm}$.

Before isolation

After isolation

Figure C-8. Photomicrographs of Radioactive Particle A9-C62, from the Washing of Collection Plate M-15, After an Exposure of 60 Min. The Equivalent $^{238}\text{PuO}_2$ Diameter for Both Before and After Isolation is $0.41\ \mu$. The Magnification is Such That $10\ \mu$ are Equivalent to 6 mm.

Before isolation

After isolation

Figure C-9. Photomicrographs of Radioactive Particle A7-1, from the Washing of Collection Plate M-35, After an Exposure of 5 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.94 and 0.98 μ , Respectively. The Magnification is such that 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-10. Photomicrographs of Radioactive Particle A7-2, from the Washing of Collection Plate M-35, After an Exposure of 5 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.84 μ , 0.55 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-11. Photomicrographs of Radioactive Particle A7-3, from the Washing of Collection Plate M-35, After an Exposure of 5 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.72 and 0.68 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-12. Photomicrographs of Radioactive Particle A7-B3, from the Washing of Collection Plate M-35, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.43 and 0.27 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After Isolation

Figure C-13. Photomicrographs of Radioactive Particle A7-B5, from the Washing of Collection Plate M-35, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.40 and 0.31 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-14. Photomicrographs of Radioactive Particle A7-B14, from the Washing of Collection Plate M-35, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.49 and 0.26 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-15. Photomicrographs of Radioactive particle A7-BB54 from the Washing of Collection Plate M-35. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.40 and 0.38μ , Respectively. The Magnification is Such That 10μ are Equivalent to 5 mm .

Before isolation

Larger of two
fragments isolated

Figure C-16. Photomicrographs of Radioactive Particle C7-C-38 from the Washing of Collection Plate M-35. The Equivalent $^{238}\text{PuO}_2$ Diameter for the Particle Before Isolation is 0.28μ and After Isolation, 0.16 and 0.11μ for the Fragments Isolated. The Magnification is Such That 10μ is Equivalent to 5 mm .

Before isolation

After isolation

Figure C-17. Photomicrographs of Radioactive Particle C7-CB-84 from the Washing of Collection Plate M-35. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.18 and 0.21 μ , Respectively. The Magnification is Such That 10 μ is Equivalent to 5 mm.

Before isolation

After isolation

Figure C-18. Photomicrographs of Radioactive Particle C7-CE-48 from the Washing of Collection Plate M-35. The Equivalent $^{238}\text{PuO}_2$ Diameter for Both Before and After Isolation is 0.09μ . The Magnification is Such That 10μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-19. Photomicrographs of Radioactive Particle A2-3 from the Washing of Collection Plate M-29. The Equivalent $^{238}\text{PuO}_2$ Diameter After Isolation is $13.5\ \mu$. The Magnification is Such That $10\ \mu$ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-20. Photomicrographs of Radioactive Particle A6-6 from the Washing of Collection Plate M-29. Before Isolation, This Radioactive Particle was Obscured by the Much Larger Particle, as Shown Above. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 2.7 and $3.0\ \mu$, Respectively. The Magnification is Such That $10\ \mu$ is Equivalent to $5\ \text{mm}$.

Before isolation

After isolation

Figure C-21. Photomicrographs of Radioactive Particle A6-11, from the Washing of Collection Plate M-29, After an Exposure of 1/4 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 7.8 and 9.1 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm. This Particle was Examined by Electron Microprobe Analysis. The Results are Presented in Appendix A.

Before isolation

After isolation

Figure C-22. Photomicrographs of Radioactive Particle A6-B3 from the Washing of Collection Plate M-29. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 1.3 and 0.14 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-23. Photomicrographs of Radioactive Particle A6-B6 from the Washing of Collection Plate M-29. The Equivalent $^{238}\text{PuO}_2$ Diameter for Both Before and After Isolation is 1.7μ . The Magnification is Such That 10μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-24. Photomicrographs of Radioactive Particle A6-B9 from the Washing of Collection Plate M-29. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 1.8 and 1.9μ , Respectively. The Magnification is Such That 10μ are Equivalent to 5 mm .

Before isolation

After isolation

Figure C-25. Photomicrographs of Radioactive Particle A6-BB2, from the Washing of Collection Plate M-29, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation 0.9 and 1.1 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-26. Photomicrographs of Radioactive Particle A2-1 from the Washing of Collection Plate M-37. The Equivalent $^{238}\text{PuO}_2$ Diameter for Both Before and After Isolation is 2.8μ .

Before isolation

After isolation

Figure C-27. Photomicrographs of Radioactive Particle A2-3 from the Washing of Collection Plate M-37. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 6.2 and 7.2 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-28. Photomicrographs of Radioactive Particle A2-7 from the Washing of Collection Plate M-37. The Equivalent $^{238}\text{PuO}_2$ Diameter After Isolation is 6.9μ . The Magnification is Such That 10μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-29. Photomicrographs of Radioactive Particle A2-C3 from the Washing of Collection Plate M-37. Before Isolation, the Radioactive Particle was Obscured by the Much Larger Particle, as Shown Above. The Equivalent $^{238}\text{PuO}_2$ Diameter Both Before and After Isolation is $1.3\ \mu$. The Magnification is Such That $10\ \mu$ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-30. Photomicrographs of Radioactive Particle A2-C12, from the Washing of Collection Plate M-37, After an Exposure of 2 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 1.3 and 1.4 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-31. Photomicrographs of Radioactive Particle A2-CC2, from the Washing of Collection Plate M-37, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.36 and 0.39 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-32. Photomicrographs of Radioactive Particle A2-CC3, from the Washing of Collection Plate M-37, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.43 and 0.38 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-33. Photomicrographs of Radioactive Particle A2-CC4, from the Washing of Collection Plate M-37, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.40 and 0.41 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-34. Photomicrographs of Radioactive Particle A2-CC6, from the Washing of Collection Plate M-37, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.78 and 0.72 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-35. Photomicrographs of Radioactive Particle A7-6 from the Washing of Collection Plate M-31. The Equivalent $^{238}\text{PuO}_2$ Diameters Before and After Isolation are 0.58 and 0.52μ , Respectively. The Magnification is Such That 10μ are Equivalent to 5 mm .

Before isolation

After isolation

Figure C-36. Photomicrographs of Radioactive Particle A7-7, from the Washing of Collection Plate M-31, After an Exposure of 10 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.48 and 0.47 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-37. Photomicrographs of Radioactive Particle A7-10, from the Washing of Collection Plate M-31, After an Exposure of 10 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.39 and 0.38 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-38. Photomicrographs of Radioactive Particle A7-B2, from the Washing of Collection Plate M-31, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.27 and 0.20 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-39. Photomicrographs of Radioactive Particle A7-B5, from the Washing of Collection Plate M-31, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.24 and 0.19 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-40. Photomicrographs of Radioactive Particle A7-B6, from the Washing of Collection Plate M-31, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.26 and 0.22 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-41. Photomicrographs of Radioactive Particle A7-B10, from the Washing of Collection Plate M-31, After an Exposure of 30 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.26 and 0.22 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

After isolation

Figure C-42. Photomicrographs of Radioactive Particle A7-BB1, from the Washing of Collection Plate M-31, After an Exposure of 60 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are 0.16 and 0.15 μ , Respectively. The Magnification is Such That 10 μ are Equivalent to 5 mm.

Before isolation

One of two fragments isolated

Figure C-43. Photomicrographs of Radioactive Particle A7-BB3, from the Washing of Collection Plate M-31, After an Exposure of 60 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for the Particles Designated Above are 0.15 and $0.11\ \mu$, Respectively. The Magnification is Such That $10\ \mu$ are Equivalent to $5\ \text{mm}$.

Before isolation

After isolation

Figure C-44. Photomicrographs of Radioactive Particle A7-BB4, from the Washing of Collection Plate M-31, After an Exposure of 60 Min. The Equivalent $^{238}\text{PuO}_2$ Diameters for Before and After Isolation are $0.22\ \mu$ and $0.21\ \mu$, Respectively. The Magnification is Such That $10\ \mu$ are Equivalent to 5 mm.

100X

200X

Figure C-45. Photomicrographs Prepared to Simplify Location of Particle A5-D1 from M-3, Which was Selected for Microprobe Analysis. The Two Scratches ("eyes") Around the Particle Designate it as the One Selected. Figure C-4 Shows This Particle at 600X.

July 28, 1967

TO: I. B. White, Sandia Corp.

FROM: C. F. Metz, Group Leader CMB-1

SUBJECT: Electron microprobe examination of particle in metallographic mount.

SAMPLE DESIGNATION: CMB-1-IBW-Sandia-M-29

SAMPLE DESCRIPTION: The sample was described as a particle mounted in a metallographic mount; possibly contaminated with Pu²³⁸.

ANALYSES REQUESTED: Determine whether silicon or any elements of atomic number greater than 20 are present in the particle.

ANALYTICAL RESULTS AND DISCUSSION:

The particle contains two phases: (1) ends of the particle contain iron, chromium, nickel, and possibly manganese, and (2) the center of the particle is a heterogeneous mixture predominantly of plutonium, chromium, silicon, calcium, and manganese with some iron and nickel. One small inclusion, less than 5 microns in diameter, was found to contain copper. Scandium, titanium, vanadium, cobalt, zinc, gallium, germanium, arsenic, bromine, rubidium, yttrium, zirconium, niobium, molybdenum, ruthenium, rhodium, palladium, silver, cadmium, indium, tin, antimony, tellurium, iodine, cesium, barium, lanthanum, cerium, hafnium, tantalum, tungsten, rhenium, osmium, iridium, platinum, gold, mercury, thallium, lead, bismuth, thorium, uranium, neptunium, and americium were sought but not detected in the sample. Detection limits for these elements in the sample are estimated to be in the range 0.1 to 1 percent.

ANALYST: E. A. Hakkila

DISTRIBUTION:

I. B. White, Sandia (2)
E. A. Hakkila
File

TO: I. B. White, Sandia

December 4, 1967

FROM: C. F. Metz, Group Leader CMB-1

SUBJECT: Electron microprobe examination of particles from $^{238}\text{PuO}_2$ ablation tests.

SAMPLE DESIGNATION: CMB-1-CFM-Sandia-IBW-M-29-A-2-3, -M-37-A-2-7

SAMPLE DESCRIPTION: The samples consist of particles from $^{238}\text{PuO}_2$ ablation tests which have been mounted to bakelite discs with collodion.

ANALYSES REQUESTED: Determine whether plutonium, iron, chromium, silicon, copper, tungsten, silver, nickel, or manganese are present in the particles.

ANALYTICAL RESULTS AND DISCUSSION:

M-29-A-2-3: The particle is approximately 20 microns in diameter (Figure 1a) and contains predominantly plutonium (Figure 1b), some silicon (Figure 1c), and trace amounts of iron (Figure 1d) and chromium (Figure 1e). Iron and chromium intensities from the particle are only slightly greater than background intensity. Note the small iron-chromium particle to the left of the particle examined. Silver, tungsten, copper, nickel, and manganese were not detected in this particle. Detection limits are estimated to be in the range between 0.1 and 1 percent.

M-37-A-2-7: The particle is approximately 12 microns in diameter (Figure 2a) and contains predominantly plutonium (Figure 2b) with trace amounts of iron (Figure 2c). Iron x-ray intensity from the particle is only slightly greater than background intensity. Silicon, chromium, manganese, nickel, copper, and tungsten were not detected in this particle. Detection limits are estimated to be in the range between 0.1 and 1 percent.

ANALYSTS: E. A. Hakkila and H. L. Barker

DISTRIBUTION:

I. B. White, Sandia
E. A. Hakkila
File

6 microns

Figure 1a. Backscattered electron intensity, Sample CMB-1-CFM-Sandia-IEW-M-29-A-2-3 (1500x). Particle is approximately 20 microns in diameter.

Figure 1b. Pu x-ray intensity, same area. Pu intensity outside of particle is equivalent to background intensity.

Figure 1c. Si x-ray intensity, same area. Trace amounts of Si are also present in plastic.

Figure 1d. Fe x-ray intensity, same area. Fe intensity in plastic is equivalent to background intensity. Note Fe-rich particle in plastic to the left of the particle examined.

Figure 1e. Cr x-ray intensity, same area. Cr x-ray intensity in plastic is equivalent to background intensity. Note Fe-rich and Cr-rich particle to the left of the particle examined.

6 microns

Figure 2a. Backscattered electron intensity, Sample CMB-1-CFM-Sandia-IBW-M-37-A-2-7 (1500x). Particle is approximately 12 microns in diameter.

Figure 2b. Pu x-ray intensity, same area. Pu intensity outside of particle is equivalent to background intensity.

Figure 2c. Fe x-ray intensity, same area. Note small amount of Fe to left of particle.

TO: I. B. White, Sandia

December 7, 1967

FROM: C. F. Metz, Group Leader CMB-1

SUBJECT: Electron microprobe examination of particles from $^{238}\text{PuO}_2$ ablation tests.

SAMPLE DESIGNATION: CMB-1-CFM-Sandia-IBW-M-3-A5-101, -M-15-A9-C8, -M-31-A-7-6.

