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IMPACT TESTS OF PLASMA FIRED
 $^{238}\text{PuO}_2$ MICROSPHERES

W. J. Dalby, 9512

AEROSPACE NUCLEAR SAFETY DEPARTMENT 9510

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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

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FIRED $^{238}\text{PuO}_2$ MICROSPHERES

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Sandia Laboratories, Albuquerque

July 1969

Approved by:

A. J. Clark, Jr., 9510

ABSTRACT

Several rigid capsules containing production grade plasma fired $^{238}\text{PuO}_2$ microspheres were impacted at velocities up to 456 feet per second against various targets. Particle size analyses of the impacted microspheres showed that from 0.01 to 13 weight percent was in the less than 10 micrometer range and from 0.01 to 6 weight percent was in the less than 3.5 micrometer range.

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SUMMARY

As a partial fulfillment of Sandia Laboratories responsibility to the AEC for the safety analyses of nuclear power supplies for aerospace applications, a series of tests was conducted to generate data concerning the degradation of fuel particle size resulting from the impact of a capsule containing plasma fired $^{238}\text{PuO}_2$ microspheres.

Fourteen capsules fabricated by Sandia Laboratories were fueled by Mound Laboratory (operated by Monsanto Research Corporation). The fueled capsules were returned to Sandia Laboratories, and 13 of them were heated to approximately 1000°F and impacted into a variety of targets (granite, sand, concrete, and graphite over granite and concrete) at velocities ranging from 100 to 460 feet per second. Mound opened the 14 capsules and performed a series of sieving operations to separate the fuel into several size ranges. For the various tests, Mound reported that from 0.01 to 13 percent of the fuel in each capsule was reduced to particles less than 10 micrometers in size. Mound prepared samples from the less than 20 and from the less than 10 micrometer fractions for additional particle size analyses by Tracerlab (a Division of Laboratory for Electronics, Inc.).

Tracerlab analyzed samples from six of the impacted capsules. Their data were combined with Mound data to calculate the percentage of the total inventory of the impacted capsule for each of the size fractions in these samples. These data indicated that from 0.01 to 6 percent of the fuel inventory of each of these six capsules was reduced to particles less than 3.5 micrometers in size.

ACKNOWLEDGMENTS

Mound Laboratory fueled the capsules, opened them after impact, and performed the particle size analyses down to 10 micrometers. Tracerlab, a Division of Laboratory for Electronics, Inc., performed additional particle size analyses in the smaller size ranges. The Mound work was performed under contract to the United States Government, and the Tracerlab work was performed under contracts to Mound and Sandia. The results were reported by Messrs. Richard E. Vallée and Paul H. Bonnell of Mound and Walter D. Holland of Tracerlab. These reports are included in the appendices of this report to provide maximum information on all of the microsphere impact tests.

Mr. B. S. Hill was the Sandia Laboratories project engineer for these tests. Mr. J. F. Hudson, Jr., assisted in the refinement of the analytical techniques. Test Projects Division, 7336, and Track and Gun Division, 7344, were responsible for test coordination and completion of the impact testing of the capsules. Health Physics Division, 3312, was responsible for the environmental health considerations at Sandia.

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IMPACT TESTS OF PLASMA FIRED $^{238}\text{PuO}_2$ MICROSPHERES

Introduction

A prerequisite for the application of radioisotope fuels to aerospace missions is that no portion of the world's population will be exposed to a biological hazard as the result of any possible sequence of events. For fuels emitting low penetrating alpha radiation, the hazard may be eliminated by providing complete containment under all circumstances. However, containment may be exceedingly difficult to accomplish and/or demonstrate for particularly severe environments or for fuels with long half-lives. An insoluble fuel form of controlled particle size to preclude hazards from ingestion and inhalation appeared to be a suitable alternative to absolute containment. Plasma fired $^{238}\text{PuO}_2$ microspheres produced to a controlled particle size range appeared to be a candidate to implement this alternative. Tests were deemed necessary to determine if the particle size of the fuel as manufactured would be altered by the impact of the encapsulated fuel. A capsule configuration was selected to provide a high degree of confidence of containment of the fuel upon impact and to minimize the fuel required per test (Figure 1).

Test Facility

The capsules were impacted in Sandia Laboratories' Area III using the 300-foot long monorail test track. The capsules were heated in a manually controlled oven (Figure 2) to a predetermined temperature. The capsules were removed from the oven and placed between the guide rails of the test track (Figure 3) and driven by a vertical member of a sled which traveled on an I-beam embedded in concrete. The sled was propelled by one or more rocket motors. The sled was braked by rigid plastic foam (Figure 4); the capsule continued in the guide rails to enter the partially evacuated plexiglass box through a small hole and impacted the target. Negative pressure was maintained within the box by a blower (Figure 5). An absolute filter was inserted between the box and the blower to entrap any fuel which might have escaped from the capsule if it had ruptured at impact.

Test Procedure

Capsule Impact

Inert -- To ensure that the capsule design was adequate, 13 capsules were filled with a fuel simulant, heated to 1200°F, and impacted end-on into a granite target at velocities up to approximately 650 feet per second. The impact velocities for these tests are given in Table I. The properties of the granite target are given in Figure 6.

The capsules were heated to 1200°F to achieve a capsule impact temperature of 1000°F. Approximately 45 seconds elapsed from removal of the capsule from the oven until rocket sled firing. This time interval was experimentally verified to result in the desired 1000°F capsule temperature at impact.¹ The test setup yielded impacts within 1 degree of the specified end-on impact attitude.² These preliminary impact test capsules also served to establish welding techniques and parameters and handling procedures for subsequent capsules containing $^{238}\text{PuO}_2$ microspheres.

Active -- Mound fueled 14 of the capsules, which had been fabricated by Sandia, with plasma fired $^{238}\text{PuO}_2$ microspheres from several different batches (Table II). The fueled capsules were shipped to Sandia and impacted into granite, concrete, Ottawa sand (Figure 7), graphite over granite (Figure 8), and graphite over concrete (Figure 9) targets. All of the capsules were at approximately 1000°F at impact. The impact velocities ranged from 102 to 456 feet per second. The initial impact test was conducted in January 1967, and the final test of this series was completed on August 29, 1968. Capsule 5 was subjected to the same thermal cycle as the other 13 capsules but it was not impacted. Immediately after impact the capsules were returned to Mound where they were opened and the fuel particle size distributions analyzed.

Analyses

Mound Laboratory -- The analytical techniques at Mound were refined throughout the program. However, they were generally as shown in Figures 10 and 11. Before the capsules were fueled, the microspheres were washed and screened into several size ranges from 44 to 210 micrometers, the weight percentages of the various fractions were determined, the size fractions were thoroughly remixed, and the capsules were fueled from this mixture. In addition, a sample was removed from this mixture for photographing³ (Appendix A).

After impact, Mound opened the capsules and separated the contents of each capsule into two fractions (>44 and <44 micrometers) by screening the dry material. The >44 micrometer fraction was screened dry into several size ranges, and the weight percentage of each fraction was determined. The <44 micrometer fraction was placed in ethyl alcohol, screened into three size ranges, and the weight percentage of each size fraction was determined.³ To obtain a more accurate mass balance, the analytical procedures were modified for Test 14 (capsule 15)⁴ (Appendix B).

Mound also prepared samples from the <20 micrometer fractions (Tests 7, 10, and 11) and from the <10 micrometer fractions (Tests 4, 5, and 14) for additional particle size distribution analyses by Tracerlab. The procedures for preparation of these samples were also modified during the program. However they were generally as follows.³ All particles <20 or 10 micrometers in size were contained in an ethyl alcohol wash in a large crystallizing dish. The solution was stirred with a medicine dropper, and one drop was placed on a slide. This procedure was repeated, using a clean dropper each time, until 10 slides were prepared from each sample. The slides were allowed to dry, and a thin film of methyl methacrylate solution was applied for permanent mounting. The methyl methacrylate solution was supplied by Tracerlab and applied according to their directions.

Tracerlab -- The Tracerlab analytical procedure is shown schematically in Figure 12. This procedure consisted of exposing the slides to nuclear emulsion plates for a minimum time to locate the larger particles. These particles were isolated or removed from the slides to prevent them from obscuring the smaller particles during the subsequent longer exposures. The smaller particles remaining on each slide were then washed from the slide using amyl acetate, and the suspension from each slide was then distributed evenly on four slides. A longer duration exposure was then used to locate smaller particles, and the entire procedure was repeated until the practical limit of the particle size which could be isolated in this manner was attained. The quantities of active material were determined by electronic counting, radiochemistry, and Tracerlab's hollow-star technique. A summary relating test numbers, capsule numbers, test dates, impact velocities, fuel identities, and targets is given in Table II.

Test Results

General

Significant proportions of plasma fired $^{238}\text{PuO}_2$ microspheres contained in small rigid capsules (Figure 1) fractured at impact to produce particle sizes which were smaller than those present at encapsulation.

Mound Analyses

The results of the sieve analyses by Mound are given in Table III. These data were obtained by telecons with Mound personnel, from Mound monthly reports, and from References 3 and 4 (included as Appendices A and B, respectively). Table IV relates the test number, the Tracerlab sample number, and the sieve fraction from which Mound prepared the sample for analyses by Tracerlab.

Tracerlab Analyses

The results of the analyses by Tracerlab of the samples furnished by Mound from six of the impacted capsules are shown in Table V. The total number of particles and the equivalent $^{238}\text{PuO}_2$ masses in grams for each of these six samples are included in Table IV. These results were

reported by Tracerlab in References 6, 7, 8, 9, and 10 (included as Appendices C, D, E, F, and G, respectively).

The results of the Tracerlab analyses of these samples from the six capsules were related to the total inventory of the capsule by multiplying the percentages reported by Tracerlab for a particular size range by the percentage of the total capsule inventory representing the sieve fraction from which Mound took the sample for analysis by Tracerlab. For example, note that for the sample from Test 7 the Tracerlab analysis determined that 1.4 weight percent was in the 3.60 to 5.30 micrometer size range (Table V). The sample from the capsule for Test 7 for Tracerlab analysis was taken from the <20 micrometer fraction by Mound (Table IV). Mound reported that the <20 micrometer fraction was 0.73 percent of the total inventory of the fuel in the capsule (Table III). Then, $1.4/100 \times 0.73/100 \times 100 = 0.0102$ weight percent of the total fuel inventory of the capsule for Test 7 was in the 3.60 to 5.30 micrometers particle size range. Corresponding values for all of the size ranges reported by Tracerlab for the six samples analyzed were calculated and are shown in Table VI. The cumulative totals are also included in Table VI.

Discussion

General

Mound also performed sieve analyses of two samples of fuel used to fill the capsules and the fuel from the capsule fueled for Test 8. The capsule for Test 8 was not impacted but was heated to the same temperature as the capsules which were impacted. These data are included in Table IV and are plotted as percent weight of particles less than or equal to diameter versus particle diameter in micrometers (Figure 13).

To permit a comparison of the fuel particle size distribution of fuel which had not been impacted to that from comparatively "soft" impacts, the data from the analyses of Tests 7 and 9 were superposed on Figure 13 to give Figure 14. No particularly significant aspect of this comparison is evident.

The particle size distributions reported by Mound (Table III) and the distributions resulting from the combination of the Tracerlab and Mound data (Table VI) were plotted as percent weight of particles less than or equal to diameter versus particle diameter in micrometers for the six tests for which samples were analyzed by Tracerlab (Figure 15).