SAMPLE DESCRIPTION: The samples consist of particles from $^{238}\text{PuO}_2$ ablation tests which have been mounted on bakelite discs with colodion.

ANALYSES REQUESTED: Determine whether plutonium, iron, chromium, silicon, copper, tungsten, silver, nickel, or manganese are present in the particles.

ANALYTICAL RESULTS AND DISCUSSION:

M-3-A5-101: The particle has an irregular shape, approximately 30 microns in length (Figure 1a), and contains predominantly silicon (Figure 1b) and some iron (Figure 1c). Manganese, chromium, nickel, copper, silver, tungsten, and plutonium were not detected in this particle. Detection limits are estimated to be in the range between 0.1 and 1 percent.

M-15-A9-C8: The particle is approximately 50 microns in diameter (Figure 2a) and contains predominantly silicon (Figure 2b), iron (Figure 2c), and copper (Figure 2d). Each element is heterogeneously distributed in the particle. A trace of chromium is present in one area of the particle (Figure 2e). Manganese, nickel, silver, tungsten, and plutonium were not detected in this particle. Detection limits are estimated to be in the range between 0.1 and 1 percent.

M-31-A-7-6: The particle has an irregular shape, approximately 100 microns in length (Figure 3a), and contains predominantly silicon (Figure 3b) and copper (Figure 3c), heterogeneously distributed, and trace amounts of iron (Figure 3d). Manganese, chromium, nickel, silver, tungsten, and plutonium were not detected in this sample. Detection limits are estimated to be in the range between 0.1 and 1 percent. A high density area on the sample was not identified.

I. B. White, Sandia

- 2 -

December 7, 1967

Photographs showing the intensity distributions of the elements that were not detected were not meaningful and were not included.

ANALYSTS: E. A. Hakkila and H. L. Barker

DISTRIBUTION:

I. B. White, Sandia

E. A. Hakkila

File

Figure 1a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-3-A5-101 (1500x). Particle has an irregular shape, approximately 30 microns in length.

Figure 1b. Si x-ray intensity distribution, same area. Si x-ray intensity from plastic is equal to background.

Figure 1c. Fe x-ray intensity distribution, same area. Fe x-ray intensity from plastic is equal to background.

6 microns

Figure 2a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-15-A9-C8 (1500x). Particle is approximately 50 microns in diameter.

Figure 2b. Si x-ray intensity distribution, same area. Si x-ray intensity from plastic is equal to background.

Figure 2c. Fe x-ray intensity distribution, same area. Fe x-ray intensity from plastic is equal to background.

Figure 2d. Cu x-ray intensity distribution, same area. Cu x-ray intensity from plastic is equal to background.

Figure 2e. Cr x-ray intensity distribution, same area. Cr is present only in area near lower center of photograph. Cr x-ray intensity from remainder of sample is equal to background.

12, microns

Figure 3a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-31-A-7-6 (800x). Particle has an irregular shape, approximately 100 microns in length. Note small high-density area at lower right of center in photograph.

Figure 3b. Si x-ray intensity distribution, same area. Si x-ray intensity from plastic is equal to background.

Figure 3c. Cu x-ray intensity distribution, same area. Cu x-ray intensity from plastic is equal to background.

Figure 3d. Fe x-ray intensity distribution, same area. Fe x-ray intensity from plastic is equal to background.

TO: I. B. White, Sandia

December 7, 1967

FROM: C. F. Metz, Group Leader CMB-1

SUBJECT: Electron microprobe examination of particles from $^{238}\text{PuO}_2$ ablation tests.

SAMPLE DESIGNATION: CMB-1-CFM-Sandia-IBW-M-37-A-2-1, -M-37-A-2-3, -M-37-A2-C3.

SAMPLE DESCRIPTION: The samples were described as particles from $^{238}\text{PuO}_2$ ablation tests which had been mounted on bakelite with collodion.

ANALYSES REQUESTED: Determine if plutonium, iron, chromium, silicon, copper, tungsten, silver, nickel, or manganese are present in the particles.

ANALYTICAL RESULTS AND DISCUSSION:

M-37-A-2-1: The particle is approximately 6 microns in diameter (Figure 1a) and contains predominantly plutonium (Figure 1b), with some silicon (Figure 1c), and chromium (Figure 1d), and trace amounts of iron (Figure 1e), manganese (Figure 1f), and copper (Figure 1g). Silver, nickel, and tungsten were not detected in this particle. Detection limits are estimated to be in the range between 0.1 and 1 percent.

M-37-A-2-3: The particle is approximately 15 microns in diameter (Figure 2a) and contains predominantly plutonium (Figure 2b) with some silicon (Figure 2c), chromium (Figure 2d), iron (Figure 2e), and manganese (Figure 2f). Nickel, copper, silver, and tungsten were not detected in this particle. Detection limits are estimated to be in the range between 0.1 and 1 percent.

M-37-A2-C3: The particle is approximately 3 microns in diameter (Figure 3a) and contains predominantly plutonium (Figure 3b) and chromium (Figure 3c), with some iron (Figure 3d), and trace amounts of silicon (Figure 3e). Manganese, nickel, copper, silver, and tungsten were not detected in this particle. Detection limits are estimated to be in the range between 0.1 and 1 percent.

Photographs showing the x-ray intensity distributions of the elements that were not detected are not meaningful and are not included.

ANALYSTS: E. A. Hakkila and H. L. Barker

DISTRIBUTION:

I. B. White, Sandia

E. A. Hakkila

File

Figure 1a. Backscatter electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-37-A-2-1 (3000x). Particle is approximately 6 microns in diameter.

Figure 1b. Pu x-ray intensity distribution, same area. Pu x-ray intensity from plastic is equal to background intensity.

Figure 1c. Si x-ray intensity distribution, same area. Si x-ray intensity from plastic is equal to background intensity.

Figure 1d. Cr x-ray intensity distribution, same area. Cr x-ray intensity from plastic is equal to background intensity.

Figure 1e. Fe x-ray intensity distribution, same area. Some Fe is also present in plastic above particle; Fe x-ray intensity from remainder of plastic is equal to background.

Figure 1f. Mn x-ray intensity distribution, same area. Mn x-ray intensity from plastic is equal to background.

Figure 1g. Cu x-ray intensity distribution, same area. Cu x-ray intensity from plastic is equal to background.

3 microns

Figure 2a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-37-A-2-3 (3000x). Particle is approximately 15 microns in diameter.

Figure 2b. Pu x-ray intensity distribution, same area. Pu x-ray intensity from plastic is equal to background.

Figure 2c. Si x-ray intensity distribution, same area. Trace amounts of Si are also present in plastic.

Figure 2d. Cr x-ray intensity distribution, same area. Cr x-ray intensity from plastic is equal to background.

Figure 2e. Fe x-ray intensity distribution, same area. Fe x-ray intensity from plastic is equal to background.

Figure 2f. Mn x-ray intensity distribution, same area. Mn x-ray intensity from plastic is equal to background.

Figure 3a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-37-A2-C3 (3000x). Particle is approximately 3 microns in diameter.

Figure 3b. Pu x-ray intensity distribution, same area. Pu x-ray intensity from plastic is equal to background.

Figure 3c. Cr x-ray intensity distribution, same area. Cr x-ray intensity from plastic is equal to background.

Figure 3d. Fe x-ray intensity distribution, same area. Fe x-ray intensity from plastic is equal to background.

Figure 3e. Si x-ray intensity distribution, same area. Si x-ray intensity from plastic is equal to background.

TO: I. B. White, Sandia

December 12, 1967

FROM: C. F. Metz, Group Leader CMB-1

SUBJECT: Electron microprobe examination of particles from $^{238}\text{PuO}_2$ ablation tests.

SAMPLE DESIGNATION: CMB-1-CFM-Sandia-IBW-M-29-A6-B3, -M-29-A6-6, -M-29-A6-B6.

SAMPLE DESCRIPTION: The samples were described as particles from ablation tests which had been mounted on bakelite with collodion.

ANALYSES REQUESTED: Determine if plutonium, iron, chromium, silicon, copper, tungsten, silver, nickel, or manganese are present in the particles.

ANALYTICAL RESULTS AND DISCUSSION:

M-29-A6-B3: The particle is approximately 5 microns in diameter and is partially surrounded by a lower density material (Figure 1a). The particle contains predominantly plutonium (Figure 1b), with some silicon (Figure 1c) and chromium (Figure 1d), and trace amounts of iron (Figure 1e), copper (Figure 1f), and manganese (Figure 1g). The low density material contains predominantly silicon and trace amounts of iron and copper. Nickel, silver, and tungsten were not detected in this sample. Detection limits are estimated to be in the range between 0.1 and 1 percent.

M-29-A6-6: The particle is approximately 8 microns in diameter (Figure 2a) and contains predominantly plutonium (Figure 2b), some silicon (Figure 2c) and chromium (Figure 2d), and trace amounts of iron (Figure 2e) and manganese (Figure 2f). Nickel, copper, silver, and tungsten were not detected in this sample. Detection limits are estimated to be in the range between 0.1 and 1 percent.

M-29-A6-B6: The particle is approximately 4 microns in diameter and is partially surrounded by a low density material (Figure 3a). The particle contains predominantly plutonium (Figure 3b) and trace amounts of silicon (Figure 3c) and iron (Figure 3d). The low density material contains some plutonium, silicon,

I. B. White, Sandia

- 2 -

December 12, 1967

and iron. Chromium, manganese, nickel, copper, silver, and tungsten were not detected in this sample. Detection limits are estimated to be in the range between 0.1 and 1 percent.

Photographs showing the x-ray intensity distributions of the elements that were not detected were not meaningful and were not included.

ANALYSTS: E. A. Hakkila and H. L. Barker

DISTRIBUTION:

I. B. White, Sandia

E. A. Hakkila

File

3 microns

Figure 1a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-29-A6-B3 (3000x). Particle is approximately 5 microns in diameter. Note lower density material partially surrounding particle.

Figure 1b. Pu x-ray intensity distribution, same area. Pu x-ray intensity from plastic is equal to background.

Figure 1c. Si x-ray intensity distribution, same area. Note Si is present in particle and in low density material, with trace amounts of Si also detected in some of the plastic.

Figure 1d. Cr x-ray intensity distribution, same area. Cr x-ray intensity from plastic is equal to background.

Figure 1e. Fe x-ray intensity distribution, same area. Trace amounts of Fe are present in the particle and low density material. Fe x-ray intensity from plastic is equal to background.

Figure 1f. Cu x-ray intensity distribution, same area. Trace amounts of Cu are present in the particle and some of the low density material. Cu x-ray intensity from plastic is equal to background.

Figure 1g. Mn x-ray intensity distribution, same area. Trace amounts of Mn are present in the particle. Mn x-ray intensity from the low density material and from the plastic is equal to background.

3 microns

Figure 2a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-29-A6-6 (3000x). Particle is approximately 8 microns in diameter.

Figure 2b. Pu x-ray intensity distribution, same area. Pu x-ray intensity from plastic is equal to background.

Figure 2c. Si x-ray intensity distribution, same area. Si is a major constituent of the particle, with trace amounts of Si also present in the plastic.

Figure 2d. Cr x-ray intensity distribution, same area. Cr x-ray intensity from plastic is equal to background.

Figure 2e. Fe x-ray intensity distribution, same area. Fe x-ray intensity from plastic is equal to background.

Figure 2f. Mn x-ray intensity distribution, same area. Mn x-ray intensity from plastic is equal to background.

3 microns

Figure 3a. Backscattered electron intensity distribution, Sample CMB-1-CFM-Sandia-IBW-M-29-A6-B6 (3000x). Particle is approximately 4 microns in diameter. Note lower density material partially surrounding particle.

Figure 3b. Pu x-ray intensity distribution, same area. Note trace amounts of Pu are present in low density material. Pu x-ray intensity from plastic is equal to background.

Figure 3c. Si x-ray intensity distribution, same area. Trace amounts of Se are present in particle and low density material. Si x-ray intensity from plastic is equal to background.

Figure 3d. Fe x-ray intensity distribution, same area. Trace amounts of Fe are present in particle and low density material. Fe x-ray intensity from plastic is equal to background.

APPENDIX D

NUMERICAL SIZE DISTRIBUTION HISTOGRAMS
OF $^{238}\text{PuO}_2$ PARTICLES DETERMINED BY
THE HOLLOW STAR TECHNIQUE

Figure D-1. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found on Collection Plate M-3 Before Washing. A Total of 23 Particles was Found.

Figure D-2. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found on Collection Plate M-35 Before Washing. A Total of 125 Particles wa Found.

Figure D-3. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found on Collection Plate M-29 Before Washing. A Total of 1164 Particles was Found.

Figure D-4. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found on Collection Plate M-37 Before Washing. A Total of 517 Particles was Removed.

Figure D-5. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found in the Washing from Collection Plate M-3. The Total Equivalent $^{238}\text{PuO}_2$ Mass for the 316 Particles Found is 9.3×10^{-11} g.

Figure D-6. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found in the Centrifuged Residue of the Washing of Collection Plate M-15. The Total Equivalent $^{238}\text{PuO}_2$ Mass for the 1954 Particles Found is 1.1×10^{-9} g.