The compressive strengths of the concrete targets were not determined; however, mixes and minimum curing times are usually specified to yield 5000 psi minimum compressive strengths. Specimens from concrete targets impacted in an earlier capsule impact program yielded 6600 to 7700 psi compressive strengths. The capsule penetrated the sand target (Test 9) approximately 36 inches. The 0.75-inch thick supertemp graphite over the granite target (Test 13) was broken into several pieces by the impact of the capsule (Figure 16). The concrete

block behind the 0.75-inch thick Hitco graphite sustained minor cratering (Figure 17), and the capsule penetrated the graphite (Figure 18).

Problem Areas

Several questions concerning these tests are discussed below.

Application and Interpretation of Test Results -- No analytical model exists for the determination of fines generated by the impact of a fueled capsule or for the extrapolation of the data from the small rigid capsule to a full scale flight configuration. The data obtained in this test series can only provide an experimental basis for an engineering judgment.

Breakup of the fuel particles resulting from the impact of a capsule may be attributed to either one or a combination of the following: (1) inertia loading from the length of the fuel column, (2) crushing by the deformation of the capsule, or (3) propagation through the fuel of the stress wave initiated by the impact of the capsule into the target. The impact of a rigid container which has a solid end cap end-on into a hard target may reasonably be expected to generate the most severe conditions for (1) and (3).

Mass Balance Discrepancy -- With the exception of the analyses for the fuel from the Test 14 capsule, for which special procedures were used, mass balance discrepancies were reported for the major portion of the Mound analyses. These discrepancies ranged from a 4 percent gain to a 13 percent loss. These reported discrepancies may be factored into the analyses in several ways:

1. Assume the reported values for the various size fractions of interest were correct as reported,
2. Assume the missing material was distributed in some manner throughout the various size fractions, and
3. Assume the missing material to consist entirely of the smallest size fraction reported.

Assumptions 1 and 3 establish the bounds, and this band for the six impact tests for which the Tracerlab evaluations were made is shown in Figure 19.

There are several possible explanations for the mass losses.

1. Entrapment of particles in the mesh of the sieves. This was observed during the sizing of glass microspheres for some impact tests conducted previously.
2. All of the fuel may not have been removed from the capsules. The fuel cavity was comparatively long and had a small diameter. In addition, the fuel was compacted by the higher velocity impacts (Figure 20) and was difficult to remove from the capsules.¹¹

3. The very fine fuel particles may have been dispersed during the initial sieving following the opening of the capsule after impact. This sieving was performed with the fuel dry for all except Test 14. The following excerpt was taken from Reference 11 and may indicate the release of very fine fuel particles upon the initial opening of the capsule.

The impact capsule* was opened in a fume hood on December 6, 1966. Some alpha activity was detected above the screw plug which was loose. When the plug was completely removed, there was a rapid increase in the airborne activity and the capsule was then placed in a glove box.

Tracerlab Samples -- For Tests 7, 10, and 11, both Mound and Tracerlab completed analyses from which the relative quantities of <10 and <20 micrometer size particles may be compared. For example, Mound reported that for Test 7 (Table III) 0.007 percent of the total capsule inventory was <10 micrometers versus 0.73 percent for that fraction <20 micrometers. Thus, approximately 1 percent of the <20 micrometer fraction was <10 micrometers. Tracerlab reported (Appendix C) that approximately 6 percent of the total sample furnished for their analyses was <10 micrometers and that more than 58 percent was >20 micrometers. The samples from Tests 7, 10, and 11 for Tracerlab analyses were all prepared from the <20 micrometer sieve fractions. The corresponding relationships for Tests 10 and 11 are as follows:
(1) Test 10: 30 percent of <20 micrometers fraction reported by Mound as <10 micrometers versus 89 percent reported by Tracerlab, and
(2) Test 11: 23 percent of <20 micrometers fraction reported by Mound as <10 micrometers versus 100 percent reported by Tracerlab.

No satisfactory explanation for this apparent discrepancy is known. Sampling techniques and sample handling may have been a factor. However, it should be noted that in all three samples Tracerlab reported a higher percentage of the <20 micrometers fraction to be <10 micrometers than did Mound.

Particle Size Measurements -- Three particle size measurements were employed. Mound used sieves and equivalent circular diameter, and Tracerlab used equivalent circular diameter and equivalent $^{238}\text{PuO}_2$ spherical diameter.

Mound separated the impacted fuel particles into several size ranges by a series of sievings. No tolerances were specified for the sieve openings. The following is from Reference 3.

Photomicrographs were taken of samples from the smaller sieve size ranges. A film negative was placed over a light source which was circular and could be varied continuously and the particle sizes were determined and the particles counted simultaneously using this apparatus (Carl Zeiss particle size counter). Since the particles in these size ranges arise from the break-up of larger spheres or from chips which break off larger spheres,

they cannot be measured with the same accuracy as spheres. The particles had many shapes, including rod-like. These rod-like particles may easily pass through a screen end-on and lie flat for photographing. With a circular light source and irregular shaped particles, personal discretion had to be exercised as to when the portion of the particle outside light circle was equal to the void inside the circle. Measurements could be made only in 1 plane; therefore the error of total particle size could vary from 20 to 100 percent.

Tracerlab analytical techniques determined equivalent spherical diameters. The larger particles of the samples furnished for Tracerlab analyses were isolated and numerically counted employing radiological and visual techniques. The activity of the various fractions isolated was determined. The equivalent spherical diameters were calculated using the density reported as typical for the fuel. For the smaller size ranges, each particle was isolated and the activity was measured using the Tracerlab hollow star technique. This method yielded equivalent spherical diameters by comparing the "tracks" produced on an emulsion-coated slide by particles from the sample undergoing evaluation with previously prepared "standards."

Tracerlab attempted to provide a correlation between equivalent circular diameter and equivalent spherical diameter data by isolating a series of particles from the sample from capsule 7 and performing both measurements. The values reported are shown in Table VII and plotted in Figure 21. These data were used to calculate the particle thicknesses plotted in Figure 22. Although the data are believed to be inadequate to permit a firm conclusion regarding the particle thickness, a trend is visible. However, an attempt to substantiate the trend by assuming a nominal 1-micrometer particle thickness and using the reported equivalent circular diameters of particles from a subsequent analysis to calculate quantities of fuel for comparison with the analytically determined quantities was unsuccessful.

Recommendations

An investigation to ascertain and refine techniques and equipment for particle size analysis of both radioactive and inert materials should be vigorously pursued.

A program to investigate the possibility of extrapolating from particle size distributions resulting from the impact of analog materials in full scale capsules to active materials in full scale capsules should be considered.

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9. Walter D. Holland, Particle Analysis of Radioactive Fragments with Diameters of Less Than 20μ From Impacted Sample 5, TLW-6026 (SC-CR-68-3749), Tracerlab, Sept. 17, 1968.
10. Letter, Walter Holland, Tracerlab, to Walter Dalby, Sandia, Analysis of Particulate Fragments in the Less Than 10μ Sieved Fraction of Sample 15, Ref. Contract No. 48-6716, TLW-12762, Oct. 23, 1968.
11. Neal Goldberg, SNAP-19/27 Fuel Impact and Vibration Test, Monsanto Research Corporation, Mound Laboratory, Dec. 29, 1966, (CRD).

TABLE I
Test Capsule Evaluation
Impact Velocity Data*

<u>Shot Number</u>	<u>Capsule Number</u>	<u>Impact Velocity (ft/sec)</u>	<u>Rebound Velocity (ft/sec)</u>
1	2	402.8	--
2	4	418.0	--
3	3	402.9	--
4	5	410.3	--
5	6	411.2	27.7
6	7	642.2	--
7	9	486.8	22.8
8	10	486.8	28.5
9	11	495.0	30.3
10	12	490.0	--
11	13	637.5	--
12	15	620.8	35.7
13	16	626.0	60.3
*All capsules heated to 1200°F and impacted into granite targets; see Figure 6 for properties of granite targets.			

TABLE II
Microsphere Impact Test Summary

Test Number	Capsule Number	Test Date	Impact Velocity (ft/sec)	Fuel ⁽²⁾	Target
1	(1)	1/67	377	(1)	Granite ⁽³⁾
2	(1)	1/67	247	(1)	Granite ⁽³⁾
3	(1)	5/17/67	107	(1)	Granite ⁽³⁾
4	20	6/28/67	102	MRL batch 116; produced 6/20/67.	Granite ⁽³⁾
5	18	6/28/67	106	MRL batch 116; produced 6/20/67.	Granite ⁽³⁾
6	24	6/28/67	106	MRL batch 116; produced 6/20/67.	Granite ⁽³⁾
7	4	10/20/67	116	MRL batch 120; produced 8/10/67.	Concrete ⁽⁴⁾
8	8		No impact	MRL batch 120; produced 8/10/67.	No impact
9	11	1/16/68	103	MRL batch 120; produced 8/10/67.	Ottawa sand ⁽⁵⁾
10	3	5/28/68	104	Aged and thermally cycled; produced 5/66.	Granite ⁽³⁾
11	5	5/28/68	103	Aged and thermally cycled; produced 5/66.	Granite ⁽³⁾
12	16	5/29/68	317	MRL batch 120; produced 8/10/67.	Granite ⁽³⁾
13	12	7/16/68	442	Aged and thermally cycled; produced 5/66.	0.75-inch thick Supertemp graphite over granite ⁽³⁾
14	15	8/29/68	456	MRL batch 120; produced 8/10/67.	0.75-inch thick Hitco graphite over concrete ⁽⁴⁾

(1) No information.

(2) Information obtained via telephone conversations, P. H. Bonnel, Mound/B. S. Hill, Sandia, and Reference 5.

(3) See Figure 6 for target properties.

(4) Nominally 5000 psi compressive strength concrete specified. Although this specific target was not evaluated, the results of the evaluation of concrete targets used for previous impact tests are shown in Table III.

(5) See Figure 7 for target description.

TABLE III
Weight Percentage Distribution of Fuel Particle Sizes
Sieve Analyses by Mound Laboratory

Particle Size (um)	Production				Test 1		Test 2		Test 3		Test 4		Test 5		Test 6		Test 7		Test 8		Test 9		Test 10		Test 11		Test 12		Test 13		Test 14	
	Sample A		Sample B																													
	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative	Reported	Cumulative
177 - 210	8.16	99.99	16.7	100.1							7.68	97.99	9.4	98.81	5.68	98.55	24.1	103.93	14.2	99.3	8.8	98.3	13.0	96.5	12.8	95.65	5.4	86.6	5.5	91.4		100.0
149 - 177	21.33	91.83	16.7	83.4							18.30	90.31	25.2	89.41	14.8	92.87	15.9	79.83	34.7	85.1	16.5	89.5	16.5	83.5	14.5	82.85	7.0	81.2	5.9	85.9		
125 - 149	16.33	70.61	20.0	66.7							15.85	72.01	18.1	64.21	12.1	78.07	18.7	63.93	21.9	50.4	22.1	73.0	17.8	67.0	17.8	68.35	7.1	74.2	7.7	80.0		
105 - 125	18.98	54.08	16.7	46.7							14.05	56.16	12.6	46.11	14.2	65.97	13.6	45.23	6.6	28.5	18.6	50.9	15.5	49.2	14.5	50.55	5.2	67.1	5.8	72.3		
> 105					28		33																									
<105					72		67																									
77 - 105	21.63	35.10									24.34	42.11	19.8	33.51	24.6	51.77	11.9	31.63	10.9	21.9	19.1	32.3										
63 - 105			25.0	30.0																			23.2	33.7	25.1	36.05	13.9	61.9	14.2	66.5		
44 - 77	13.47	13.47									16.90	17.77	10.4	13.71	23.1	27.17	14.4	19.73	11.0	11.0	13.2	13.2										
44 - 63			5.0	5.0																			7.5	10.5	7.2	10.95	8.5	48.0	5.4	52.3		
< 53					51		31																									
> 44																																
20 - 44								10.4			0.73	0.87	3.02	3.31	3.80	4.07	4.6	5.33					2.5	3.0	3.1	3.75	24.9	39.5	34.0	46.9	50.27 21.41	49.73
< 37					16		17																									
< 20	10 ⁻⁴																															
10 - 20											0.08	0.14	0.26	0.29	0.19	0.27	0.72	0.73					0.35	0.50	0.50	0.65	9.8	14.6	6.5	12.9	15.60	28.32
< 10							0.03				0.06	0.06	0.025	0.025	0.08	0.08	0.007	0.007					0.15	0.15	0.15	0.15	4.8	4.8	6.4	6.4	12.72	17.72
Discrepancy		0.01*		0.1**								2.01*		1.19*		1.45*		3.92**		0.7*		1.7*		3.5*		4.35*		13.4*		8.6*		0.00

Sample: A - Fuel from Mound batch 116; produced 6/20/67.
B - Sample from fuel used to fill capsules for tests Nos. 10, 11, and 13.