Figure D-7. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found in the Centrifuged Residue of the Washing from Collection Plate M-35. The Total Equivalent $^{238}\text{PuO}_2$ Mass for the 2705 Particles Found is 1.4×10^{-9} g.

Figure D-8. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found in the Centrifuged Residue of the Washing from Collection Plate M-29. The Total Equivalent $^{238}\text{PuO}_2$ Mass for the 2260 Particles Found is 6.7×10^{-8} g.

Figure D-9. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found in the Centrifuged Residue of the Washing from Collection Plate M-37. The Total Equivalent $^{238}\text{PuO}_2$ Mass for the 3460 Particles Found is 1.8×10^{-8} g.

Figure D-10. Numerical Size Distribution Histogram of Equivalent $^{238}\text{PuO}_2$ Particles Found in the Washing from Collection Plate M-31. The Total Equivalent $^{238}\text{PuO}_2$ Mass for the 352 Particles Found is 8.0×10^{-11} g.

BLANK

APPENDIX E

CALCULATION OF THE CUMULATIVE MASS DISTRIBUTION VALUES FOR EQUIVALENT $^{238}\text{PuO}_2$ PARTICLES COLLECTED ON IMPACTORS

E-1. CALCULATION OF THE MASS DISTRIBUTION FOR PARTICLES COLLECTED ON IMPACTOR M-3

The results of analyses of radioactive particles collected on impactor M-3 are tabulated in Table E-I.

Table E-I. Results of Radiochemical Analyses		
Portion Analyzed	$^{238}\text{PuO}_2$ α Activity (dpm)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
Centrifuged Residue	9.7×10^5	2.9×10^{-2}
Centrifuged Supernate	1.3×10^6	3.8×10^{-2}
Washed Impactor	1.3×10^{-5}	4×10^{-3}

From the analysis of single particles by the hollow-star and electronic-counting techniques, it was determined that for radioactive particles in the centrifuged residue (those which were transferred to the microscope slides) the equivalent $^{238}\text{PuO}_2$ diameter size range is $0.3 - 0.9 \mu$, and the equivalent $^{238}\text{PuO}_2$ mass is $9.3 \times 10^{-5} \mu\text{g}$.

The equivalent $^{238}\text{PuO}_2$ mass of particles with diameters smaller than 0.3μ which are on the microscope slides is calculated as follows:

$$\begin{array}{rcl}
 \text{Centrifuged Residue} & 0.029 & \mu\text{g} \\
 \text{Total for Individual} & & \\
 \text{Particles Measured} & -0.000093 & \mu\text{g} \\
 & \hline
 & \sim 0.029 & \mu\text{g}
 \end{array}$$

The amount of plutonium found on the washed impactor must be included with the masses listed in Table E-II:

Assuming that the ^{238}Pu remaining on the washed impactor was in the form of particles with the same equivalent $^{238}\text{PuO}_2$ size distribution as those washed from the impactor, the percent mass values for the particles washed from the impactor, shown in Table E-II, will be the same as the percent mass values of the particles collected on impactor M-3. (The percentages for the overlapping ranges $0 - 0.09 \mu$ and $0 - 0.3 \mu$ are combined to produce a total of 99.86% for the 0 to 0.3μ range.)

Table E-II. Breakdown of Plutonium Found on Microscope Slides and in Centrifuged Residue			
Source	Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	% of Subtotal
Supernate	0 - 0.09	3.8×10^{-2}	56.66
Slides	0 - 0.3	2.9×10^{-3}	43.20
Slides	0.3 - 0.9	9.3×10^{-5}	0.14
		<hr/>	
		Total 0.067	

To calculate the equivalent $^{238}\text{PuO}_2$ masses for the size ranges of 0 - 0.3 μ and 0.3 - 0.9 μ for the particles collected on impactor M-3, the amount washed from the impactor, 0.067 μg , is added to 4×10^{-3} μg , the amount that remained on the impactor after washing. This total, 0.07 μg , is multiplied by each of the mass percents in Table E-II to obtain the desired values. These values are listed in Table E-III.

Table E-III. Percentages of Total Mass for Two Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-3			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0 - 0.3	6.99×10^{-2}	99.4	99.9
0.3 - 0.9	1×10^{-4}	0.14	100

E-2. CALCULATION OF THE MASS DISTRIBUTION FOR PARTICLES COLLECTED ON IMPACTOR M-15

The results of analyses of radioactive particles collected on impactor M-15 are tabulated in Table E-IV.

Table E-IV. Results of Radiochemical Analyses		
Portion Analyzed	^{238}Pu α Activity (dpm)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
Centrifuged Residue	6.7×10^6	2.0×10^{-1}
Centrifuged Supernate	1.3×10^6	4.1×10^{-2}
Washed Impactor	1.9×10^5	5.6×10^{-1}

From the analysis of single particles by the hollow-star and electronic-counting techniques, it was determined for radioactive particles in the centrifuged residue (those that were transferred to the microscope slides) that the equivalent $^{238}\text{PuO}_2$ diameter size range is 0.2 to 1.4 μ , and the equivalent $^{238}\text{PuO}_2$ mass is 1.1×10^{-3} μg .

The equivalent $^{238}\text{PuO}_2$ mass of particles with diameters smaller than 0.2 μ that are on the microscope slides is calculated as follows:

Centrifuged Residue 0.2000 μg
Total for Individual
Particles Measured -0.0011 μg
0.1989 μg

The amount of plutonium on the washed impactor must be included with the masses listed in Table E-V:

Table E-V. Breakdown of Plutonium Found on Microscope Slides and in Centrifuged Residue			
Source	Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	% of Subtotal
Supernate	0 - ~0.09	4.1×10^{-2}	16.9
Slides	0 - 0.2	2.0×10^{-1}	82.6
Slides	0.2 - 1.4	1.1×10^{-3}	0.5
		Total 2.421×10^{-1}	

Assuming that the ^{238}Pu remaining on the washed impactor was in the form of particles with the same equivalent $^{238}\text{PuO}_2$ size distribution as those washed from the impactor, the percent mass values for the particles washed from the impactor, shown in Table E-V, will be the same as the percent mass values of the particles collected on impactor M-15. (The percentages for the overlapping ranges 0 - 0.09 μ and 0 - 0.2 μ are combined to produce a total of 99.5% for the 0 - 0.2 μ range.)

To calculate the equivalent $^{238}\text{PuO}_2$ masses for the size ranges of 0 - 0.2 μ and 0.2 - 1.4 μ for the particles collected on impactor M-15, the amount washed from the impactor, 5.6×10^{-1} μg , is added to 2.42×10^{-1} μg , the amount that remained on the impactor after washing. This total (8.02×10^{-1} μg) is multiplied by each of the mass percents in Table E-V to obtain the desired values. These values are listed in Table E-VI.

Table E-VI. Percentages of Total Mass for Two Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-15			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0 - 0.2	8.0×10^{-1}	99.5	99.5
0.2 - 1.4	4×10^{-3}	0.5	100

E. 3. CALCULATION OF THE MASS DISTRIBUTION FOR PARTICLES COLLECTED ON IMPACTOR M-35

The results of analyses of radioactive particles collected on impactor M-35 are tabulated in Table E-VII.

From the analysis of single particles by the hollow-star and electronic-counting techniques, it was determined for radioactive particles in the centrifuged residue (those that were transferred to the microscope slides) that:

Table E-VII. Results of Radiochemical Analyses		
Portion Analyzed	^{238}Pu α Activity (dpm)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
Centrifuged Residue	9.81×10^6	0.29
Centrifuged Supernate	9.84×10^5	0.03
Washed Impactor	1.01×10^6	0.03

Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
0.05 - 0.1	0.0112
0.1 - 1.6	0.0025
	<hr/>
Total	0.0137

The equivalent $^{238}\text{PuO}_2$ mass of particles with diameters smaller than 0.05μ that are on the microscope slides is calculated as follows:

Centrifuged Residue	0.29 μg
Total for Individual	
Particles Measured	$\frac{-0.0137 \mu\text{g}}{0.2863 \mu\text{g}}$

The amount of plutonium found on the washed impactor plate must be included with the masses listed in Table E-VIII:

Table E-VIII. Breakdown of Plutonium Found on Microscope Slides and in Centrifuged Residue			
Source	Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	% of Subtotal
Slides	0 - 0.05	0.2863	86.76
Slides	0.05 - 0.1	1.12×10^{-2}	3.39
Slides	0.1 - 1.6	2.5×10^{-3}	0.76
Centrifuged Supernate	0 - 0.05	3.0×10^{-2}	9.09

Assuming that the ^{238}Pu remaining on the washed impactor was in the form of particles with the same equivalent $^{238}\text{PuO}_2$ size distribution as those washed from the impactor, the percent mass values for the particles washed from the impactor, (shown in Table E-VIII) will be the same as the percent mass values for the particles collected on impactor M-35. (The percentages for the range of 0 - 0.05μ for the slides and the range of 0 - 0.05μ for the supernate are added to produce a total of 95.85 percent.)

To calculate the equivalent $^{238}\text{PuO}_2$ masses for the size ranges of 0 - 0.05μ , 0.05 - 0.1μ , and 0.1 - 1.6μ for the particles collected on impactor M-35, the amount washed from the impactor, $0.33 \mu\text{g}$, is added to $0.03 \mu\text{g}$, the amount that remained on the impactor after washing. This total, $0.36 \mu\text{g}$, is multiplied by each of the mass percents in Table E-VIII to obtain the desired values. These values are listed in Table E-IX:

Table E-IX. Percentages of Total Mass for Three Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-35			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0 - 0.05	3.4×10^{-1}	95.8	95.8
0.05 - 0.1	1.2×10^{-2}	3.39	99.2
0.1 - 1.6	2.7×10^{-3}	0.76	100

E-4. CALCULATION OF THE MASS DISTRIBUTION OF PARTICLES COLLECTED ON IMPACTOR M-29

The results of analyses of radioactive particles collected on impactor M-29 are tabulated in Table E-X.

Table E-X. Results of Radiochemical Analyses		
Portion Analyzed	^{238}Pu Activity (dpm)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
Centrifuged Residue	1.16×10^7	0.35
Centrifuged Supernate	3.4×10^7	1.0
Washed Impactor	1.18×10^7	0.35

From the analyses of single particles by the hollow-star and electronic-counting techniques, it was determined for radioactive particles in the centrifuged residue (those transferred to microscope slides) that:

Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
0.2 - 0.6	8.0×10^{-4}
0.6 - 1.0	1.8×10^{-3}
1.0 - 14.0	6.47×10^{-2}
Total	6.73×10^{-2}

The equivalent $^{238}\text{PuO}_2$ mass of particles with equivalent diameters smaller than 0.2μ that is on the microscope slides is calculated as follows:

Centrifuged Residue	0.35 μg
Total for Individual	
Particles Measured	$- 0.0673 \mu\text{g}$
	$0.2827 \mu\text{g}$

The amount of plutonium found on the washed impactor must be added to the masses listed in Table E-XI:

Table E-XI. Breakdown of Plutonium Found on Microscope Slides and in the Centrifuged Residue			
Source	Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	Cumulative %
Supernate	0 - 0.09	1.0	74.1
Slides	0 - 0.2	0.2827	21.0
Slides	0.2 - 0.6	8×10^{-4}	0.06
Slides	0.6 - 1.0	1.8×10^{-3}	0.1
Slides	1.0 - 14.0	6.47×10^{-2}	4.8
		Total 1.350	

Assuming that the $^{238}\text{PuO}_2$ remaining on the washed impactor was in the form of particles with the same equivalent $^{238}\text{PuO}_2$ size distribution as that of the particles washed from the impactor, the percent mass values for the particles washed from the impactor (shown in Table E-XI) will be the same as the percent mass values for the particles collected on impactor M-29. (The percentages for the 0 - 0.09 μ and 0 - 0.2 μ ranges are added to produce a total of 95.1%.)

To calculate the equivalent $^{238}\text{PuO}_2$ masses for the size ranges of 0 - 0.2 μ , 0.2 - 0.6 μ , 0.6 - 1.0 μ , and 1.0 - 14.0 μ for the particles collected on impactor M-29, the amount washed from this impactor, 1.350 μg , is added to 0.35 μg , the amount that remained on the impactor after washing. This total, 1.70 μg , is multiplied by each of the mass percents in Table E-XI to obtain the desired values, which are listed in Table E-XII.

Table E-XII. Percentages of Total Mass for Four Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-29			
Size Range of Particle Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0 - 0.2	1.62	95.1	95.1
0.2 - 0.6	1.02×10^{-3}	0.06	95.2
0.6 - 1.0	1.7×10^{-3}	0.1	95.3
1.0 - 14.0	8.16×10^{-2}	4.8	100

E-5. CALCULATION OF THE MASS DISTRIBUTION FOR PARTICLES COLLECTED ON IMPACTOR M-37

The results of analyses of radioactive particles collected on impactor M-37 are tabulated in Table E-XIII.