*Missing material
**Excess material.

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TABLE IV

Samples Furnished by Mound Laboratory
for Analyses by Tracerlab

<u>Test</u>	<u>Tracerlab Sample Reference*</u>	<u>Sample from MRL Sieve Fraction (μm)</u>	<u>Total Number of Particles in Sample</u>	<u>Equivalent $^{238}\text{PuO}_2$ Mass (10^{-8} gm)</u>
4	20	<10	9,165	5.6
5	18	<10	8,491	3.5
7	4	<20	950	7.3442
10	3	<20	396,958	22.8405
11	5	<20	261,214	16.38
14	15	<10	28,982	9.1725
*Capsule number, column 2, Table II.				

TABLE V

Numerical and Weight Percent Distribution of Fuel Particle Sizes
in Samples Furnished by Mound Laboratory for Analyses by Tracerlab

Particle Size* (μm)	Test 4		Test 5		Test 7		Test 10		Test 11		Test 14	
	Numerical	Weight	Numerical	Weight	Numerical	Weight	Numerical	Weight	Numerical	Weight	Numerical	Weight
10.80 - 11.60 7.60 - 10.90 7.50 - 10.80	0.01	8.5			0.5 0.5	58.4 31.4						
6.78 - 7.46 6.75 - 7.83 5.92 - 6.78 5.31 - 6.75							0.0025 0.0033	10.5 8.1			0.01 0.02	9.46 12.20
5.30 - 7.60 5.30 - 7.50 4.81 - 5.92 3.70 - 5.30	0.02 0.10	5.8 10.9			0.2	4.2					0.06	16.57
3.63 - 5.31 3.60 - 5.30 3.50 - 4.81 2.60 - 3.70	0.40	11.6	0.1	4.8	0.2	1.4	0.0088	7.6			0.08	11.79
2.60 - 3.60 2.34 - 3.63 2.24 - 3.50 1.80 - 2.60	1.1	11.1	1.1	16.6	0.8 1.2	1.9 0.91	0.022	4.8			0.22	8.85
1.59 - 2.29 1.30 - 2.34 1.30 - 2.24 1.30 - 1.80	6.3	22.0	5.5	28.9			0.091	3.2	0.12	4.3	1.75	9.41
1.20 - 1.80 0.88 - 1.59 0.88 - 1.30 0.87 - 1.30	13.1	15.5	17.8	31.5	2.7	0.73	0.6	8.1	0.92	9.5	2.29	5.62
0.87 - 1.20 0.61 - 0.88 0.61 - 0.87 0.60 - 0.87	21.7	8.7	17.5	10.5	3.2 10.2	0.28 0.32	2.95	12.7	4.50	17.8	5.93	4.56
0.42 - 0.61 0.42 - 0.60 0.30 - 0.42 0.21 - 0.30	37.6 18.4 1.3	5.1 0.8 0.02	28.7 26.3 2.9	5.8 1.8 0.37	19.2 33.9 26.2	0.20 0.12 0.032	8.24 6.82 14.51	11.7 3.3 2.5	10.41 17.40 29.89	13.6 7.8 4.7	11.90 12.59 16.01	3.17 1.12 0.53
0.19 - 0.21 0.14 - 0.21 0.10 - 0.14 < 0.10					1.3	0.00054	37.40 29.34	2.1 0.5 24.5	28.91 7.84	1.5 0.1 40.7	26.17 22.97	0.29 0.08 16.35
Σ	100.03	100.02	99.90	99.97	100.10	99.89	99.99	99.6	99.99	100.00	100.00	100.00

*Equivalent $^{238}\text{PuO}_2$ spherical diameter.

TABLE VI
Weight Percent Distribution of Fuel Particle Sizes
Tracerlab Analyses

Particle Size* (µm)	Test 4			Test 5			Test 7			Test 10			Test 11			Test 14		
	Tracerlab Sample	Total Fuel in Capsule		Tracerlab Sample	Total Fuel in Capsule		Tracerlab Sample	Total Fuel in Capsule		Tracerlab Sample	Total Fuel in Capsule		Tracerlab Sample	Total Fuel in Capsule		Tracerlab Sample	Total Fuel in Capsule	
			Cumulative			Cumulative			Cumulative			Cumulative			Cumulative			Cumulative
10.80 - 11.60 7.60 - 10.90 7.50 - 10.80 6.78 - 7.46	8.5	0.0051	0.0600				58.4 31.4	0.4246 0.2283	0.7262 0.3017							9.46	1.2033	12.72
6.75 - 7.83 5.92 - 6.78 5.31 - 6.75 5.30 - 7.60 5.30 - 7.50	5.8	0.0035	0.0549				4.2	0.0353	0.0734	10.5 8.1	0.0525 0.0405	0.4980 0.4455				12.20	1.5518	11.5167
4.81 - 5.92 3.70 - 5.30 3.63 - 5.31 3.60 - 5.30 3.50 - 4.81	10.9	0.0065	0.0514				1.4	0.0102	0.0428	7.6	0.038	0.405				16.57 11.79	2.1077 1.4997	9.9648 7.8571
2.60 - 3.70 2.60 - 3.60 2.34 - 3.63 2.24 - 3.50 1.80 - 2.60	11.6 11.1	0.0070 0.0067	0.0449 0.0379	4.8 16.6	0.0012 0.0042	0.0250 0.0238	1.9 0.91	0.01381 0.0066	0.0327 0.0188	4.8	0.024	0.367				8.85	1.1257	6.3575
1.59 - 2.29 1.30 - 2.34 1.30 - 2.24 1.30 - 1.80 1.20 - 1.80	22.0	0.0132	0.0313	28.9	0.0072	0.0196	0.73	0.0053	0.0122	3.2	0.016	0.343	4.3	0.0280	0.65	9.41	1.1970	5.2317
0.88 - 1.59 0.88 - 1.30 0.87 - 1.30 0.87 - 1.20 0.61 - 0.88	15.5	0.0093	0.0181	31.5	0.0079	0.0124	0.28	0.0020	0.0069	8.1 12.7	0.0405 0.0635	0.3270 0.2865	9.5 17.8	0.0618 0.1157	0.6221 0.5603	5.62 4.56	0.7149 0.5800	4.0348 3.3199
0.61 - 0.87 0.60 - 0.87 0.42 - 0.61 0.42 - 0.60 0.30 - 0.42	8.7 5.1 0.8	0.0052 0.0031 0.0005	0.0088 0.0036 0.0005	10.5 5.8 1.8	0.0026 0.0015 0.0005	0.0045 0.0019 0.0005	0.32 0.20 0.12	0.0023 0.0145 0.0009	0.0049 0.0026 0.0011	11.7 3.3	0.0585 0.0165	0.223 0.1645	13.6 7.8	0.0884 0.0507	0.4446 0.3562	3.17 1.12	0.4032 0.1425	2.7399 2.3366
0.21 - 0.30 0.19 - 0.21 0.14 - 0.21 0.10 - 0.14 < 0.10	0.02			0.07			0.032 0.0005	0.0002	0.0002	2.5 2.1 0.5 24.5	0.0125 0.0105 0.0025 0.1225	0.148 0.1355 0.125	4.7 1.5 0.1 40.7	0.0306 0.0098 0.0007 0.2646	0.3055 0.2750 0.2652	0.53 0.29 0.08 16.35	0.0674 0.0369 0.0102 2.0797	2.1942 2.1268 2.0899
Σ	100.02			99.97			99.89			99.6			100.00			100.00		

*Equivalent $^{238}\text{PuO}_2$ spherical diameter.

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TABLE VII

Equivalent Spherical $^{238}\text{PuO}_2$ and Circular
Diameters for Radioactive Particles Isolated
from Test No. 7--Analyses by Tracerlab

Equivalent Spherical $^{238}\text{PuO}_2$ Diameters (μm)	Equivalent Circular Diameters (μm)
10.6	30
3.14	5
1.4	2
11.2	26
11	26
11	28
10	26
10	28
4.8	8
5.4	12
3.6	5
3.9	6
6.6	13
1.5	3
3.5	5
1.4	2
2.5	3
1.1	1.1
1.1	1.1
10.7	28
11.5	28
8.6	21

Figure 1. Capsule configuration

Figure 2. Oven and rheostat used for heating capsules

Figure 3. Rocket sled and guide rail

Figure 4. Impact test setup

Figure 5. Target in plexiglass box with filter and blower

Specimen				Ultimate Load (lb)	Ultimate Strength (psi)	Maximum Deflection at Fracture (in.)	Modulus of Elasticity (psi x 10 ⁶)
Number	Size	Area	Length				
From Target A (SG 2.62)							
1	0.500 x 0.500	0.250	1.000	5350	21,400	0.0052	4.115
2	0.500 x 0.501	0.2505	1.000	4275	17,060	0.006	2.843
3	0.500 x 0.501	0.2505	1.000	4840	19,320	0.006	3.220
4	0.500 x 0.500	0.250	1.000	3720	14,880	0.007	2.126
5	0.499 x 0.500	0.2495	1.000	5100	20,440	0.0068	3.006
Average					18,620	0.0062	3.003
From Target B (SG 2.64)							
1-1	0.501 x 0.501	0.251	1.000	3600	14,340	0.0068	2.109
1-2	0.500 x 0.500	0.250	1.000	4700	18,800	0.007	2.686
2-1	0.500 x 0.501	0.2505	1.000	3580	14,290	0.0075	1.905
2-2	0.500 x 0.500	0.250	1.000	5000	20,000	0.0075	2.667
3-1	0.500 x 0.501	0.2505	1.000	5120	20,440	0.0073	2.800
3-2	0.500 x 0.501	0.2505	1.000	4700	18,760	0.0073	2.570
Average					17,770	0.0072	2.456

Figure 6. Typical granite target properties

Figure 7. Target for Test 9: open end wooden box filled with 950 pounds of 20-30 standard sand, ASTM designation C-190, from Ottawa Silica Company, Ottawa, Illinois

Figure 8. Target for Test 13, pretest

Figure 9. Target for Test 14, pretest

Figure 10. Mound Research Laboratory procedure before capsules were fueled (Reference 3)

Figure 11. Mound Laboratory procedure after impact (Reference 3)

Figure 12. Diagram showing microscope slides with sample X particles which were scanned for hollow stars; a total of $(N_1 + 10N_2 + 80N_3)$ normalized particle measurements was obtained; the slides designated by an asterisk were not scanned

Figure 13. Particle size distribution before impact

Figure 14. Particle size distribution before impact and "soft" target, low velocity impacts

Figure 15. Postimpact particle size distributions for tests for which Tracerlab analyses were made