Table E-XIII. Results of Radiochemical Analyses		
Portion Analyzed	^{238}Pu α Activity (dpm)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
Centrifuged Residue	7.51×10^6	2.3×10^{-1}
Centrifuged Supernate	1.07×10^6	3.2×10^{-2}
Washed Impactor	8.21×10^6	2.5×10^{-1}

From the analysis of single particles by the hollow-star and electronic-counting techniques, it was determined for radioactive particles in the centrifuged residue (those that were transferred to microscope slides) that:

Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)
0.2 - 0.6	1.5×10^{-3}
0.6 - 1.0	8.0×10^{-4}
1.0 - 8.0	1.62×10^{-2}
Total	1.85×10^{-2}

The equivalent $^{238}\text{PuO}_2$ mass of particles with diameters smaller than 0.2μ which were on the microscope slides is calculated as follows:

Centrifuged	$2.3 \times 10^{-1} \mu\text{g}$
Total for Individual Particles Measured	$-0.185 \times 10^{-1} \mu\text{g}$
	$2.115 \times 10^{-1} \mu\text{g}$

The amount of plutonium found on the washed impactor must be included with the masses listed in Table E-XIV:

Table E-XIV. Breakdown of Plutonium Found on Microscope Slides and in Centrifuged Residue			
Source	Equivalent $^{238}\text{PuO}_2$ Diameter Size Range (μ)	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	% of Subtotal
Microscope Slides	0.0 - 0.2	2.115×10^{-1}	80.7
Microscope Slides	0.2 - 0.6	1.50×10^{-3}	0.57
Microscope Slides	0.6 - 1.0	8.0×10^{-4}	0.31
Microscope Slides	1.0 - 8.0	1.62×10^{-2}	6.18
Microscope Slides	0.0 - 0.09	3.20×10^{-2}	12.2

Assuming that the ^{238}Pu that remained on the washed impactor was in the form of particles with the same equivalent $^{238}\text{PuO}_2$ size distribution as that of those washed from the impactor, the percent mass values for the particles washed from the impactor (shown in Table E-XIV) will be the same as the percent mass values of the particles collected on impactor M-37. (The percentages for the overlapping ranges $0 - 0.09 \mu$ and $0 - 0.2 \mu$ are combined to produce a total of 92.9% for the 0 to 0.2μ range.

To calculate the equivalent $^{238}\text{PuO}_2$ masses for the size ranges of $0 - 0.2 \mu$, $0.2 - 0.6 \mu$, $0.6 - 1.0 \mu$, and $1.0 - 8.0 \mu$ for the particles collected on impactor M-37, the amount washed from the impactor, $2.6 \times 10^{-1} \mu\text{g}$, is added to $2.5 \times 10^{-1} \mu\text{g}$, the amount that remained on the impactor after washing. This total, $5.12 \times 10^{-1} \mu\text{g}$, is multiplied by each of the mass percents in Table E-XIV to obtain the desired values. These values are listed in Table E-XV.

Table E-XV. Percentages of Total Mass for Four Size Ranges of Equivalent $^{238}\text{PuO}_2$ Particles Collected on Impactor M-37

Size Range of Particle Diameters	Equivalent $^{238}\text{PuO}_2$ Mass (μg)	%	Cumulative %
0.0 - 0.2	4.8×10^{-1}	92.9	92.9
0.2 - 0.6	2.9×10^{-3}	0.57	93.5
0.6 - 1.0	1.6×10^{-3}	0.31	93.8
1.0 - 8.0	3.2×10^{-2}	6.18	100

APPENDIX F

STANDARD DEVIATIONS OF ALPHA ACTIVITIES FOR PARTICLES AFTER ISOLATION, AND PERCENTAGES OF INITIAL ALPHA ACTIVITIES RECOVERED

For particle A7-B14 from M-35, the standard deviation of the alpha activity after isolation is obtained as follows:

The standard deviations of the alpha activities found for each of the isolated fragments were:

$\sigma\%$	DPM	σ^*	σ^2
14.7	3.64	0.535	0.29
22.3	1.58	0.35	0.12
31.6	0.79	0.25	0.06
33.3	<u>0.71</u>	0.24	<u>0.06</u>
	$Q = 6.72$		$\sum_{i=1}^n \sigma_i^2 = 0.53$

*These standard deviations were based on the Poisson statistics of the actual track counts of the individual hollow stars.

The formula used to calculate the standard deviation associated with the total alpha activity of the fragments is ^(F1):

$$\sigma_Q = \left(\sum_{i=1}^n \sigma_i^2 \right)^{\frac{1}{2}}$$

where σ_Q is the standard deviation of the sum Q

and σ_i is the standard deviation of each independent value that is added to calculate Q.

Thus σ_Q for A7-B14 from M-35 is:

$$\sigma_Q = (0.53)^{\frac{1}{2}} = 0.73$$

and

$$\sigma_Q \% = \frac{\sigma_Q \times 100}{Q} = \frac{0.73 \times 100}{6.72} = 11\%$$

To calculate the standard deviation of the percentage of ²³⁸Pu recovered after the particle was isolated, the following formula ^(F1) was used:

$$\sigma_Q = \left[\left(\frac{x}{y} \right)^2 \left(\frac{\sigma_x^2}{x^2} + \frac{\sigma_y^2}{y^2} \right) \right]^{\frac{1}{2}}$$

^(F1)Overman, R., and Clark, H., Radioisotope Techniques, McGraw-Hill, New York (1960), page 108.

where x is the dpm of ^{238}Pu after isolation
 and y is the dpm of ^{238}Pu before isolation.

thus

$$\begin{aligned}x &= 6.7 \\y &= 24 \\ \sigma_x &= 6.7 \times 11\% = 0.74 \\ \sigma_y &= 24 \times 6\% = 1.4\end{aligned}$$

also

$$\begin{aligned}\sigma_Q^2 &= (0.28)^2 \frac{0.55}{45} + \frac{2.0}{570} \\ &= (0.08)(1.2 \times 10^{-2} + 3.54 \times 10^{-3}) \\ &= (0.08)(1.5 \times 10^{-2}) = 1.2 \times 10^{-2} \\ \sigma_Q &= 0.11 \text{ or } 11\%\end{aligned}$$

APPENDIX G

SPECIAL PROCEDURE FOR RADIOCHEMICAL ANALYSIS OF PLUTONIUM COLLECTED ON IMPACTOR PLATES*

1. Removal of the Plutonium-Containing Particles from the Microscope Slides

Add a few drops of 50% collodion to each of the slides that contain radioactive particles covered by 4% collodion. After the collodion has dried, strip it from the slides with a razor blade and place it in a teflon beaker. Add fuming HNO_3 to dissolve the collodion. Then treat repeatedly with HF to dispel the silica. Boil off all HF and transfer residue to a glass beaker with distilled water. Evaporate the solution to ~5 ml and proceed to Step 4.

2. Removal of the Plutonium from the Washed Impactor Plate

Hold the impactor plate over a 1000-ml beaker and pipette HNO_3 over the impactor and rinse with H_2O , testing occasionally with an α -monitor to see if the plutonium has been completely removed. Final volume of HNO_3 and H_2O is about 500 ml. Evaporate the solution to ~5 ml and proceed to Step 4.

3. Dissolution of Plutonium-Coating Particles Remaining in the Centrifuged Supernate

Evaporate the centrifuged residue to ~5 ml and proceed to Step 4.

4. To each of the residues suspended in the 5-ml portions of liquid, add 200 ml of fuming HNO_3 and evaporate to near dryness. Add 120 ml of fuming H_2SO_4 and boil to near dryness. Add more fuming H_2SO_4 and boil to near dryness. If no residue remains, proceed to Step 5. If after several additions of the fuming H_2SO_4 and boiling to near dryness, some residue remains, treat it as follows:

- a. Transfer it to a small boat and ash in a Tracerlab LTA-600 Low-Temperature Asher.
- b. With fuming H_2SO_4 , transfer the ash to a clean 250-ml beaker. Boil to near dryness and add 20-ml portions of fuming H_2SO_4 , boiling to near dryness each time until the residue is completely dissolved. Now proceed to Step 5.

5. Add 2 ml of HNO_3 and transfer with water and HNO_3 to an appropriate-sized flask. For samples with a count rate of $\sim 10^6$ dpm, a one-liter volumetric flask is appropriate. Add H_2O to dilute to one liter.

6. Stipple a 10 λ (0.01 ml) portion of each sample on a platinum disc and alpha count in a scaler. From this approximation of the concentration of the alpha activity, select an aliquot with a dpm of ~100 for radiochemical decontamination.

* The work in Steps 1 through 6 is performed in a high-level radiochemical laboratory. The remaining work with the aliquoted samples will be performed in a low-level laboratory.

7. Transfer each aliquot to a 100-ml beaker that contains ^{236}Pu tracer. Boil in 20 ml of aqua regia until the volume is about 2 ml to equilibrate the sample and tracer activities.
8. Transfer the samples to 40-ml centrifuge cones with 2 ml HNO_3 and 2 ml H_2O . Add 5 mg of Fe scavenge to each.
9. Add NH_4OH until the solutions are basic. Digest in a hot H_2O bath for 5 minutes. Centrifuge and discard the supernate.
10. Wash each precipitate with 5 ml of H_2O that contains 2 drops of NH_4OH . Centrifuge and discard the supernate.
11. Dissolve each precipitate in a minimum of HNO_3 . Dilute to 10 ml. Repeat Steps 9 and 10 once.
12. Dissolve each precipitate in 5 ml of 6N HNO_3 . Add 5 drops of saturated NaBrO_4 and digest in a hot H_2O bath for 5 minutes.
13. Saturate the solutions with crystalline NH_4NO_3 . Extract the plutonium with 5 ml of hexone by stirring 3 minutes, centrifuging, and pipetting the hexone phases into clean 40-ml centrifuge cones.
14. Repeat the extraction twice, combining the hexone phases.
15. Wash each combined phase by stirring for one minute with 5 ml of 6N HNO_3 . Centrifuge and pipette off the aqueous phases, combining them with the original aqueous phases from Step 13.
16. Back extract the plutonium by stirring for 3 minutes with 5 ml of 0.1N HNO_3 , centrifuging, and pipetting off the aqueous phases into clean 50-ml beakers.
17. Repeat Step 16 twice, combining all the aqueous phases in the 50-ml beakers.
18. Boil the aqueous phases to near dryness, take up in 3 ml of HCl and 1 ml of HNO_3 , and boil to wet dryness. Repeat the aqua regia boiling until the ammonium salts are removed.
19. Boil in HCl until all HNO_3 is dispelled and continue boiling to wet dryness. Add 1 ml of HCl and boil to 1/2 ml.
20. Transfer samples to standard Tracerlab electroplating cells by pipetting. Wash beakers with 10 drops of HCl , then with 8 drops of HCl , and last with 1 ml of H_2O .
21. Add one drop of methyl red and titrate to yellow with NH_4OH . Back titrate to red with 2N HCl . Then add one drop extra of 2N HCl .
22. Electroplate for 20 minutes at 2.5 amps.
23. Send platinum discs to counting room for counting of $^{236, 8, 9}\text{Pu}$.

APPENDIX H

PRELIMINARY REPORT OF STUDIES WITH PARTICLES COLLECTED ON COPPER PLATES
FROM ABLATION OF $^{238}\text{PuO}_2$ FUEL PARTICLES NOS. 1-25-3 AND 1-25-20
UNDER CONDITIONS SIMULATING ATMOSPHERIC RE-ENTRY

5 May 1967

CONTRACT NO. 48-5713

Note to Appendix H:

- The Sandia designation for this report is SC-CR-67-2581.

I. INTRODUCTION

These particle size analyses were conducted as a continuing effort in support of Sandia's investigation of the thermal response and ablation behavior of plutonium dioxide microspheres.⁽¹⁾ The particulates being analyzed were generated in a hyperthermal arc tunnel where behavior studies were being conducted on single plutonium dioxide microspheres undergoing aerodynamic heating and ablation. The copper impactor plates were positioned in the arc tunnel to collect the plutonium ablation debris.

The immediate objectives of this program are to determine the size distribution and the total amount of plutonium on the collection plates. Also an effort will be made to determine the chemical nature of the plutonium in the particles.

II. DESCRIPTION OF SAMPLES

The plutonium particles being investigated were collected on special copper disc which are shown in Figure 1. A further description of the two collection plates analyzed is presented as Table I. Also described in this table are the single particulates of plutonium dioxide which were ablated under conditions simulating atmospheric re-entry and their fragments collected on the respective collection plates which were positioned downstream of the particles.

(1) Holland, W., Preliminary Report of Studies with Plutonium Particles on Copper Collection Plates from Ablation of a $\text{Pu}^{238}\text{O}_2$ Fuel Particle under Conditions Simulating Atmospheric Re-entry, Tracerlab, a Division of Laboratory for Electronics, Sandia Contract No. 48-5713, 24 March 1967.

Figure 1. Special Copper Collection Plate (1-25-3) in Lucite Holder for Shipping.

TABLE I. Description of Collection Plates Analyzed
and Pu O₂ Particles Ablated*

Impactor Number	Original Particle Diameter (μ)	Particle Weight (μ g)	Density (gm/cc)	Impactor Coating
1-25-3	234.4	69	10.3	PVC
1-25-20	253.0	87	10.3	PVC

The surfaces of both collection plates on the sides containing the alpha activity appeared only slightly affected by exposure to the plasma. There was a slight amount of charring and some decomposition of the polyvinylchloride (PVC) coatings which is evident from the radioautographs shown in Figures 2 and 3.

III. REMOVAL OF PARTICLES FROM COLLECTION PLATES 1-25-3 AND 1-25-20

Collection plates No. 1-25-3 and 1-25-20 were removed from the shipping container and the pieces of mylar which were in contact with them during shipment. Then the plates were monitored at a 2 cm distance by an Eberline Model PAC-3G portable alpha counter which, in each case, indicated a value greater than its maximum reading which is 10^5 cpm. Each collection plate was placed in a 400 ml beaker containing 100 ml of methyl ethyl ketone and scrubbed with a policeman. After the collection plates were removed, the pieces of mylar which also contained some plutonium particles were placed in the beakers and likewise washed. The wash solutions were utilized for further particle study. Even though it appeared that most of the coating originally present on the collection plates had been removed, monitoring the plates with the portable counter indicated the same reading as before. Figures 4 and 5 are radioautographs of the exposed plates which show the residual activity to be mainly on the edges of the plates.

The washed plates were covered with a layer of mylar and exposed to NTA nuclear emulsion slides. Optical examination of these slides after a 5 minute period indicated radioactive particles on the edges of the plates, no larger than those found in the washings. The residual plutonium on the plates will be removed when the total amounts originally collected is determined.

*The density of Pu O₂ is 11.46 according to the latest Chemical Rubber Handbook. The values of density listed in this table were calculated from the values determined experimentally for particle diameter and weight. The difference is possibly voids in the particles.

Figure 2. Autoradiograph of Impactor 1-25-3 (3X, 3 hr. exposure).

Figure 3. Autoradiograph of Impactor 1-25-20 (3X, 48 hr. exposure).

Figure 4. Radioautographs of the Collection Plates After Washing. The exposure times are shown.

The wash solutions were then transferred to centrifuge tubes and centrifuged for 30 minutes at approximately 2000 rpm. All of the supernate except for about 3 ml for each sample was removed. The two - 3 ml suspensions were sonic shaken for 10 minutes. For each of the samples, six drop portions of the suspensions were added to microscope slides which were designated as 1-25-3 A-1 to A-15 and 1-25-20 A-1 to A-10, respectively. After drying, these slides were ashed in a low temperature asher to remove the small particles of polymerized PVC which were not dissolved in the solvent. After a few drops of 4% collodion were added, each slide was placed in a special holder using 76 μ shims and exposed for 5 minutes to a NTA nuclear emulsion slide in order to observe the hollow stars produced by the plutonium containing particles. ⁽²⁾

IV. PARTICLE ANALYSIS

A. Radioactive Particles from Collection Plate 1-25-3

Optical Microscopy and Radioautography

After photographic processing, the NTA slides with their corresponding 1-25-3 slides were examined in their special holders by optical microscopy at 150 x. Each recognizable hollow star was scribed. The numbers of hollow stars found on each slide are recorded in Table II.

(2) Holland, W., Fourth Quarter of SNAP 9A Particle Studies during 15 April 1966 to 15 July 1966, Tracerlab, a Division of Laboratory for Electronics, HASL Contract No. AT(30-1) 3547 Mod. 3. pp.18-23, 14 September 1966.

TABLE II - Numbers of Hollow Stars Found in NTA Slides
Exposed for Five Minutes to Slides Containing
1-25-3 Plutonium Particles

<u>Slide Designation</u>	<u>Hollow Stars Found</u>	<u>Remarks</u>
A-1	27	The numbers of alpha tracks in the largest and smallest hollow stars were determined and found to indicate equivalent Pu ²³⁸ O ₂ particle diameters of 3.4μ and 0.56 μ, respectively
A-2	22	
A-3	20	
A-4	17	
A-5	5	
A-6	12	
A-7	19	
A-8	16	
A-9	13	
A-10	12	
A-11	15	
A-12	23	
A-13	5	
A-14	13	
A-15	8	
<u>227</u> total hollow stars		

As indicated in Table II the hollow star which contained the most alpha tracks and the star which contained the fewest tracks were counted and found to represent equivalent Pu²³⁸ O₂ particle diameters of 3.4μ and 0.56μ, respectively. Photomicrographs of the largest hollow star and the particle itself before and after isolation are shown in Figures 5 through 7. Note in Figure 6 the large amount of extraneous material present on the slides. As implied in Figure 5, the particle may be pure Pu²³⁸ O₂. Unfortunately, the particle was lost before it was counted under more accurate conditions or analyzed by an electron microprobe. The smallest hollow star from the 5 minute exposure of the 1-25-3 slides is shown in Figure 8. It was counted and found to represent an equivalent Pu²³⁸ O₂ diameter of 0.56μ. An attempt was made to isolate the particle which produced this star. The isolated particle and its hollow star are shown in Figure 9. The equivalent Pu²³⁸ O₂ diameter of the isolated particle is only 0.17μ. The isolated particles contained only approximately 3% of the plutonium contained in the original particle before isolation.

5 minute exposure

15 second exposure

Figure 5. Hollow stars produced by the "hottest" particle from 1-25-3 before isolation. The lower star indicated an equivalent Pu-²³⁸O₂ diameter of 3.4μ; however, this diameter likely should be much larger because absorption effects of the 30% collodion and self absorption in the particle were not considered.

In 4% collodion

After 30% collodion
added for isolation

Figure 6. Photomicrographs of "hottest" 1-25-3 particle before isolation. This particle produced the hollow stars shown in Figure 5. Note that another particle became attached to the radioactive particle after the 30% collodion was added.

15 second exposure

Figure 7. Photomicrograph of hollow star and "hottest" 1-25-3 particle after isolation. The hollow star indicated an equivalent $\text{Pu-}^{238}\text{O}_2$ diameter of 3.5μ . See the caption for Figure 5 for reason this value should be much larger. The particle diameter as measured from the lower picture is 65.7μ .

Figure 8. The smallest hollow star produced by 1-25-3 particles during a 5 minute exposure. It represents an equivalent $\text{Pu}^{238}\text{O}_2$ diameter of $0.56\ \mu$. Even though the parent particle was not recognizable beneath the center of this star, an attempt was made to isolate it.

Exposed for
1 hour

Figure 9. Photomicrographs of a radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 8. The equivalent Pu-²³⁸ O₂ diameter is only 0.17μ and the actual diameter of the particle is approximately 6μ.

B. Radioactive Particles from Collection Plate 1-25-20

Optical Microscopy and Radioautography

After photographic processing, the NTA slides which were exposed for 5 minutes to slides 1-25-20 A-1 through A-10 were examined by optical microscopy for hollow stars which were then scribed. The number of these stars found is recorded in Table III. After adding 30% collodion to slides A-1 through A-4, all the particles which produced the hollow stars during the five minute exposure were removed by our usual isolation technique. After low temperature ashing to remove the 30% collodion and adding more 4% collodion, the four slides were exposed to NTA nuclear emulsion slides for 15 minutes and scanned for hollow stars which were then scribed and enumerated. If the particles which produced the hollow stars during the five minute exposure had not been removed, they would have obscured many of the 15 minute hollow stars. The number of the 15 minute stars normalized to 10 slides is included in Table III. All the radioactive particles which produced the hollow stars during the 15 minute exposure of slide A-1 were isolated after a few drops of 30% collodion were added. The thick collodion was removed from A-1 by low temperature ashing and more 4% collodion added. This slide was exposed to an NTA slide for 30 minutes and the hollow stars produced were scribed and counted, the normalized results being listed in Table III. More 30% collodion was added to the slide and the radioactive particles which produced the hollow stars during the 30 minute exposure were removed from half the slide. After ashing and adding more 4% collodion, an exposure of 1 hour was made to a NTA slide. Optical microscopic examination of this NTA slide revealed a background of alpha tracks sufficient to obscure the hollow stars produced.

As shown in Figures 10 through 26, an attempt was made to isolate the radioactive particles producing the largest and smallest hollow stars for each of the exposure periods of 5, 15, and 30 minutes. Their equivalent $\text{Pu}^{238}\text{O}_2$ diameters are shown in Table III.

TABLE III - Numbers of Hollow Stars Found on NTA
Slides Exposed to Slides Containing 1-25-20
Plutonium Particles

Exposure Time (Min.)	Number of Hollow Stars Found	Equivalent Pu ²³⁸ O ₂ Diameters (μ)	
		Largest	Smallest
5	65	0.91	0.49
15	540	0.71	0.39
30	370	0.58	0.31

Figure 10. Photomicrograph of largest hollow star found in sample 1-25-20 during a 5 minute exposure. The magnification is 150 x, that is $100\ \mu$ is equivalent to 1.5 cm. Using a shim distance of $76\ \mu$, the equivalent Pu- 238 O $_2$ diameter for the particle producing this star was calculated to be $0.91\ \mu$.

30 minute exposure

Figure 11. Photomicrographs of a radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 10. The equivalent $\text{Pu-}^{238}\text{O}_2$ diameter is 0.39μ . The particle appears to have an actual diameter of approx. 25μ ; however, it is a flake with a thickness of approximately 5μ .

Figure 12. Photomicrograph of smallest hollow star from 1-25-20 during a 5 minute exposure. An equivalent $\text{Pu}^{238} \text{O}_2$ diameter of 0.49μ was indicated.

One hour
exposure

Figure 13. Photomicrographs of a radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 12. The equivalent $\text{Pu}^{238} \text{O}_2$ diameter is 0.18μ and the actual diameter approximately 3μ .

Figure 14. Photomicrograph of the largest hollow star from 1-25-20 during a 15 minute exposure. An equivalent $\text{Pu}^{238} \text{O}_2$ diameter of 0.71μ was indicated.

One hour
exposure

Figure 15. Photomicrographs of a radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 14. The equivalent $\text{Pu}^{238} \text{O}_2$ diameter is 0.46μ and the actual diameter, 6.6μ .

Figure 16. Photomicrograph of the smallest hollow star from 1-25-20 during a 15 minute exposure. An equivalent $\text{Pu}^{238} \text{O}_2$ diameter of 0.39μ was indicated.

One hour
exposure

Figure 17. Photomicrographs of a radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 16. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.26μ and the actual dimensions of the particle, $16\mu \times 6\mu$.

One hour
exposure

Figure 18. Photomicrographs of a second radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 17. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.19μ and the actual dimensions of the particle, $27\mu \times 15\mu$.

Figure 19. Photomicrograph of the largest hollow star from 1-25-20 during a 30 minute exposure. An equivalent $\text{Pu}^{238} \text{O}_2$ diameter of 0.58μ was indicated.

One hour
exposure

Figure 20. Photomicrographs of a radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 19. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.18μ and the actual diameter, approximately 7μ .

Figure 21. Photomicrographs of a second radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 19. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.15μ and the actual diameter, approximately 11μ .

One hour
exposure

Figure 22. Photomicrographs of a third radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 19. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.20μ and the actual diameter, approximately 21μ .

Figure 23. Photomicrograph of the smallest hollow star from 1-25-20 during a 30 minute exposure. An equivalent $\text{Pu}^{238} \text{O}_2$ diameter of 0.31μ was indicated.

One hour
exposure

Figure 24. Photomicrographs of a radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 23. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.18μ and the actual dimensions of the particle, approximately $1.5\mu \times 5\mu$.

One hour
exposure

Figure 25. Photomicrographs of a second radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 23. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.22μ and the actual diameter, approximately 3μ .

One hour
exposure

Figure 26. Photomicrographs of a third radioactive particle and its hollow star obtained in an attempt to isolate the particle which produced the hollow star shown in Figure 23. The equivalent $\text{Pu}^{238}\text{O}_2$ diameter is 0.18μ and the actual diameter, approximately 5μ .

Even though there is considerable overlapping of the equivalent $\text{Pu}^{238}\text{O}_2$ size ranges for the different exposure periods shown in Table III, it is evident that there is probably a numerical peak in the range from 0.7 to 0.39 μ . In order to obtain more detailed information regarding the size distribution based on equivalent $\text{Pu}^{238}\text{O}_2$ diameters, it is recommended that the alpha tracks in each hollow star be counted or estimated. It is planned to send the radioactive particles shown in Figures 9, 11, 15, and 22 to the Los Alamos Scientific Laboratory for qualitative electron microprobe analysis.

In Table IV is presented a comparison of the equivalent $\text{Pu}^{238}\text{O}_2$ quantities obtained before and after the radioactive particles were isolated. Unfortunately, the amount of extraneous particles on these slides prevents recognizing the parent particles before isolation. This background is illustrated in Figure 27.