Figure 16. Target for Test 13, posttest

Figure 17. Target for Test 14, posttest

Figure 18. Test 14 capsule and graphite, posttest

Figure 19. Postimpact range of particle size distributions for tests for which Tracerlab analyses were made

Figure 20. Test 1 capsule after impact

Figure 21. Equivalent circular diameter vs particle thickness for radioactive particles isolated from Test 7 by Tracerlab

$$\text{PARTICLE THICKNESS} = \frac{1/6\pi (\text{EQUIVALENT } ^{238}\text{PuO}_2 \text{ SPHERICAL DIAMETER})^3}{1/4\pi (\text{EQUIVALENT CIRCULAR DIAMETER})^2}$$

Figure 22. Equivalent circular diameter vs equivalent $^{238}\text{PuO}_2$ spherical diameter for radioactive particles isolated from Test No. 7 by Tracerlab

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APPENDIX A

P. H. Bonnell, Particle Sizing of Plutonium
Dioxide Microspheres After Impact, MLM-1499,
Dated Dec. 11, 1967, Issued June 21, 1968

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C O P Y

MLM-1499
TID-4500
UC-36
Aerospace Safety

**PARTICLE SIZING OF PLUTONIUM DIOXIDE
MICROSPHERES AFTER IMPACT**

P H Bonnell

Date December 11, 1967
Issued: June 21, 1968

MONSANTO RESEARCH CORPORATION

A Subsidiary of **Monsanto** Company

MOUND LABORATORY

Miamisburg, Ohio

operated for

UNITED STATES ATOMIC ENERGY COMMISSION

U S GOVERNMENT CONTRACT NO AT 33-1 GEN 53

ABSTRACT

Three capsules containing sized plutonium-238 dioxide microspheres were impacted onto a granite block. The fines (particles less than 44 μm in diameter) produced by damage to the micropsheres were sized, and the percentage in each size range was determined.

INTRODUCTION

In order to evaluate a fuel form for space applications in SNAP sources, it is important that the impact characteristics of the fuel be known. Microscopic particles of the radioactive fuel which could be released are a potentially serious health hazard. These particles could be released either through melting of the capsule (as could occur during launch pad explosion and/or fire) or fracture of the containers on impact of the capsule with the earth. Impact could occur after either suborbital or orbital abort of the vehicle. Therefore, a cooperative program was set up between Mound Laboratory and Sandia Corporation to study the impact characteristics of $^{238}\text{PuO}_2$ microspheres. Mound Laboratory was to load the fuel into impact capsules and characterize the fuel both before and after impact. Sandia was to impact the capsules at their facilities.

These microscopic particles, or fines, are present to some extent in production grade microspheres. In a previous study at Mound Laboratory the percentage of fines, but not particle size, removed from production grade microspheres was determined. In another study, particle sizes less than $10\text{ }\mu\text{m}$ were measured with an electron-microscope. A sample of fines was sent to Oak Ridge National Laboratory to be mounted for study. During storage and transportation of the sample, the fines settled out and agglomerated. No definite particle size distribution was obtained; however, a few definite particles could be seen (Figure 1).

Two capsules were previously impacted at Sandia at velocities of 377 and 248 fps (114.9 mps and 75.6 mps). The material in these capsules compacted to such a degree that measurements were not conclusive. The data that were obtained are listed in Table 1.

In studies covered by this report, all capsules were impacted end-on onto a granite block. It was felt that this mode of impact would produce the greatest number of fines (particles less than $44\text{ }\mu\text{m}$ in a loaded capsule. The capsules used in these fuel impact studies are shown in Figure 2.

Table 1

PARTICLE SIZE DISTRIBUTION

Particle Size (μm)	Percentage Before Impact (wt %)	Percentage After Impact (wt %) at 248 fps (75.6 mps)	Percentage After Impact (wt %) at 377 fps (114.9 mps)
>105	79.6	33	28
>53 <105	20.3	36	21
>37 <53	0.1	14	35
<37		17	16
<10		0.03

FIGURE 1 - ORNL photomicrograph of fines.

FIGURE 2 - Drawing of impact capsule.

PROCEDURE

All work was completed according to the following schedule:

A. Before the capsules were loaded

1. The microspheres were washed.
2. The microspheres were screened and each size range weighed. This gave the percentage in each range before impact.
3. The microspheres were thoroughly remixed. A sample was removed for a photomicrograph (Figure 3).

B. After impact

1. The capsules were opened and the contents separated into two fractions:
 - a. 44 - 200 μm
 - b. <44 μm .
2. The fraction above 44 μm was screened into the same ranges examined before impact. Each range was weighed.
3. Photomicrographs were taken of a sample from each range:

177 - 200 μm (Figure 4)	105 - 125 μm (Figure 7)
149 - 177 μm (Figure 5)	77 - 105 μm (Figure 8)
125 - 149 μm (Figure 6)	44 - 77 μm (Figure 9)
4. The fraction below 44 μm was placed in ethyl alcohol and screened into the following ranges:

20 - 44 μm
10 - 20 μm
<10 μm
5. Samples from the two ranges above 10 μm were photomicrographed (Figures 10 and 11) and the particles sized.
6. Total weight determination of the particles in these two ranges was made (includes samples on microscope slides).
7. Samples from the <10 μm range were mounted on slides for the particle size determination by Tracerlab.*
8. The weight of the particles remaining after slide samples were removed was determined. This procedure was followed for two of the impact capsules. For a total weight determination all particles <10 μm from the third capsule were dissolved and weighed.

This program gave both a complete size spectrum and the total percentage in each size range.

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Richmond, California.

FIGURE 3 - Composite of microspheres before impact.

FIGURE 4 - Composite of the 177 - 210 μm size range after impact.

FIGURE 5 - Composite of the 149 - 177 μm size range after impact.

FIGURE 6. Composite of the 125 - 149 μm size range after impact.

FIGURE 7 - Composite of the 105 - 125 μm size range after impact.

FIGURE 8 - Composite of the 74 - 105 μm size range after impact.

FIGURE 9 - Composite of the 44 - 74 μm size range after impact.

FIGURE 10 - Composite of the 20 - 44 μm size range after impact.

FIGURE 11 - Composite of the 10 - 20 μm size range after impact.

All microspheres received from production were washed by the following method:

The spheres were placed in a Petri dish and approximately 100 ml of ethyl alcohol added. After a few minutes of ultrasonic cleaning, the liquid was decanted. This process was repeated three or four times. After each wash, a drop of liquid was placed on a filter paper and dried. The filter paper was then counted using an Eberline PACLSA. Washing was stopped when the counting rates stabilized. Then 5 ml ethyl alcohol was added and the dish gently rocked back and forth by hand. This liquid was decanted and added to the previous supernatant liquid.

The spheres were then allowed to dry by self-heating, weighed, and placed in an impact capsule. The capsule cap was electron beam welded. The capsule was helium leak tested, radio-graphed to determine weld integrity and fuel level, and calorimetered. The capsules were then shipped to Sandia Corporation in Albuquerque, New Mexico, for impact into granite at approximately 105 fps (32 mps), the calculated impact velocity of an aborted space capsule. The capsules were impacted end-on, long axis perpendicular to face of target.

IMPACTED FUEL EXAMINATION

Upon receipt of the impacted capsules from Sandia Corporation, the end caps were cut off and the screw plug removed. The fuel was sieved into two sizes, those above 44 μm and those less than 44 μm . Those above 44 μm were sieved into the various size ranges and weighed. A sample was removed, placed on a microscope slide, and photomicrographed. Fines below 44 μm in size were placed in ethyl alcohol and sieved into three size ranges. Fines in the range 0 - 10 μm were analyzed by Tracerlab* for particle size.

The weight of the various sizes of fines was determined radiochemically. The alcohol containing fines of a particular range was evaporated to dryness and the particulate matter dissolved in a HNO_3 -HF mixture. The HF was evaporated and sufficient 2M or 8M HNO_3 was added to give 50 ml of solution. A 10- λ aliquot was pipetted onto a disk for counting. The weight was calculated from the counted results.

Photomicrographs were taken of a sample which was removed before the solution was taken for chemical analysis. This sample was placed on a microscope slide, dried, and placed in a special Lucite glovebox. A 40-power magnification was used for the 20 - 44 μm range and 100-power magnification for the 10 - 20 μm range. Several pictures were taken of each sample. A Carl Zeiss particle size counter was then used to simultaneously count and size the particles. Sizing was done by placing the film negative over a light source. This source is circular and can be varied in size continuously. Since the particles in these size ranges

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arise from the break-up of larger spheres or from chips which break off larger spheres, they cannot be measured with the same accuracy as spheres.

The particles have many shapes, including rod-like. These rod-like particles may easily pass through a screen end-on and lie flat when mounted. With a circular light source and irregular shaped particles, personal discretion had to be exercised as to when the portion of the particle outside the light circle is equal to the void inside the circle. Measurements can be made in only one plane; therefore, error of total particle size could vary from as little as 20% to as much as 100%.

By dividing the number of particles of each size by the total number counted, a numerical percentage was determined (Table 2). Sizes larger than 44 μm were determined by weight. Typical curves are shown in Figure 12. Figure 13 is a cumulative plot of the same data. Figure 14 is a cumulative plot curve of all size ranges.

Table 2
PARTICLE SIZES

Diameter Range (μm)	Distribution Before Impact (wt %)	Distribution After Impact (wt %)		
		in Cap. 18	in Cap. 20	in Cap. 24
177-210	8.16	9.4	7.7	5.7
149-177	21.22	25.2	18.3	14.8
125-149	16.53	18.1	15.9	12.1
105-125	18.98	12.6	14.1	14.2
77-105	21.63	19.8	24.3	24.6
44-77	13.47	10.4	16.9	23.1
20-44	0.00	3.02	0.73	3.8
10-20	0.00	0.26	0.08	0.19
<10	0.00	0.03	0.06	0.08
<u>Impact Velocity (fps)</u>		<u>Capsule Number</u>		
105.9 (32.2 mps)		18		
101.7 (31. mps)		20		
105.8 (32.2 mps)		24		

TRACERLAB WORK AND RESULTS

All particles less than 10 μm were contained in the alcohol wash in a large crystallizing dish. A medicine dropper was used to stir the solution; a sample was taken from about a half-depth of the solution and one drop placed on a slide. This procedure was used, using a clean dropper each time, until 10 slides were prepared. The slides were allowed to dry and a thin film of methyl methacrylate solution applied for permanent mounting. The methyl methacrylate solution was supplied by Tracerlab and applied according to their directions.

FIGURE 12 - Particles in Capsule 24 impacted at 32 mps.

FIGURE 13 - Cumulative particle size distribution.

FIGURE 14 - Cumulative particle size distribution of all size ranges.

The mounted slides were shipped to Tracerlab where the particles were counted and sized by the "Hollow Star" technique.¹ Table 3 gives the numerical frequency percentages for the two samples sent them. Figures 15 and 16 are numerical histograms of the two samples. Figures 17 and 18 are curves showing the cumulative plot of numerical count and equivalent plutonium dioxide particle mass as a function of particle diameter.

In Figure 19, the particle size was taken from the midpoint of the histogram ranges and plotted as a function of the percentage for that range.

Table 3

NUMERICAL FREQUENCY PERCENTAGES OF
EQUIVALENT $^{238}\text{PuO}_2$ PARTICLE MEASUREMENTS
OBTAINED FOR SAMPLES ANALYZED

Size Ranges (μm)	Numerical Frequency (%)	
	in Cap. 20	in Cap. 18
0.1 - 0.144	0	0
0.145 - 0.206	0	0
0.207 - 0.295	1.34	2.96
0.296 - 0.422	18.81	26.62
0.423 - 0.608	38.50	29.09
0.609 - 0.872	22.22	17.75
0.873 - 1.25	13.19	17.91
1.26 - 1.79	5.11	4.82
1.80 - 2.57	0.553	0.78
2.58 - 3.70	0.195	0.07
3.71 - 5.32	0.061	0
5.33 - 7.61	0.01	0
7.62 - 10.90	0.006	0

¹Special Particle Techniques, Brochure, Tracerlab.