TABLE IV - Comparison of Equivalent Pu²³⁸O₂ Volumes Found
Before and After Particle Isolations²

Hollow Star Designations	Equivalent Pu ²³⁸ O ₂ Volumes (μ ³)*		Number of Radioactive Fragments Isolated	% of Pu ²³⁸ O ₂ Recovered After Particles Isolated
	Before Isolation	After Isolation		
Largest in 5 min. expos.	0.39	5.66 x 10 ⁻²	2	14.4
Smallest " " "	6.09 x 10 ⁻²	2.86 x 10 ⁻³	1	4.7
Largest in 15 min. "	0.184	5.07 x 10 ⁻²	3	27.6
Smallest in " " "	3.15 x 10 ⁻²	3.08 x 10 ⁻²	8	97.8
Largest in 30 " "	0.101	9.12 x 10 ⁻³	3	9.0
Smallest in " " "	1.52 x 10 ⁻²	1.12 x 10 ⁻²	3	73.9

*See Appendix.

Figure 27. Typical background of alpha tracks produced by slide 1-25-3 A-1 which was exposed for one hour. The hollow stars were not sufficiently recognizable in this background to permit their being counted.

V. ESTIMATE OF TOTAL PLUTONIUM COLLECTED ON IMPACTOR PLATES

A. 1-25-3

The total plutonium collected on the impactor plate 1-25-3 is estimated by averaging the equivalent $\text{Pu}^{238}\text{O}_2$ volumes corresponding to the largest and smallest hollow stars and multiplying this value by the number of hollow stars found:

$$10.34 \mu^2 \times 227 = 2.35 \times 10^3 \mu^3$$

Dividing this value by the volume of the original 1-25-3 particle,

$$\frac{2.35 \times 10^3}{6.71 \times 10^6} = 3.5 \times 10^{-4}$$

reveals the fraction of the original plutonium dioxide collected. It is pointed out that we have not considered the contribution of the particles having equivalent $\text{Pu}^{238}\text{O}_2$ diameters smaller than 0.3μ which were collected.

As implied earlier, the 65.7μ particle from 1-25-3 may be entirely $\text{Pu}^{238}\text{O}_2$. Making this assumption and neglecting the contribution of other radioactive particles collected from this sample, comparison of the volume of the 65.7μ particle to the volume of particle 1-25-3 before ablation indicates

$$\frac{1.49 \times 10^5 \mu^3}{6.71 \times 10^6 \mu^3} = 2.2 \times 10^{-2}$$

B. 1-25-20

The total plutonium collected on impactor plate 1-25-20 is estimated by calculating the average equivalent $\text{Pu}^{238}\text{O}_2$ volumes corresponding to the largest and smallest hollow stars for each size range and multiplying these values by their respective number of hollow stars found on the slides. These results are added to estimate the total volume of plutonium dioxide collected on the impactor plate:

$$\begin{array}{rcl} 0.228\mu^3 \times 65 & = & 14.8\mu^3 \\ 0.109\mu^3 \times 540 & = & 59.0\mu^3 \\ 0.059\mu^3 \times 370 & = & 22.0\mu^3 \\ \hline & & 95.8\mu^3 \end{array}$$

Dividing this total by the original volume of the 1-25-20 particle indicates the fraction collected:

$$\frac{95.8 \mu^3}{8.48 \times 10^6 \mu^3} = 1.13 \times 10^{-5}$$

VI. CONCLUSIONS

A. For sample 1-25-3 the radioactive particles were found with equivalent $\text{Pu}^{238}\text{O}_2$ diameters from 3.4 to 0.56 μ and actual particle diameters from 65.7 to approximately 6 μ .

B. For sample 1-25-20 the radioactive particles had equivalent $\text{Pu}^{238}\text{O}_2$ diameters from 0.91 to 0.31 μ with a numerical peak between 0.71 and 0.39 μ . The actual particle diameters varied from approximately 20 μ to approximately 3 μ having no correlation to the equivalent $\text{Pu}^{238}\text{O}_2$ diameters.

C. During attempts to isolate radioactive particles from sample 1-25-20 several radioactive fragments were usually obtained. See Table IV. These fragments were either dispersed from initial clusters or produced when the original particles were crushed during isolation.

VII. RECOMMENDATIONS

A. To obtain more detailed information regarding the size distribution based on equivalent $\text{Pu}^{238}\text{O}_2$ diameters, it is recommended that the number of alpha tracks be counted or estimated.

B. To reduce the problem of nonradioactive particles on the original slides from obscuring the radioactive particles, the material on each slide will be dispersed to four slides after the large radioactive particles are removed. Then these slides will be exposed to NTA nuclear emulsion slides for a longer period.

APPENDIX
TABLE RELATING DIAMETERS OF SPHERES TO VOLUMES

D(μ)	V(μ^3)		
.010	5.24×10^{-7}	10	524
.020	4.19×10^{-6}	20	4.19×10^3
.030	1.41×10^{-5}	30	1.41×10^4
.040	3.35×10^{-5}	40	3.35×10^4
.050	6.54×10^{-5}	50	6.54×10^4
.060	1.13×10^{-4}	60	1.13×10^5
.070	1.80×10^{-4}	70	1.80×10^5
.080	2.68×10^{-4}	80	2.68×10^5
.090	3.82×10^{-4}	90	3.82×10^5
.100	5.24×10^{-4}	100	5.24×10^5
.200	4.19×10^{-3}		
.300	1.41×10^{-2}		
.400	3.35×10^{-2}		
.500	6.54×10^{-2}		
.600	.113		
.700	.180		
.800	.268		
.900	.382		
1.00	.524		
2.00	4.19		
3.00	14.1		
4.00	33.5		
5.00	65.4		
6.00	113		
7.00	180		
8.00	268		
9.00	382		

	0	1	2	3	4	5	6	7	8	9
10	5236	5395	5556	5721	5890	6061	6236	6414	6596	6781
11	6969	7161	7356	7555	7752	7962	8173	8386	8628	8824
12	9048	9276	9508	9744	9980	<u>1023</u>	1049	1073	1098	1124
13	1150	1177	1204	1232	1260	1288	1317	1346	1376	1407
14	1437	1468	1499	1531	1563	1596	1629	1665	1697	1732
15	1767	1803	1839	1875	1912	1950	1988	2026	2065	2104
16	2145	2185	2226	2268	2310	2352	2395	2439	2483	2527
17	2572	2618	2664	2711	2758	2806	2855	2903	2953	3003
18	3054	3105	3156	3209	3262	3315	3369	3424	3479	3535
19	3591	3648	3706	3764	3823	3882	3942	4003	4064	4126
20	4189	4252	4316	4380	4445	4511	4577	4644	4712	4780
21	4849	4919	4989	5060	5131	5203	5276	5350	5424	5499
22	5575	5651	5728	5806	5884	5964	6044	6124	6206	6288
23	6370	6454	6538	6623	6709	6795	6882	6965	7058	7148
24	7238	7326	7420	7513	7606	7700	7794	7890	7986	8083
25	8181	8299	8378	8479	8580	8681	8784	8887	8991	9097
26	9203	9309	9417	9524	9634	9744	9854	9966	<u>1008</u>	1019
27	1031	1042	1054	1065	1077	1089	1101	1113	1125	1137
28	1149	1162	1174	1187	1199	1213	1225	1238	1251	1264
29	1277	1290	1304	1317	1331	1344	1358	1372	1386	1400
30	1414	1428	1442	1457	1471	1486	1500	1515	1530	1545
31	1560	1575	1590	1604	1621	1637	1652	1668	1684	1700
32	1716	1732	1748	1764	1781	1798	1814	1831	1848	1865
33	1882	1899	1916	1933	1952	1968	1986	2004	2022	2040
34	2058	2076	2094	2113	2131	2150	2169	2188	2207	2226
35	2245	2264	2284	2303	2323	2342	2362	2382	2402	2423
36	2443	2463	2484	2504	2525	2546	2567	2588	2609	2631
37	2652	2674	2695	2717	2739	2761	2783	2806	2828	2850
38	2873	2896	2919	2942	2968	2988	3011	3035	3058	3082
39	3106	3130	3154	3178	3202	3227	3251	3276	3301	3326
40	3351	3376	3401	3427	3459	3478	3504	3530	3556	3582
41	3609	3635	3662	3688	3715	3742	3769	3797	3824	3851
42	3879	3907	3935	3963	3991	4019	4048	4076	4105	4134
43	4163	4192	4221	4246	4280	4310	4340	4370	4400	4230

	0	1	2	3	4	5	6	7	8	9
44	4460	4491	4521	4526	4583	4614	4645	4676	4708	4739
45	4771	4803	4835	4867	4872	4932	4965	4997	5030	5063
46	5096	5130	5163	5197	5231	5263	5298	5333	5367	5401
47	5436	5471	5506	5541	5576	5612	5647	5683	5719	5754
48	5790	5815	5863	5900	5937	5973	6010	6048	6085	6122
49	6160	6198	6236	6274	6312	6351	6389	6428	6467	6506
50	6545	6584	6624	6664	6701	6743	6783	6824	6864	6905
51	6945	6986	7028	7069	7110	7152	7193	7236	7277	7320
52	7362	7405	7447	7490	7533	7577	7620	7663	7707	7751
53	7795	7839	7884	7928	7973	8018	8063	8108	8153	8199
54	8245	8290	8337	8383	8429	8476	8523	8569	8617	8664
55	8711	8759	8807	8855	8903	8951	8999	9048	9097	9143
56	9195	9244	9294	9344	9394	9444	9492	9544	9595	9646
57	9697	9748	9799	9851	9902	9954	<u>1001</u>	1006	1011	1016
58	1022	1027	1032	1038	1043	1048	1054	1059	1064	1070
59	1075	1081	1086	1092	1097	1103	1108	1114	1120	1125
60	1131	1137	1142	1148	1154	1159	1165	1171	1177	1183
61	1188	1194	1200	1206	1212	1218	1224	1230	1236	1242
62	1248	1254	1260	1266	1272	1278	1284	1291	1297	1303
63	1309	1315	1322	1328	1334	1341	1347	1353	1360	1366
64	1373	1379	1385	1392	1398	1405	1412	1417	1425	1431
65	1438	1445	1451	1458	1465	1471	1478	1485	1492	1498
66	1505	1512	1519	1526	1533	1540	1547	1554	1561	1568
67	1575	1582	1589	1596	1603	1610	1617	1625	1632	1639
68	1646	1654	1661	1668	1676	1683	1690	1698	1705	1713
69	1720	1728	1735	1743	1750	1758	1765	1773	1781	1788
70	1796	1804	1811	1819	1827	1835	1843	1850	1858	1866
71	1874	1882	1890	1898	1906	1914	1922	1930	1938	1946
72	1954	1962	1971	1979	1987	1995	2004	2012	2020	2029
73	2037	2045	2054	2062	2071	2079	2087	2096	2105	2113
74	2122	2130	2139	2148	2156	2165	2174	2182	2191	2200
75	2209	2218	2227	2236	2244	2254	2262	2271	2280	2289
76	2298	2308	2317	2326	2335	2344	2353	2364	2372	2381
77	2390	2400	2409	2418	2428	2437	2447	2456	2466	2475
78	2485	2494	2504	2513	2523	2533	2542	2552	2562	2572
79	2582	2591	2601	2611	2621	2631	2641	2651	2661	2671
80	2681	2691	2701	2711	2721	2731	2742	2752	2762	2772

	0	1	2	3	4	5	6	7	8	9
81	2783	2793	2803	2814	2824	2834	2845	2855	2866	2876
82	2887	2897	2908	2919	2929	2940	2951	2961	2972	2983
83	2994	3005	3016	3026	3038	3048	3059	3070	3081	3092
84	3103	3114	3126	3137	3148	3159	3170	3182	3193	3204
85	3216	3227	3238	3250	3261	3273	3284	3296	3307	3319
86	3330	3342	3354	3365	3377	3389	3401	3412	3424	3436
87	3448	3460	3472	3484	3496	3508	3520	3532	3544	3560
88	3568	3580	3593	3605	3617	3629	3642	3654	3666	3679
89	3691	3704	3716	3728	3741	3754	3766	3779	3792	3804
90	3817	3830	3842	3844	3868	3881	3894	3907	3920	3933
91	3946	3959	3972	3988	3998	4011	4024	4037	4051	4064
92	4077	4090	4104	4117	4131	4144	4157	4171	4185	4198
93	4212	4225	4239	4252	4266	4280	4294	4307	4321	4335
94	4349	4363	4377	4391	4405	4419	4433	4447	4461	4475
95	4489	4503	4518	4527	4546	4560	4575	4589	4604	4618
96	4632	4647	4661	4676	4691	4705	4720	4735	4749	4764
97	4779	4794	4808	4823	4838	4853	4868	4883	4898	4913
98	4928	4943	4958	4973	4989	5004	5019	5034	5050	5065
99	5080	5096	5111	5127	5142	5158	5173	5189	5205	5220

APPENDIX I

PRELIMINARY REPORT OF STUDIES WITH PLUTONIUM PARTICLES ON COPPER
COLLECTION OF PLATES FROM ABLATION OF A $^{238}\text{PuO}_2$ FUEL PARTICLE
UNDER CONDITIONS SIMULATING ATMOSPHERIC RE-ENTRY

24 March 1967

CONTRACT NO. 48-5713

I.

INTRODUCTION

This particle size analysis is being conducted in support of Sandia's investigation of the thermal response and ablation behavior of plutonium dioxide microspheres. The particulates being analyzed were generated in a hyperthermal arc tunnel where behavior studies were being conducted on single plutonium dioxide microspheres undergoing aerodynamic heating and ablation. The copper impactor plates were positioned in the arc tunnel to collect the plutonium ablation debris.

The immediate objectives of this program are to determine the size distribution and the total amount of plutonium on the collection plates. Also an effort will be made to determine the chemical nature of the plutonium in the particles.

II.

DESCRIPTION OF SAMPLES

The plutonium particles being investigated were collected on copper discs which had $3/4$ inch diameters and 0.04 inch thicknesses with $1/8$ inch diameter holes at their centers. A further description of the two collection plates analyzed is presented as Table I. Also described in this table are the single particles of plutonium dioxide which were ablated under conditions simulating atmospheric re-entry and their fragments collected on the respective collection plates which were positioned downstream of the particles.

TABLE I. DESCRIPTION OF COLLECTION PLATES ANALYZED
AND Pu O₂ PARTICLES ABLATED*

Designation	Description of Collection Surface	PuO ₂ Particles		
		Dia. (μ)	Wt. (μg)	Density (g/cc)
1-25-6	Uncoated Copper	256.8	92	10.4
1-24-14	Dow Corning 269 Adhesive Coating	235.4	71	10.4

The surfaces of both collection plates on the sides containing the alpha activity appeared black as though coated with a thin film of dried automotive grease.

* The density of Pu O₂ is 11.46 according to the latest Chemical Rubber Handbook. The value of density listed in this table was calculated from the values determined experimentally for particle diameter and weight. The difference is attributed to voids in the particles.

III. DESCRIPTION OF WORK WITH RADIOACTIVE PARTICLES COLLECTED ON UNCOATED PLATE NO. 1-25-6

A. Removal of Particles from Collection Plate

Collection plate No. 1-25-6 was removed from the shipping container and the ~ 2 inch² piece of mylar which was in contact with it during shipment. It was monitored by an Eberline model PAC-3G portable alpha counter which indicated a value greater than its maximum reading which is 10⁵ cpm at 1 cm distance. The collection plate was placed in a 150 ml beaker containing 20 ml of 4% collodion in amyl acetate and scrubbed with a policeman. After the collection plate was removed, the ~ 2 inch piece of mylar which also contained some plutonium particles was placed in the beaker and likewise washed. The wash solution was retained for further particle study. The collection plate was attached to a microscope slide by applying a few drops of 50% collodion to the edge of the plate. Even though it appeared that ~ 3/4 of the dark

coating originally present on the collection plate had been removed, monitoring the plate with the portable counter indicated the same reading as before. This is interpreted as the original alpha activity on the slide being considerable larger than the maximum which the instrument could indicate. Also since the reading obtained from monitoring the washed collection plate is again greater than the maximum for the instrument, one can only crudely estimate the amount of plutonium removed.

A piece of mylar was placed over the collection plate and a Kodak NTA nuclear emulsion slide was placed in contact with it. The radioautograph is shown in Figure 1. Because such a large amount of plutonium remains on the collection plate, we will investigate it thoroughly.

B. Examination of Particles Washed from Collection Plate

1. Optical Microscopy and Radioautography

In order to view the radioactive particles which were removed from the uncoated collection plate No. 1-25-6, 2 drops of the wash solution were added to each of sixteen clean glass microscope slides which were numbered 1-25-6-A through 1-25-6-P.

Slides No. 1-25-6-B, C, D, and F were exposed for five minutes to Kodak NTA nuclear emulsion slides in an attempt to produce hollow stars⁽¹⁾ with alpha tracks. Numerous hollow stars such as the one shown in Figure 2 were observed when the NTA slides were examined with an optical microscope. This hollow star was found to be produced by a clear particle from slide No. 1-25-6-C. This particle is shown in Figure 3. Proper alignment of the NTA slides in their original positions over the slides containing the plutonium activity disclosed one or more irregular shaped particles

(1) Holland, W., Fourth Quarter of SNAP 9A Particle Studies during 15 April 1966 to 15 July 1966, Tracerlab, a Division of Laboratory for Electronics, HASL Contract No. AT(30-1) 3547 Mod. 3, pp 18-23, 14 September 1966.

Figure 1. Radioautograph produced by the washed uncoated collection plate which was placed in contact with this NTA slide for 5 minutes. The cleared "cross" may indicate the pattern of the original plutonium deposit or it may be due to insufficient scrubbing during washing.

Figure 2. Hollow star produced by the particle shown below during a 5 minute exposure. The streak on the right which continues across the top of the photomicrograph is a marking made with a dissecting needle to enable easy relocation of this star on the slide.

Figure 3. The clear particle on slide 1-25-6-C which produced the hollow star shown above is typical of those containing a majority of the alpha activity from the 1-25-6 collection plate.

of a clear colorless material beneath the centers of each of the large hollow stars. The hollow stars contain insufficient alpha tracks for plutonium to be considered as a major constituent in the clear particles although some is obviously present. See Table II. Further optical microscopic examination of these particles using polarized, dark field, phase contrast, and reflected illumination produced no additional information.

In order to isolate the particle shown in Figure 3, a few drops of 50% collodion were added, and the particle was transferred with a dissecting needle to a clean microscope slide. The isolated particle is shown in Figure 4. The original particle was very brittle as evidenced by the numerous fragments produced during isolation. The slide containing the isolated particle was exposed for 30 minutes to the oxygen plasma of a low temperature asher, but no changes occurred as shown by the photomicrographs of Figure 4. Likewise, solubility tests with decalin, freon, and prepsol produced no effect. The results of the solubility test with decalin are illustrated in Figure 5. Organic solvents were selected because it is suspected that the unknown matrix material containing the plutonium may be a silicone.

The unknown matrix material may originate from the Dow Corning Adhesive 269 which is used to insulate the needle holding the SNAP fuel particle upstream of the collection plate during the simulated atmospheric ablation. It is indicated that a portion of this adhesive which is a silicone was probably ablated and deposited with the plutonium on the collection plate. Also it would be expected that the intense heat might char and polymerize the silicone rendering it insoluble in the usual solvents for DC 269 adhesive.*

The other surfaces inside the reaction chamber are either metallic or ceramic and thus not likely candidates for the unknown matrix material.

* We have found that Dow Corning vacuum grease is affected in this manner by prolonged exposure to the oxygen plasma of a Low Temperature Asher.

Before Ashing

After Ashing in
LTA at $\sim 200^{\circ}\text{C}$

Figure 4. Fragments of the clear particle shown in Figure 3 which was isolated from slide No 1-25-6-C. These photomicrographs show ashing has no effect on the clear particles.

After decalin
evaporated

Figure 5. Photomicrograph showing the results of the solubility test to determine if the particle shown in Figure 4 is dissolved by decalin.

Table II. Alpha Activities for Pu-238 O₂ Particles of Various Diameters*

Diameter (μ)	DPM
100	2×10^8
50	2.5×10^7
20	1.6×10^6
10	2×10^5
5	2.5×10^4
2	1.6×10^3
1	200
0.5	25
0.2	1.6
0.1	0.2

*These values are not corrected for self-absorption which is significant for particles $>2\mu$ in diameter

In order to investigate further the size of the plutonium particles deposited on the collection plate, the "silicone" particles shown in Figure 5 (lower) were crushed with a dissecting needle, spread over a 1 cm^2 area, and covered with a few drops of 4% collodion. After the collodion dried, the slide was placed in a special holder with a Kodak NTA nuclear emulsion slide for 5 minutes. The NTA slide was processed and examined by optical microscopy. Several hollow stars were found. The largest hollow star shown in Figure 6 was produced by the particle shown in Figure 7. This hollow star was calculated to have been produced by an amount of plutonium (in the clear particle of Figure 7) equivalent to a Pu-238 O₂ particle with a diameter of 0.5μ and an alpha activity of $\sim 21 \text{ dpm}$.

Figure 6. Photomicrograph of the hollow star which has more alpha tracks than any other produced by fragments obtained by further crushing of the clear particles shown in Figure 5. This was a 5 minute exposure at a shim distance of 76μ .

Figure 7. This clear particle which was made by further crushing the particles shown in Figure 5. Its hollow star is shown above. The alpha activity shown here indicates an equivalent Pu-238 O_2 particle diameter of 0.5μ and is approx. 21 dpm.

Also alpha counted by the hollow star method were two metallic spheres which were isolated from slide No. 1-25-6-C and are shown in Figure 8. Note the larger particle had the smaller alpha activity.

These results are adequate for revealing information regarding typical radioactive particles washed from the plate; however, additional work is necessary to determine the maximum size of plutonium particles removed. Placing all of the wash solution (~ 20 ml) on microscope slides for examination as described previously would be both time consuming and expensive. A more practical approach is to concentrate the particles $> 1 \mu$ in diameter by centrifugation.

The remaining wash solution from the collection plate No. 1-25-6 was removed from its beaker which was washed with 2-10 ml portions of 4% collodion. These solutions were added to a 40 ml centrifuge cone which was centrifuged at $\sim 2,000$ rpm for 30 minutes. All of the supernate except for 1 ml was removed. The remaining 1 ml of collodion solution and suspended particles were placed on nine microscope slides.

After drying, these slides were placed in special holders and exposed for one minute to nuclear emulsion slides using 76μ shims. After the nuclear emulsion slides were processed, all were examined by optical microscopy along with the slides containing the particles. An effort was made to locate the largest hollow stars on the nuclear emulsion slides. Most of the hollow stars were formed by clear "silicone" particles which have been described previously. The largest hollow stars were produced by a few metallic particles. The largest star found on the slides is shown in Figures 9 and 10. This star which indicates an equivalent Pu-238 O_2 particle diameter of 3.5μ and 8,000 dpm is illustrated in Figure 11. Also found were similar metallic particles illustrated in Figures 12, 13, and 14 which had little or no alpha activity.

Figure 8. Photomicrographs of metallic spheres isolated from slide no. 1-25-6-C. The amounts of alpha activity were 3.3 and 617 dpm or equivalent Pu-238 O₂ diameters of 0.25 and 1.5μ for the particles in the upper and lower pictures, respectively.

Figure 9. Photomicrograph of hollow star produced by a 1 minute exposure using 76μ shims. This hollow star was produced by the particle shown in Figure 11 which was obtained from the centrifuged wash solution from collection plate no. 1-25-6.

Figure 10. Photomicrograph of hollow star shown in Figure 9. Also shown is the particle which produced it.

Transmitted
illumination
used

Reflected and
transmitted
illumination
used

Figure 11. Photomicrographs of the metallic particle from centrifuged wash solution from collection plate no. 1-25-6 which produced the hollow stars shown in Figures 9 and 10. Figure 9 indicated this particle contains 8000 dpm of alpha activity or an equivalent Pu-238 O₂ particle diameter of 3.5μ.

Figure 12. Photomicrograph of metallic particle obtained from centrifuged wash solution from collection plate no. 1-25-6. Also shown are the alpha tracks produced by this particle in a nuclear emulsion slide exposed for one minute using 76μ shims.

Figure 13. Photomicrograph of the metallic particle shown in Figure 12. Transmitted and reflected illumination was used to obtain this picture.

Figure 14. Photomicrographs of metallic particles obtained from the centrifuged wash solution from collection plate no. 1-25-6. Hollow star exposures of these particles failed to indicate alpha activity. Transmitted and reflected illumination was used in obtaining these photomicrographs.

2. Electron Microscopy

In order to view submicron pieces of the silicone material in the electron microscope and locate embedded particles of plutonium, the photoreversal technique was utilized.⁽²⁾ See Figure 15. Slides 1-25-6-B, G, H, I, and J were ashed in the Low Temperature Asher to remove the collodion. After the addition of 1% PVC (polyvinylchloride) and a thin film of Ilford L-4 nuclear emulsion, the slides were exposed for 2-1/2, 20, and 90 hours before processing. Although optical microscopy of the slide which was photoreversed after 2-1/2 hours showed no cleared areas, a few were found surrounding some of the larger particles on the slides photoreversed after 20 hours and longer. By means of a recently developed technique⁽³⁾, small portions of these films were transferred from the microscope slide to blank electron microscope grids. The results of electron microscopy of these grids are illustrated in Figure 16.

Slide No. 1-25-6-H was developed normally after a 90 hour exposure. This was done to determine more accurately the degree to which the alpha activity was recorded in the thin nuclear emulsion films. Optical microscopy of these films revealed numerous clear particles with alpha tracks, which are illustrated in Figure 17.

Portions of the normally developed films were examined by electron microscopy. The results are shown in Figures 18 and 19.

The $< 0.1\mu$ particles covering the surface of the silicone fragment shown in Figure 19 may be plutonium or possibly an artifact. We plan to decide this by determining the plutonium in a few of these "silicone" fragments using the

(2) Ibid. p.17

(3) Holland, W., Second Quarterly Report of SNAP-9A Studies During 15 January 1966 to 15 April 1966, Tracerlab, a Division of Laboratory of Electronics, HASL Contract No. AT (30-1) 3547 Mod. 3, p30, 23 May 1966.