FIGURE 15 - Numerical histogram of the 12,171 equivalent plutonium-238 dioxide particle measurements obtained from Mound sample No. 18.

FIGURE 16 - Numerical histogram of the 17,875 equivalent particle measurements obtained from the Mound sample No. 20.

FIGURE 17

Cumulative numerical count and equivalent plutonium-238 dioxide particle mass as a function of particle diameter for Mound sample No. 18. A total of 12,171 particles with a total mass of 4.49×10^{-8} g was obtained.

FIGURE 18

Cumulative numerical count and equivalent plutonium-238 dioxide particle mass as a function of particle diameter for Mound sample No. 20. A total of 17,873 particles with a total mass of 7.95×10^{-8} g was obtained.

FIGURE 19

Particle size as a function of percentage in that range (Tracerlab data).

DISCUSSION

Some plutonium-238 dioxide particles were reported as being in the range of 5- μ m equivalent size and 20- μ m microscopic size. Microprobe analysis by C. F. Metz at Los Alamos (Appendix) showed that the particle had a diameter of approximately 10 μ m of plutonium and the remainder was iron, chromium, and nickel. Evidently there are a few small stainless steel particles that are not removed after the capsule is cut open and the plutonium-238 dioxide fines become attached to them. This occurs more often with submicron particles than it does with larger ones. The microprobe analysis also showed that the plutonium was pure or that any contamination was below detectable limits.

No valid conclusions can be reached from the limited number of capsules that were impacted. While the Tracerlab data show very few or no particles in the range from approximately 4 μ m to 10 μ m, this is not actually true. Tracerlab particle sizes are reported as "equivalent spherical;" in this method all particles are assumed to be perfect spheres. Since the particles are actually chips or flakes, this method of reporting sizes will be off by a factor of roughly 2 to 2.5 at 4 μ m. This factor was determined by measuring particle size from photomicrographs and comparing this size to the "equivalent spherical" size reported. This difference factor is not constant but diminishes as the

particle size gets smaller. Mound particle sizes are reported as "equivalent circular," which is the same as that measured on photographs.

Particle weight can be calculated from "equivalent spherical" sizes, but not from "equivalent circular" sizes. This is due to the fact that "equivalent circular" size does not give a thickness to a particle. Therefore, circular size must be converted to spherical size to obtain a particle weight and spherical size must be converted to circular size to obtain the surface area of a flake or chip that may be in contact with lung tissue if inhaled. No particle size reported by either method is absolute but is in a range of approximately $\pm 30\%$.

The data also indicated that the percentage of fines produced in the 20 - 44 μm range was different in capsules impacted only 4 fps (1.2 mps) slower than others. Since all three capsules were impacted the same day and on the same machine, it is evident that very close velocity control is virtually impossible. Whether this difference in fines production is actual or only an anomaly would require further study.

Future Work:

A program is now underway to determine the fines production when capsules are impacted into targets other than granite. Since only one capsule will be impacted at each velocity into each target, only a general indication of fines production will be determined. Costs of materials prohibits any in-depth research at the present time.

ACKNOWLEDGMENTS

Grateful acknowledgment is made to Mr. Walter Holland of Tracerlab for his work and cooperation in sizing particles less than 10 μm ; to C. F. Metz, E. A. Hakkila, and H. L. Barker of Los Alamos Scientific Laboratory for their work on the microprobe analysis; and to R. E. Hermetz, R. L. Wise, and W. H. Johnson of Mound Laboratory for their work in sizing and photographing the particles.

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APPENDIX

ELECTRON MICROPROBE EXAMINATION
OF PARTICLES

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TO: Walter Holland, Tracerlab
Paul Bonnell, Mound
Ed Hansen, 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-Mound-PB-A-5-45, -46, -67, -74, -82

SAMPLE DESCRIPTION: The samples were described as plutonium-containing
particles mounted with collodion on bakelite.

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

ANALYTICAL RESULTS AND DISCUSSION:

A-5-45: The particle is approximately 10 microns in diameter (Figure 1a) and contains predominantly plutonium (Figure 1b), with trace amounts of iron (Figure 1c). Silicon, chromium, manganese, nickel, copper, silver, and tungsten were not detected. Detection limits are estimated to be in the range between 0.1 and 1 percent.

A-5-46: The particle is approximately 15 microns in diameter, and there is a lower density material to the left of the particle (Figure 2a). The high density particle contains predominantly plutonium (Figure 2b), and trace amounts of silicon (Figure 2e). The low density material contains predominantly iron (Figure 2c) and chromium (Figure 2d), with some plutonium (Figure 2b), silicon (Figure 2e), and nickel (Figure 2f). Manganese, 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.

A-5-67: The sample consists of several high density particles; the largest is triangular and is approximately 10 microns wide. A low density material (Figure 3a) partially surrounds the particles. The high density particles are predominantly plutonium (Figure 3b) and contain trace amounts of silicon (Figure 3g). The low density material contains predominantly iron (Figure 3c), some plutonium (Figure 3b), chromium (Figure 3d), and nickel (Figure 3e), and trace amounts of tungsten (Figure 3f) and silicon (Figure 3g). Manganese, copper, and silver were not detected in this sample. Detection limits are estimated to be in the range between 0.1 and 1 percent.

A-5-74: The particle is approximately 10 by 15 microns (Figure 4a) and contains only plutonium (Figure 4b). Silicon, manganese, chromium, iron, 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.

A-5-82: The sample consists of two large irregularly shaped particles and two small particles approximately 1 micron in diameter (Figure 5a). The large particles and the small particle near the center contain only plutonium (Figure 5b). The small particle in the lower right quadrant contains chromium (Figure 5c) and iron (Figure 5d). Silicon, 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 intensity distributions of the elements that were not detected were not meaningful and were not included.

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

6 microns

FIGURE 1a - Backscattered electron intensity distribution, Sample CMB-1-CFM-Mound-PB-A-5-45 (1500x). Particle is approximately 10 microns in diameter.

FIGURE 1b - Pu x-ray intensity distribution, same area. Pu 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.

FIGURE 2a - Backscattered electron intensity distribution, Sample CMB-1-CFM-Mound-PB-A-5-46 (1500x). Particle is approximately 15 microns in diameter with lower density area to left.

FIGURE 2b - Pu x-ray intensity distribution, same area. Note low Pu concentration in low density area. Pu x-ray intensity from plastic is equal to background.

FIGURE 2c - Fe x-ray intensity distribution, same area. Fe is only present in low density area. Fe x-ray intensity from high density particle and plastic is equal to background.

FIGURE 2d - Cr x-ray intensity distribution, same area. Cr is only present in low density area. Cr x-ray intensity from high density particle and plastic is equal to background.

FIGURE 2e - Si x-ray intensity distribution, same area. Si is present in low and high density areas. Si x-ray intensity from plastic is equal to background.

FIGURE 2f - Ni x-ray intensity distribution, same area. Ni is only present in low density area at left. Ni x-ray intensity from high density particle and plastic is equal to background.

6 microns

FIGURE 3a - Backscattered electron intensity distribution, Sample CMB-1-CFM-Mound-PB-A-5-67 (1500x). Largest high density particle near center is approximately 10 microns wide. Note low density material surrounding particles.

FIGURE 3b - Pu x-ray intensity distribution, same area. Note Pu in high density particles with lesser amount in low density material. Pu x-ray intensity from plastic is equal to background.

FIGURE 3c - Fe x-ray intensity distribution, same area. Fe is only present in low density material. Fe x-ray intensity from high density particles and plastic is equal to background.

FIGURE 3d - Cr x-ray intensity distribution, same area. Cr is only present in low density area. Cr x-ray intensity from high density particle and plastic is equal to background.

FIGURE 3e - Ni x-ray intensity distribution, same area. Ni is only present in low density area. Ni x-ray intensity from high density particle and plastic is equal to background.

FIGURE 3f - W x-ray intensity distribution, same area. W is only present as a trace constituent in the low density area immediately to the left of the large particle. W x-ray intensity from plastic is equal to background.

FIGURE 3g - Si x-ray intensity distribution, same area. Si is present as a trace constituent in the large high density particle and the low density material. Si x-ray intensity from plastic is equal to background.

6 microns
| |

FIGURE 4a - Backscattered electron intensity distribution, Sample CMB-1-CFM-Mound-PB-A-5-74 (1500x). High density particle is approximately 10 x 15 microns.

FIGURE 4b - Pu x-ray intensity distribution, same area. Pu x-ray intensity from plastic is equal to background.

6 microns

FIGURE 5a - Backscattered electron intensity distribution, Sample CMB-1-CFM-Mound-PB-A-5-82 (1500x). Note two large particles and two small particles approximately 1 micron in diameter.

FIGURE 5b - Pu x-ray intensity distribution, same area. Note Pu in large particles and in small particle near center. Pu x-ray intensity from plastic and other small particle is equal to background.

FIGURE 5c - Cr x-ray intensity distribution, same area. Cr is present only in small particle, lower right quadrant. Cr x-ray intensity from remainder of sample is equal to background.

FIGURE 5d - Fe x-ray intensity distribution, same area. Fe is present only in small particle, lower right quadrant. Fe x-ray intensity from remainder of sample is equal to background.

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APPENDIX B

Paul H. Bonnell and Richard E. Vallée,
Analysis of Fines From Impact Capsule #15,
Sept. 26, 1968, Monsanto Research Corporation,
Mound Laboratory, Miamisburg, Ohio

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C O P Y

ANALYSIS OF FINES FROM IMPACT CAPSULE #15

Paul H. Bonnell
Richard E. Vallée

MONSANTO RESEARCH CORPORATION

A SUBSIDIARY OF MONSANTO COMPANY

M O U N D L A B O R A T O R Y

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

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ANALYSIS OF FINES FROM IMPACT CAPSULE #15

Prepared by: Paul H. Bonnell

Approved by: Richard E. Vallée

September 26, 1968

Monsanto Research Corporation
Mound Laboratory
Miamisburg, Ohio

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ANALYSIS OF FINES FROM IMPACT CAPSULE #15

Capsule #15 was impacted by Sandia Corporation at 456.1 fps into a concrete target with a 3/4 inch thick piece of Hitco graphite taped over the target area; the fibers in the graphite ran parallel to the target face. Upon impact the capsule stuck in the graphite and knocked a shallow area out of the target.

As stated in the TWX, J. A. Powers to S. A. Upson (Reference 1, Page 5) dated September 11, 1968, the information from this impact test was urgently needed for inclusion in the GE SNAP 27 safety analysis report. It was also essential that every precaution be taken to obtain as accurate a material balance as possible within the time limit imposed.

While the entire program was conducted on a crash basis, every step was conducted with all care that would have been exercised during normal work. This fact is evidenced by the excellent material balance achieved; there was no gross material loss at any point and there was no gross contamination outside of working zones.

All procedures (Appendix A) were written on a cooperative basis between Mr. J. F. Hudson, Jr., Sandia Corporation and Mound Laboratory and agreed to by both parties. Due to the time limit imposed, some compromises with Sandia suggested procedures were necessary. It is not felt that these compromises affected the results within the errors of the measurements. During the analysis, various problems dictated deviations from the procedures dated September 11, 1968. These deviations were discussed before the fact and agreed to by both parties (Appendix B).

The capsule was opened by cutting off the electron beam welded end cap using a new Con-O-Saw in an uncontaminated glovebox. Two air samplers were set up inside the glovebox and a 4 hour background sample was taken before opening the capsule and a 24 hour sample was taken after opening the capsule. For all air sample and wipe results see the health physics procedures in Appendix C. The first cut was through solid metal and the second cut just touched the top of the screw cap. The cotton tip of a Q-Tip was used to wipe around the area between the screw cap and the capsule wall. This was bagged out and counted on a PAC-1SA hand monitor. The Q-Tip read 1500 cpm. The capsule was bagged out of the glovebox and transferred to a specially built plastic enclosure inside a fumehood.

A special made tool was used to remove the screw plug under ethyl alcohol. A pair of forceps were bent to hold the capsule while the fuel was removed. Some fuel poured out into the beaker of alcohol when the screw plug was removed. The remainder was removed by working the wooden portion of a Q-Tip into the fuel, with alcohol in the cavity, placing the end of the capsule under alcohol and tapping on the solid end with a pair of forceps. At first the fuel would come out very fast and then taper off to a thin stream and finally stop. The procedure would then be repeated. When experience and measurements indicated that all of the fuel had been removed, the inside of the capsule was flushed with alcohol using a 5 cc hypodermic syringe with a 20 gage needle. A clean wooden end of a Q-Tip was then worked along the sides and bottom of the fuel cavity. Visual examination of the Q-Tip showed no evidence of any fuel remaining.

The fuel was now in two beakers of alcohol. While this procedure was being carried out the screens to be used were washed with alcohol and air dried. They were then weighed on an analytical balance that had been calibrated by the standards and calibration laboratory just a few hours previously in the room it was being used in. The capsule was placed in a 100 ml graduate and bagged out for analysis by gamma scanning. The beakers containing the alcohol and fuel were then bagged out of the enclosure. The plastic bag used to transfer the capsule, forceps, Q-Tips and other tools used in opening and emptying the capsule were then bagged out of the enclosure and canned for analysis by gamma scanning. The plastic enclosure was then taken down, collapsed and canned for analysis by gamma scanning. The air samples from the plastic enclosure were removed for counting.

The entire inside of the fumehood was lined with plastic and an air sampler head was placed near the exhaust. A 44 μ m screen (44A) was fastened into a holder and placed on top of a clean 1000 ml pyrex beaker. The alcohol containing the fuel was transferred to this screen using a 25 ml Saebolt pipette. The pipette was rinsed periodically with clean alcohol to prevent any buildup of material. When one of the original beakers was empty a small amount of alcohol was added and a rubber policeman used to scrub the sides and bottom of the beaker. This was transferred to the 44 μ m screen by pouring and simultaneously washing with alcohol from a plastic squeeze bottle. This same procedure was used for the second beaker containing material. The screen, which now contained material $>44 \mu$ m plus some material $<44 \mu$ m, was placed in a 1000 ml beaker with sufficient alcohol to cover the material. This was placed, by hand, in an ultrasonic tank and agitated for 5 minutes. The screen was then removed from the beaker and washed with alcohol. Visible fines were evident. The screen was then placed in a crystallizing dish and again the material was just covered with alcohol. This was placed, by hand, in the ultrasonic tank and agitated for 6-1/2 minutes. Again fines were visible. These were transferred to a beaker by pouring and flushing with alcohol from a plastic squeeze bottle. This process was repeated five more times. At this point Mr. Hudson, Sandia, was afraid we might be breaking up large particles and suggested stopping the ultrasonic cleaning. The material was then washed with 100 ml of alcohol poured through the material and screen. This wash was sampled by placing one drop on a piece of filter paper, counting it on a PAC-1SA and calculating the counts per sphere. The equation used is

$$\frac{20 \times \text{volume of alcohol (ml)} \times \text{counts}}{\text{number of spheres}}$$

(assume number of spheres to be 80,000/g). We assumed that there were 3 g of spheres. This was done approximately six times with the counts per sphere varying between 400 and 1000. Mr. Hudson decided to cease washing.

The screen containing particles $>44 \mu$ m was placed in a pan and covered with a lid and set aside. The three beakers containing alcohol and fines were placed on a hot plate and the alcohol evaporated to a manageable level. The alcohol and fines were screened through a 20 μ m screen and the beakers cleaned in the same manner as before. The screen was placed in a crystallizing dish and ultrasonically agitated. This was

repeated several times and finally stopped because Mr. Hudson thought we might be breaking up particles. The screen was washed once more by pouring some alcohol over the material. Nothing visible came through the screen. This material, 20-44 μm range, was washed out of the screen, into a small plastic bottle, by using alcohol in a plastic squeeze bottle. The screen was set aside to air dry for later weighing. The plastic bottle was placed in a metal can with a friction lid and transferred to analytical. The fines were in the plastic bottle for no more than 15 minutes. More alcohol was evaporated and then put through the 10 μm screen and washed in exactly the same way as the 20 μm screen. The beakers were cleaned in the same manner as before. The alcohol containing the <10 μm material was evaporated to approximately 900 ml and the sample taken for the Tracerlab analysis. This sample consisted of placing one drop of the well stirred alcohol on a microscope slide that had been supplied by Tracerlab. This was repeated for ten slides. The procedure used has been used for all previous capsules and is the same as Tracerlab uses for mounting all Sandia samples they do. The alcohol was then evaporated to approximately 50 ml and transferred to analytical personnel in the same manner as the other samples.

After the beakers and other equipment used for one size range were empty or finished with they were placed in plastic bags, marked with the size range and set aside for analysis by gamma scanning. After the <10 μm material and equipment was removed from the fumehood the ultrasonic tank was bagged and removed to make room for the balance.

The weighed screens were stacked and the >44 μm material transferred from screen 44A to the 177 μm screen. To remove as much material as possible an artists brush was used to clean the screen. The stack with the pan on the bottom and the lid on top was shaken by hand to separate the material into the various size ranges. The 44A screen and pan were weighed together prior to placing the material in the stack. After the screening was complete, each screen, with material, was weighed. The material was then transferred to preweighed primary containers. The screens were weighed to determine the amount of material held up in the openings. The primary containers and material were also weighed. After all screens were emptied the pan was weighed to determine the amount of fines that had come off the >44 μm particles during screening.

After the last step was completed the plastic lining in the fumehood was removed, canned up and gamma scanned.

The material sent to analytical was transferred to 250 ml Teflon beakers and boiled to near dryness. About 25 ml of concentrated HNO_3 was added to the sample and the sample boiled for a few minutes to decompose any remaining organic material which might have been present. A few drops of HF were then added and the sample boiled until dissolution was complete. After the dissolution was complete it was diluted 50 to 1 and counted by liquid scintillation counting. This method is accurate to $\pm 1\%$.

Gamma scan results have an accuracy of $\pm 20\%$. Balance weights were accurate to $\pm 0.5\%$. The gamma scan results were prorated across the range they covered based on the weight of material in the respective size range. For example, the material in the 0 - >177 μm range was prorated for all size ranges while material in the 0 - 20 μm range was prorated for the two smallest ranges only.

Table I gives the weight distribution for all size ranges and source of weights. Based on the initial sample weight, there was a total of 6.088 grams of oxide to be accounted for. All weights were converted to oxide weight before determining totals and calculating percentages. The final calorimeter value (2.4375 watts) and the initial weight of fuel (6.088 grams) were used to calculate the conversion factor for calculating the oxide weight for the samples assayed by scintillation counting. As Table I shows a total of 6.1312 g were accounted for.

Table I
DISTRIBUTION OF FUEL FROM
CAPSULE #15 BY PARTICLE SIZE

<u>Size Range</u> <u>(μm)</u>	<u>Source</u>	<u>Amount</u> <u>(g)</u>	<u>Total</u> <u>(g)</u>	<u>% of</u> <u>Total</u>
<u>>44μm</u>	>177	retained on 177 μ screen	0.3824 ¹	
	149-177	retained on 149 μ screen	0.3998 ¹	
	125-149	retained on 125 μ screen	0.5438 ¹	
	105-125	retained on 105 μ screen	0.3486 ¹	
	74-105	retained on 74 μ screen	0.5994 ¹	
	44-74	retained on 44B μ screen	0.7013 ¹	
	44-74	retained on 44A μ screen	<u>0.0532¹</u>	
	(see Table II)	0.0538	3.0823	50.27%
<u>20-44μm</u>	removed from 20 μ m screen	1.2681 ²		
	retained on 20 μ m screen	<u>0.0153¹</u>		
	(see Table II)	0.0294	1.3128	21.41%
<u>10-20μm</u>	removed from 10 μ m screen	0.9022 ²		
	retained on 10 μ m screen	<u>0.0203¹</u>		
	(see Table II)	0.0338	0.9563	15.60%
<u><10μm</u>	passed thru 10 μ m screen	0.7268 ²		
	γ scan	0.0253		
	air samples	<u>0.0003</u>		
	(see Table II)	0.0277	<u>0.7798</u>	<u>12.72%</u>
			6.1312	100.00%
<u>Summary of Table I</u>				
Total accounted for by discrete size ranges			5.9865g	97.64%
Total prorated (see Table II)			<u>0.1447g</u>	<u>2.36%</u>
			6.1312	100.00%

¹ by weight

² by scintillation counting

Table II gives the amount of material prorated to each size range from samples containing material from mixed ranges. The vertical columns give the total added to each size range.

Table II
FUEL FROM CAPSULE #15 IN MIXED RANGES

Size Range of Fuel (μm)	Proration by Size Range				Total
	$>44 \mu\text{m}$	20-44 μm	10-20 μm	$<10 \mu\text{m}$	
0 - >177	0.0538g	0.0228g	0.0164g	0.0134g	0.1064g ^a
0 - 44	--	0.0066g	0.0048g	0.0039g	0.0153g ^b
0 - 20	--	--	0.0126g	0.0104g	0.0230g ^c
	0.0538g	0.0294g	0.0338g	0.0277g	0.1447g

^a0.0044 of this total was estimated from wipes, remainder by γ scan.

^bBy weight in pan after screening $>44 \mu$ sizes.

^cBy γ scan.

APPENDIX A

PROCEDURES FOR OPENING IMPACT CAPSULE #15 (456.1 fps)

September 11, 1968

- Item 1 - The thermal output of the capsule will be determined calorimetrically after impact. This will yield a third "bench mark" (initial weight and calorimetric determination).
- Item 2 - Welded end cap of capsule will be cut off using a new Con-O-Saw in an uncontaminated glovebox in room R-127. The walls of this glovebox will be mapped for alpha to determine background activity. A one (1) hour (minimum) air sample will be taken inside the glovebox prior to opening the capsule. A sketch of the relative position of the air sampler heads will be provided. It will be assumed that the gloveboxes are exhausted at their specified rating. If significant release of material was indicated to have occurred during end cap cut off, the box exhaust flow will be measured.
- Item 3 - Cut off the welded end of the capsule above the interior screw cap with the "Con-O-Saw" and clean the capsule of metal scraps and cutting debris with a Q-Tip. Monitor and record the CPM for the Q-Tip and the immediate work area. Wipes will be taken on inside of glovebox after cutting off end cap. If wipes show activity, the walls will be remapped for alpha. The membrane filters will be changed and a twenty-four (24) hour air sample taken. This will include the period the capsule is being cut open. All air sample counts will be recorded and the filters saved. The CPM will be converted to weight of plutonium present. If the quantity indicated to have been lost is greater than 0.001% of the total plutonium in the capsule, the filters will be sent to TracerLab for analysis.
- Item 4 - The capsule, with the screw cap in place will be placed in a plastic bag and transferred to a plastic enclosure inside a fumehood in room R-143. A background count will be taken in the plastic enclosure by air samples and swipes prior to the transfer. There are to be air sampler heads in the plastic enclosure and at the fumehood exhaust. The fumehood will be lined with plastic.