Figure 15. Electron micrographs illustrating the results of photoreversal of nuclear weapon debris particles. The clear areas surrounding the radioactive particles which are designated by arrows are areas from which silver grains that had been exposed by nuclear radiation were selectively dissolved. Then the film was exposed to light and processed normally to develop the silver grains surrounding the clear areas.

Figure 16. Electron micrograph of photoreversed radioactive "silicone" fragment from wash solution of uncoated collection plate no. 1-25-6. The cleared portions of the figure indicate alpha radiation from the particle. The very small fibrous particles surrounding the "silicone" fragment are developed silver grains which were not exposed by alpha radiation.

Figure 17. Photomicrographs of normally processed alpha tracks in a thin nuclear emulsion film produced by plutonium contained in the clear particles.

Figure 18. Electron micrographs showing two radioactive "silicone" fragments (arrows no. 1) and alpha tracks (arrows no. 2) These were processed normally.

Figure 19. Electron micrograph taken at high magnification of a radioactive "silicone" fragment.

hollow star technique and comparing these values with those obtained by estimating the total amounts of small particles attached to the same individual silicone fragments as viewed in the electron microscope.

C. Examination of Particles Remaining on Collection Plate after Washing

The washed collection plate was examined by optical microscopy by means of reflected illumination. Several areas of the clear "silicone" material were observed. These areas are illustrated by Figure 20.

An NTA nuclear emulsion slide was placed in contact with the mylar covering the collection plate No. 1-25-6. After the NTA slide was exposed for 5 minutes and processed, it was examined with the plate by optical microscopy using reflected illumination. As illustrated in Figure 21, intense alpha activity was observed above the "silicone" deposits.

The five minute exposure discussed above produced hollow stars too concentrated to permit calculation of alpha activity equivalent to Pu-238 O₂ diameters. Figures 22, 23, 25, and 26 show the two largest hollow stars which were found on the slides after the one minute exposure. The metallic particles producing these stars illustrated in Figures 24 and 27 were calculated to have 94,000 and 68,000 dpm corresponding to equivalent Pu-238 O₂ particle diameters of 7.8 and 7.0μ, respectively.

IV. DESCRIPTION OF WORK WITH RADIOACTIVE PARTICLES
COLLECTED ON COATED PLATE NO. 1-24-14

A. Removal of Particles from Collection Plate

The plutonium and DC 269 adhesive coating were removed from coated collection plate No. 1-24-14 in the following manner: First the plate was monitored with the portable alpha counter which read $\sim 10^5$ cpm at 1 cm. Then it was placed in

Figure 20. Photomicrograph of washed collection plate no. 1-25-6 showing the "silicone" material and the clean copper surface.

Figure 21. Photomicrograph of alpha tracks produced during a 5 minute exposure by plutonium in "silicone" material remaining on the collection plate after washing.

Figure 22. Photomicrograph of the largest hollow star found after a one minute contact exposure of the washed collection plate no. 1-25-6. The dark black spot designated by an arrow was produced by the top of the particle being in contact with the nuclear emulsion. The particle producing this hollow star is shown in Figure 24.

Figure 23. Photomicrograph of a hollow star designated by arrow produced by one minute exposure using a 500 μ shim. This hollow star was also produced by the particle shown in Figure 24.

Figure 24. Photomicrograph of the metallic particle designated by the arrow which produced the hollow star shown in Figures 22 and 23. Figure 23 indicated this particle contains ~94,000 dpm of alpha activity which is equivalent to a Pu-238 O₂ particle diameter of 7.8μ.

Figure 25. Photomicrograph of the second largest hollow star found after a 1 minute contact exposure to the washed collection plate No.1-25-6. The dark black spot indicated by the arrow was produced by the top of the particle touching the nuclear emulsion. The particle producing this hollow star is shown in Figure 27.

Figure 26. Photomicrograph of hollow star indicated by arrow and produced by a one minute exposure using a 500μ shim. This hollow star was also produced by the particle shown in Figure 27.

Figure 27. Photomicrograph of the metallic particle indicated by the arrow which produced the hollow stars shown in Figures 25 and 26. Figure 26 indicated this particle contains 68,000 dpm of alpha activity which is equivalent to a Pu-238 O₂ particle 7.0μ in diameter.

40 ml of decalin and scrubbed with a policeman. Upon removal, the plate counted $\sim 5 \times 10^4$ cpm at 1 cm. The wash solution was agitated with a sonic shaker for 15 minutes. An attempt was made to filter the suspension through a 2.5 cm millipore filter with 0.05 μ pores. Even though the filter was under vacuum, it became clogged after only 1 or 2 ml passed. Probably this stoppage was due to charred or polymerized DC 269 adhesive which failed to dissolve.

B. Examination of Radioactive Particles

The filter was removed and an unsuccessful effort was made to view the particles collected thereon. A standard technique for optical microscopy using transmitted light of particles deposited on millipore filters is to clarify the filter by having acetone vapor come in contact with it on a glass microscope slide. However, no clearing occurred when the filter containing particles from a portion of the collection plate No. 1-24-14 wash solution was treated in this manner. Instead the filter remained white and curled away from the slide. It is believed this problem is caused by the silicone adhesive remaining on the filter. No additional work was performed with this filter.

Two drops of the wash solution were added to a microscope slide for optical examination as performed with the wash of the uncoated washer. After a few drops of 4% collodion were added, the slide was radioautographed using a Kodak NTA slide as described earlier. Beneath the centers of the hollow stars which were found were pieces of clear flaky material which appeared identical to those observed from the uncoated collection plate.

No effort was made for further identification of these particles because of their similarity to those investigated from the uncoated collection plate. An added problem for particle analysis was presented by the extra amount of dissolved silicone adhesive in the wash liquid.

A radioautograph presented as Figure 28 was made of the coated collection plate after washing. Note that much more alpha activity was removed from this collection plate than from the uncoated one. (See Figure 1).

V.

DISCUSSION OF RESULTS

Although alpha activity has been detected both in clear and metallic particles on the washed collection plates and in the wash solutions, no particles consisting of pure plutonium or plutonium compounds have been found. This is evident from the hollow star results.

The maximum alpha activity in a "point" source was found with approximately 94,000 dpm which would be equivalent to a Pu-238 O₂ particle having a 7.8 μ diameter. In this case, the alpha activity was associated with a metallic flake $\sim$ 70 μ in diameter. Larger plutonium particles could be sufficiently encased in the metallic or the clear particles such that their alpha activity would be completely absorbed. If such large particles were present, a few "exposed" particles should be detected by the hollow star technique. We believe that no plutonium particles $>8\mu$ are present. In fact, we predict that the maximum Pu-238 O₂ particle diameter may be $<0.5\mu$ and suggest that electron microprobe analysis be performed on the large metallic particles producing the largest hollow stars. This work should reveal any concentrations $>\sim 2\mu$ in diameter which are present.

VI.

CONCLUSIONS

- (1) The majority of the plutonium appears to be encapsulated in a "silicone" and metallic particles. Measuring the size distribution and alpha activity of these particles is possible, but would not be relative to the problem of interest.

Figure 28. Radioautograph produced by the washed coated collection plate. The NTA nuclear emulsion slide shown above was exposed to the washer for 5 minutes.

- (2) The maximum size of plutonium particles originally deposited on the plates is estimated as an equivalent Pu-238 O₂ diameter of $< 8\mu$.
- (3) The procedure for removing the plutonium from the collection plates and the hollow star and photoreversal techniques for particle counting and location appear quite suitable for performing the desired work provided appropriate samples are obtained.

VII.

RECOMMENDATIONS

- (1) Perform electron microprobe analysis of the largest metallic particles producing the largest hollow stars.
- (2) Make an ablation run with the silicone materials omitted from the system to determine if analyzable collection plates may be obtained.
- (3) Substitute a plastic which would form no inorganic residues for the DC 269 adhesive provided such is appropriate after recommendation (2) is performed.
- (4) Perform radiochemical analysis of the uncoated and coated collection plates with their wash solutions to determine the total amounts of plutonium collected on each collection plate.
- (5) Determine if the small 0.1μ particles on the surface of "silicone" particles as revealed by electron microscopy are plutonium by means of the hollow star technique which will determine the total plutonium in individual silicone particles. The results should be compared with the amounts of plutonium as estimated from the total volume of the 0.1μ particles on individual "silicone" particles.

DISTRIBUTION:

Standard Distribution TID-4500 (52nd Edition) - UC-36 (176)

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

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

U.S. Atomic Energy Commission
Space Nuclear Propulsion Office
Washington, D. C. 20545
Attn: R. S. Decker, Jr., Chief,
Safety 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 (1)
F. D. Anderson, Regulation (1)

U.S. Atomic Energy Commission
Division of Biology and Medicine
Washington, D.C. 20545
Attn: J. Z. Holland, Fallout Studies Branch (1)
H. D. Bruner, Asst. Director Medical
and Health Research (1)

U.S. Atomic Energy Commission
Space Nuclear Propulsion Office
Albuquerque Extension
Albuquerque Operations Office
P.O. Box 5400
Albuquerque, New Mexico 87115
Attn: H. P. Smith (1)

U.S. Atomic Energy Commission
Albuquerque Operations Office
P.O. Box 5400
Albuquerque, New Mexico 87115
Attn: S. A. Upson, Director,
Non-Nuclear Act. Division (1)
J. F. Burke, Director,
Operational Safety Division (1)

AEC Site Representative
National Aeronautics and Space Adm.
Manned Space Center
Houston, Texas 77058
Attn: W. C. Remini, Bldg 16, Code ZS-5 (1)

Donald W. Douglas Laboratories
P.O. Box 310
Richland, Washington 99352
Attn: S. P. Gydesen (1)

Douglas Aircraft Company
Missile and Space Systems Division
3000 Ocean Park Blvd.
Santa Monica, California
Attn: Sig Gronich, Advanced Space
Technology (1).

General Electric Company
Valley Forge Space Technology Center
P.O. Box 8555
Philadelphia, Pennsylvania 19101
Attn: S. M. Scala, Room M9539,
Space Sciences Lab (1)
G. W. Rivenbark, Isotope
Power Systems Operations (1)

Deputy I. G. for Insp. and Safety,
USAF
Kirtland Air Force Base,
New Mexico 87117
Attn: Col. D. C. Jameson (AFINSR)
(1)

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

Jet Propulsion Laboratory
California Institute of Technology
4800 Oak Grove Drive
Pasadena, California 91103
Attn: A. L. Klascius, Radiation
Health and Safety (1)

Los Alamos Scientific Laboratory
P.O. Box 1663
Los Alamos, New Mexico 87544
Attn: Dr. L. D. P. King (1)
Dr. Wright Langham (1)
C. F. Metz, CMB-1 (1)
F. W. Schonfeld, CMF-5 (1)
J. A. Leary, CMB-11 (1)

Monsanto Research Corporation
Mound Laboratory
P.O. Box 32
Miamisburg, Ohio 45342
Attn: G. R. Grove (1)

Thomas B. Kerr
Code RNS
National Aeronautics and Space
Administration
Washington, D. C. 20545

National Aeronautics and Space
Administration
Ames Research Center
Moffett Field, California
Attn: Glenn Goodwin (1)

U. S. Naval Radiological Defense
Laboratory
San Francisco, California 94135
Attn: S. Z. Mikhail, Nuclear
Systems Group (1)

DISTRIBUTION: (continued)

National Aeronautics and Space Administration
Goddard Space Flight Center
Glenn Dale Road
Greenbelt, Maryland 20771
Attn: A. W. Fihelly, Nimbus Project (1)

Naval Facilities Engineering Command
Department of the Navy, Code 042
Washington, D. C. 20390
Attn: Graham Hagey (1)

Space Nuclear Propulsion Office
Lewis Research Center
21000 Brookpark Road
Cleveland, Ohio 44135
Attn: L. Nichols (1)

TRW Systems
P.O. Box 287
Redondo Beach, California 90278
Attn: Dr. Donald Jortner (1)

Union Carbide Corporation
Nuclear Division
P.O. Box X
Oak Ridge, Tennessee 37831
Attn: R. A. Robinson, Isotope Development
Center, (1)
B. R. Fish, Health Physics Division (1)

U. S. Public Health Service
Nat. CTR for Radiological Health
1901 Chapman Avenue
Rockville, Maryland 20852
Attn: Nuclear Facilities Section (1)

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

D. B. Shuster, 1200
J. R. Banister, 5270
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 (3)
S. L. Jeffers, 9512
S. McAlees, Jr., 9513
J. D. Appel, 9514
J. D. Appel, 9514 (ARPIC) (2)
R. J. Everett, 9515
A. J. Clark, 9520
J. W. McKiernan, 9521
J. Jacobs, 9522
B. F. Hefley, 8232 (5)
B. R. Allen, 3421
C. H. Sproul, 3428-2 (10)
W. J. Wagoner, 3413
Attn: M. S. Goldstein

TRACERLAB (10)
A Division of Laboratory for Electronics, Inc.
2030 Wright Avenue
Richmond, California 94804
Attn: W. D. Holland

Weiner Associates, Inc.
P.O. Box 4797
Baltimore, Maryland 21211

BLANK