Item 5 - While holding the capsule with the hand, place the capsule in a beaker (all beakers used in this procedure should be pyrex glass) so that at least the screw cap and cut end of the capsule are below the liquid level in the beaker (liquid in the beaker should be absolute alcohol). The gloves are not to come in contact with the liquid. Loosen the screw cap and remove it with an appropriate tool, keeping the open end of the capsule below the surface of the liquid. Rinse the screw cap and copper gasket with alcohol and remove the copper gasket from the screw cap. Place the screw cap in a beaker containing HNO_3 -HF and dip the copper gasket in this solution only long enough to etch the surface.

NOTE: All tools, etc., used during the experiment are to be kept on a piece of absorbent paper in the glovebox.

Item 6 - Alcohol will be squirted into the fuel cavity to remove any loose material. Compacted fuel will be removed with the wooden end of a Q-Tip and flushed with alcohol from a squeeze bottle.

A. Using a 5 CC syringe and a hypodermic needle the interior of the capsule will be flushed with alcohol. This will be repeated 3 or 4 times. The capsule will be allowed to dry and then placed in the beaker of HNO_3 -HF with the screw cap and heated for 3 hours or until no apparent capsule contamination is observed.

Item 7 - The beaker containing the material will then be removed from the plastic enclosure. The air sampler counts will be taken and recorded and the membrane filters marked and stored. The interior of the plastic enclosure will be monitored for alpha activity.

Item 8 - The plastic enclosure will be removed from the fumehood. New filters will be placed in the air samplers of the plastic enclosure. The fumehood walls will be mapped for alpha activity with a "PAC" unit.

Item 9 - Weigh the cleaned 44μ screen (constant weight?) to within ± 1 mg prior to the screening operation (record weight). Place the weighed screen in a dish containing a small volume of alcohol. Carefully transfer the fuel from the beaker onto the screen using standard analytical procedures. (When the dish becomes partially full, stop the transfer, remove the screen and dish (together) from the ultrasonic tank.

Place the dish and screen next to a clean beaker, lift the screen carefully and hold it over the beaker so that any drops will fall into the new beaker. While holding the screen above the beaker, carefully transfer the contents of the dish into the same beaker. Continue the transfer of the fuel from the original beaker until the original beaker appears clean by visual examination. Then rinse the beaker at least four times, pouring the wash over the contents of the screen. Swipe and count the interior surface of the beaker after the third and fourth rinse; if the count changes, continue the rinse procedure until the count (CPM) stabilizes or becomes negligible.

- Item 10 - When the transfer of the fuel from the original beaker to the screen has been completed, it is then necessary to wash the screen and its contents. Wash the screen and its contents with fresh fluid (this wash and rinse operation should continue until two successive ~25 CC washes of all the material yield a stabilized level of contamination not greater than 1×10^6 CPM total). Allow the screen and contents to completely dry and weigh. Record the total weight.
- Item 11 - Set up a screen stack to separate the particulate larger than 44μ into specified size range. Weigh and record the weight of each screen in the stack, including the terminal pan. Separate the greater than 44μ portion using the present procedure except that the terminal pan shall be rinsed and washed. The wash fluid shall be added to the less than 44μ portion.
- Item 12 - Weigh and record the weight of the 10μ m and 20μ m screens to ± 1 mg. Separate the less than 44μ m portion using present procedures. Check frequently for clogged screens. Wash and rinse the beakers several times screening the rinse fluid through the 10 and 20μ m screen. Swipe and count the inner surfaces of the beakers and record CPM values.
- Item 13 - Prepare samples for TracerLab analysis per agreement between Walt Holland and Paul Bonnell.
- Item 14 - Prepare the residual from Item 13 above, after samples for TracerLab, for rad-chemical analysis.
- Item 15 - Wash all glassware and tools used in hot HNO_3 -HF mixture. Perform rad-chemical analysis on the contents of this wash and the wash from capsule and screw cap to determine the weight of plutonium present.

Item 16 - Count air sampler membranes, record and save, properly identified, for possible analysis by TracerLab.

Items to be recorded and saved

1. All air sample counts
2. All membrane filters
3. Air flows of exhaust air
4. Air flow of air samples
5. Alpha monitoring results for walls of glovebox, plastic enclosure, and fumehood before and after the operations
6. Results of rad-chemical analysis of HNO_3 -HF wash solutions

Items to be weighed and recorded

1. All screens before screening
2. 44 μm screens and larger containing a size range of material
3. All screens after material has been removed
4. All primary containers before placing material in them
5. All primary containers plus the appropriate size material

Given the time limit imposed by AEC Headquarters, these procedures have been read and found to be agreeable for this analysis.

R. E. Vallée
Mound Laboratory

J. F. Hudson
Sandia Corporation

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PRIORITY

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10 AEC

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12 UNCLAS INFORMATION FROM THE IMPACT TEST PERFORMED ON LIVE FUELED
13 CAPSULE NR15, CURRENTLY AT MRC, IS URGENTLY NEEDED FOR INCLUSION

14 INTO THE GE SNAP 27 SAFETY ANALYSIS ADDENDUM TO BE RELEASED BY

15 SEPTEMBER 15, 1963. DATA IS REQUIRED ON THE MASS BALANCE AND THE
16 QUANTITIES ASSOCIATED WITH PARTICLE SIZES OF LESS THAN AND GREATER

17 THAN 10 MICRONS. PLEASE DIRECT MOUND TO PERFORM THIS ANALYSIS.

18 USING OVERTIME AS NECESSARY, TO OBTAIN AND TRANSMIT THE REQUIRED

19 DATA ON SEPTEMBER 13, 1963. RECEIPT OF THE DATA AT HQS SHOULD

20 BE NOT LATER THAN EARLY MONDAY MORNING, SEPTEMBER 15, 1963.

21 END: SNS: SEP: I & M: MMJG

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NNNN

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APPENDIX B

September 13, 1968

During the fines analysis, various problems dictated deviations from the procedures dated September 11, 1968. These deviations were discussed before the fact and because of the limited time available for the analysis were found to be acceptable.

J. F. Hudson, Jr.
Sandia Corporation

APPENDIX C

MONSANTO RESEARCH CORPORATION

Inter-Office Correspondence

From LOCATION : Mound Laboratory, Miamisburg, Ohio cc: P. C. Adams
DATE : September 20, 1968 P. H. Bonnell
SUBJECT : Health Physics Coverage During W. B. Brown
REFERENCE : Opening of Impact Capsule H. E. Meyer
File

TO : Dr. R. E. Vallee

Enclosed is a description of the procedures used in taking and evaluating the alpha wipes and air samples associated with the opening of the impact capsule by P. Bonnell's group on 9/11/68 and the subsequent processing of the PuO_2 .

In evaluating the wipes, air samples, Q-Tips, etc. for alpha contamination one of the following methods was used:

- 1) Eberline Portable Alpha Survey Meter, PAC-1SA
- 2) Eberline Single Channel Alpha Counter (SAC)
- 3) Eberline Multichannel Alpha Counter (MAC)
- 4) Gamma Counting with NaI (Tl) crystal

Where the contamination levels were not high enough to contaminate or raise the background count on our SAC or MAC these instruments were used for alpha counting. For more extensively contaminated items a PAC-1SA was used and this instrument will measure up to 4×10^6 d/m alpha. Above 4×10^6 d/m the gamma counting technique was used.

After the disintegration rate was determined, the count rate in d/m was converted to equivalent micrograms of PuO_2 . This method used and a sample calculation is given on page 9.

The airflow through the fumehood in R-143 was calculated to be 49 CFM. This was done by measuring the linear flow rate in LFM through a known area (fumehood sash open 4"). The pump on the air sampler in the fumehood drew approximately 1 CFM so the contamination as measured on the fumehood air sample was multiplied by 50 to

determine the approximate quantity of Pu-238 leaving the fumehood. The air sampler in the fumehood was a Gelman Filter Holder, Open, 47 mm and it was placed at the entrance of the 6" duct, located at the top, which exhausts the fumehood.

This type air sampler was also used in the glove box in R-127. One air sampler was placed directly in front of the filter opening through which the glove box was exhausted and one sampler was placed near the floor of the glove box, as close as possible to the capsule as it was being opened.

In the glove box in R-127, the design specification call for 25 CFM exhaust, so the contamination determined on these filters was multiplied by 25 to determine the approximate quantity of airborne Pu within the glove box in R-127.

The air samplers used in the plastic glove box were Gelman Filter Holders, In-Line, Aluminum, 47 mm. Two filter holders were used per exhaust line to maximize the chance of complete capture of all radioactive material. See Sketch 1.

The air sample filter media used was Gelman Triacetate Metrical, Type GA-4 and Gelman Glass Fiber Filter, Type E (99.97% retention efficiency for particles exceeding $.3\mu$ as measured by DOP (Diethyl Phthalate). In all cases, except the back up filters on the plastic glove box, the membrane filter was placed on top of the Type E filter (all air flowed thru membrane filter first). In the secondary filters on the plastic glove box, only Type E filter paper was used.

All alpha wipes were taken with Reeve Angel filter paper, Grade 201, 4.25 cm.

Below is a detailed account of the Health Physics aspects of the opening of the capsule from beginning to end.

GLOVE BOX IN R-127

PRIOR TO OPENING OF CAPSULE

September 20, 1968

September 11, 1968

4:15 -

5:00 PM 1) Sketches 2 and 3 show diagrams of this glove box and the location of alpha contamination found by surveying with an Eberline PAC-1SA. All surfaces were also thoroughly wiped, but no removable contamination was found (all wipes <20 d/m alpha). The highest level of fixed contamination found was 1200 d/m.

8:00 PM 2) The background air samples (2) were removed and counted for alpha contamination. The air flows at the beginning (2:50 PM) were 28 liters/min in both samplers and at 8:00 PM when the air samples were pulled (5.17 hr. sample) the air flows were 20 and 24 liters per minute. These results are given below.

		d/m
Exhaust Sample -	Membrane	0
	Type E	6
Floor Sample -	Membrane	6
	Type E	0

AFTER OPENING OF CAPSULE

September 11, 1968

8:35 PM 3) Q-Tip wipe on end of capsule was passed out and counted on a PAC-1SA. It counted 3000 d/m.

AFTER CAPSULE TRANSFERRED TO R-143

September 11, 1968

10:00 PM 4) Wipes were taken in R-127 glove box and the results given below:

	Wipe	d/m	ugm
Bottom	#1	32,550	
	2	236	
	3	176	

September 20, 1968

Left Side	#1	0	
	2	4,224	
	3	6	
Right Side	#1	62	
	2	86	
Con-o-saw		<u>4,068</u>	
Total		41,408	.002

September 12, 1968

- 8:00 PM 5) The two post-opening air samples were removed and counted (24 hour samples). These results are given below:

	d/m	μgm
Exhaust Sample - Membrane	4×10^5	
Type E	2×10^2	
Floor Sample - Membrane	8×10^4	
Type E	<u>3.2×10^5</u>	
Total	8×10^5	.03

The air flows at the beginning were 28 liter per minute and at the end were 20 and 18 liter respectively.

September 13, 1968

- 9:00 AM 6) A PAC-1SA was passed into the glove box and a contamination survey was made on all accessible surfaces. These results and the locations of contamination found are shown in Sketch 4.

	Average d/m/60 cm^2	Area cm^2	μgm
Back Surface	3.0×10^3	10800	.02
Bottom Surface	--	--	21
Right Side	1.5×10^5	8449	.05

Left Side	--	--	negl.
Con-o-saw	2×10^8	--	.7
Vacuum Pumps (2)	2×10^8	--	<u>1.4</u>
Total			23.2

- 7) Next a wipe survey was also taken on the accessible surfaces within the glove box to determine the extent of the loose contamination. These results were:

	Wipe	d/m	μgm
Glove Box Front	#1	10	
	2	14	
	3	100	
Left Side	#1	218	
	2	76	
	3	0	
Bottom	#1	9,852	
	2	<u>53,892</u>	
Total		64,162*	
	Wipe	d/m	μgm
Right Side	#1	40	
	2	6,066	
Vacuum Pump	#1	404	
	2	318	
Con-o-saw	#1	<u>8×10^5</u>	
Total		8.1×10^5 *	

*1 This contamination has already been accounted for.

PLASTIC GLOVE BOX IN R-143

PRIOR TO TRANSFER OF CAPSULE

September 11, 1968

- 8:45 PM 1) The two air samples were removed and counted to determine the background count rate. This was a 4.75 hour sample. All filter papers (4) were counted and found to be 0 d/m.
- 9:00 PM 2) Opened capsule was passed into plastic glove box.
- 12:30 PM 3) Work halted and no contamination found within the fumehood, in the room or on any personnel.

September 12, 1968

12:00 -

- 12:30 PM 4) Wipes were taken within the plastic glove box after all samples had been removed. The results are given below:

		d/m	μgm
Right Side		4×10^5	
Left Side		8×10^5	
Bottom	#1	2.4×10^5	
	2	2.4×10^5	
	3	2.2×10^5	
Back		1.0×10^5	
Right Top		6×10^5	
Left Top		<u>1.6×10^5</u>	
Total		3×10^7	1.1

- 2:00 PM 5) The air samples were removed from the plastic glove box and counted (22 hour sample). These results are given below:

		d/m	μgm
1)	Top Sample - Membrane Type E	2.63×10^7 1.5×10^6	
2)	Top Sample - Type E (back up)	696	
1)	Side Sample - Membrane Type E	3.50×10^7 1.3×10^6	
2)	Side Sample - Type E (back up)	320	
Total		6.4×10^7	2.3

- 6) These filters were replaced and allowed to run again until the plastic glove box was collapsed.

2:30 PM 7) The air sample in the fumehood was removed, counted and a new filter replaced at this time. The results were 6540 d/m and 150 d/m on the membrane and type E filters respectively.

2:45 PM 8) After collapsing the plastic glove box the air samples in the glove box and fumehood were removed and counted. The results were:

		d/m	μgm
Fumehood -	Membrane Type E	2 50	
Glove Box			
Top Sample -	Membrane Type E	8×10^3 1.5×10^6	
Top Sample -	Type E (back up)	480	
Side Sample -	Membrane Type E	4×10^4 8×10^4	

September 20, 1968

Side Sample - Type E (back up)	1800	
	<hr/>	
Total	2.8×10^6	.01

Wipes were also taken within the fumehood before and after collapsing and these indicated no contamination was released from the plastic glove box while collapsing.

- 7:30 PM 9) The air samples within the fumehood were removed, counted and new filters replaced just prior to beginning the boiling operation of the microsphere, alcohol solution. The results were:

	d/m	μgm
Membrane	3.2×10^6	
Type E	<u>2.0×10^6</u>	
Total	5.2×10^6	1.0

September 13 & 14, 1968

- 10) Routine work continued in fumehood, transferred samples to analytical, etc. There were no unusual Health Physics problems.

September 16, 1968

- 8:30 AM 11) The plastic lining in the fumehood was removed and packaged for gamma scan. When this was completed the air sample in the fumehood was removed and counted.

	d/m	μgm
Membrane	1.24×10^6	
Type E	<u>1.95×10^6</u>	
Total	1.44×10^6	260

- 9:00 AM 12) After removal of the plastic, the inside fumehood surfaces were wiped for removable contamination. Direct readings would not be meaningful. Sketches 5 and 6 show the approximate location of these wipes and their

values.

The total amount found on the 22 wipes throughout the fumehood was 1.2×10^8 d/m. Assuming that these wipes removed one hundredth of the material present the total amount on the fumehood surfaces would equal approximately 1.2×10^{11} d/m or 4360 μgm or 4.4 mgm.

SAMPLE CALCULATION

$$1 \text{ gm Pu-238} = 17.4 \text{ ci}$$

$$1 \text{ gm } ^{238}\text{PuO}_2 = [1] [.8] [.88] = .7 \text{ gm of Pu-238}$$

$$1 \text{ gm } ^{238}\text{PuO}_2 = [17.4 \text{ ci}] [.7] = 12.2 \text{ ci}$$

$$.08 \text{ } \mu\text{gm of } ^{238}\text{PuO}_2 = 1 \text{ } \mu\text{ci}$$

$$.08 \text{ } \mu\text{gm of } ^{238}\text{PuO}_2 = 2.2 \times 10^8 \text{ d/m}$$

$$\frac{X \text{ d/m}}{Y \text{ } \mu\text{gm}} = \frac{2.2 \times 10^8 \text{ d/m}}{.08 \text{ } \mu\text{gm}}$$

X d/m = the activity in d/m as measured on the air sample filter paper, wipe, etc.

Y μgm = quantity to be calculated.

D. A. Edling
D. A. Edling

DAE/no

Sketch 1. Plastic glovebox

Sketches 2 and 3

Sketch 4

Sketch 5

d/M

Sketch 6

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APPENDIX C

Three letters dated 1/18/68, 3/7/68, and 3/11/69 from Walter Holland, Tracerlab, to Burton Hill, Sandia Corporation, reporting the results of Tracerlab analysis of Sample 4

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C O P Y

TRACERLAB

A DIVISION OF LABORATORY FOR ELECTRONICS INC

2030 WRIGHT AVENUE • RICHMOND CALIF 94804

January 18, 1968

235-2633

CABLE ADDRESS
LFE

Mr. Burton Hill
Division 9312
Sandia
P.O. Box 5800
Albuquerque, New Mexico

Ref: Contract No.
48-6716
W.O. 12762

Dear Mr. Hill:

Enclosed are the results of our particle investigations with your sample 4. This information is contained in Figure 1, a numerical size distribution histogram; Figure 2, a cumulative plot of equivalent $^{238}\text{PuO}_2$ diameters versus % less than number count and mass; and Table 1, a list of the mass and numerical percentages of the particles in each size range. It is emphasized that this information describes the particles in size range from 0.19 to 11.6 μ and should not be extrapolated for particles outside these limits. Of course it can be combined with information obtained by other means for the larger and smaller particles. Specifically, it can be combined with the results regarding the larger particles obtained by Mound Laboratory and data regarding the smaller particles which we will obtain by radiochemical analysis within 2 weeks.

As was found during our earlier work with impacted samples, the radioactive particles are flakes which have equivalent $^{238}\text{PuO}_2$ diameters which are 1/2 to 1/3 smaller than the actual x and y dimensions. Photomicrographs of typical particles are shown in Figures 3 through 5. Additional pictures will be included in the final report of this work. There was no evidence of the radioactive particles agglomerating with non-radioactive particles. Considerable difficulty was encountered when an attempt was made to photomicrograph isolated radioactive particles having submicron equivalent diameters.

Should you have any questions, please contact me.

Very truly yours,

/S/ Walter Holland, Supervisor
Physical Measurements

WH/br
encl/

cc: Mr. Paul Bonnell
Mound Laboratory
Monsanto Research Cor.
Miamisburg, Ohio 45342

TABLE 1 - Mass and Numerical Percentages of Particles in Each Size Range

Top of Range (μ)	Numerical	Mass %
0.19	0	0
0.21	1.3	4.0×10^{-4}
0.30	26.2	2.4×10^{-2}
0.42	33.9	9.1×10^{-2}
0.60	19.2	1.5×10^{-1}
0.87	10.2	2.4×10^{-1}
1.2	3.2	2.1×10^{-1}
1.8	2.7	5.5×10^{-1}
2.6	1.2	3.2×10^{-1}
3.6	8.0×10^{-1}	1.5
5.3	2.0×10^{-1}	1.1
7.5	2.0×10^{-1}	3.2
10.8	5.0×10^{-1}	23.6
11.6	5.0×10^{-1}	69.1

Figure 1 Numerical size distribution histogram of equivalent $^{238}\text{PuO}_2$ particle measurements obtained from sample 4

Figure 2 - Plot of equivalent $^{238}\text{PuO}_2$ diameters versus % less than number count and mass for sample 4. The total mass for the 950 particle measurements is 9.8×10^{-8} gm.

Figure 3 - Photomicrograph of radioactive particle A-1-4 which was obtained from impacted sample 4. The equivalent $^{238}\text{PuO}_2$ diameter obtained after isolation is $10.6\ \mu$. The magnification is such that $10\ \mu$ are equivalent to $5\ \text{mm}$. The shaded circle extending about $20\ \mu$ around the particle is due to radiation damage by the high alpha activity.

Before

After

Figure 4 - Photomicrographs of a radioactive particle B-1-25 which was obtained from impacted sample 4. The equivalent $^{238}\text{PuO}_2$ diameters for before and after isolation are 3.06 and 3.14 μ , respectively. The magnification is such that 10 μ are equivalent to 5 mm.

Before

After

Figure 5 - Photomicrographs of a radioactive particle C-1-10 which was obtained from impacted sample 4. The equivalent $^{238}\text{PuO}_2$ diameters for before and after isolation are 1.34 and 1.36 μ , respectively. The magnification is such that 10 μ are equivalent to 5 mm.

TRACERLAB

A DIVISION OF LABORATORY FOR ELECTRONICS, INC.

2030 WRIGHT AVENUE • RICHMOND, CALIFORNIA

March 7, 1968

BEACON 5-2633
CABLE ADDRESS
LFE

Ref Contract 48-6716
WO #12762

Mr. Burton Hill
Division 9312
Sandia
P. O. Box 5800
Albuquerque, New Mexico 87115

Dear Mr. Hill:

I want to express how much I appreciated meeting with you and Paul Bonnell of Mound Laboratory on the 26th and 27th of February to discuss the particle work on the impacted samples. The main decision of this meeting was that I would combine the Mound and Tracerlab results using a single basis for measuring particle sizes. It was anticipated that the finalized results would contain numerical and mass histograms for particles having diameters from 250μ to 0.19μ . Following is the preliminary work which I have done for sample 4. It will be noted that I obtained the mass histogram for all the microspheres impacted and the resulting fragments; however, insufficient data were available for plotting the numerical distribution.

The results obtained by Mound and Tracerlab are summarized as follows:

Mound Results

1. Equivalent circular diameters for a representative portion of each of the sieved fractions from 44μ - 20μ and 20μ - 0 of the impacted sample 4 microspheres were obtained. See Tables I and II.
2. Each of the fractions obtained by sieving the impacted microspheres through 210μ , 177μ , 149μ , 125μ , 105μ , 74μ , 44μ , 20μ , and 10μ sieves was weighed. This information was used to calculate the percent of the total mass of the impacted particles in each size range. See Table III for the percent mass for the three smallest fractions.
3. Photomicrographs of representative portions of each of the 10 fractions were prepared.

Tracerlab Results

1. Equivalent $^{238}\text{PuO}_2$ spherical diameters for a representative portion of the $<20\ \mu$ sieved fraction are presented in Table IV. These values, varying from $0.19\ \mu$ - $11.6\ \mu$, are plotted in Figure 1 as a numerical histogram having 13 ranges. The data are also plotted in Figure 2 on a cumulative numerical and mass percentages versus particle diameter basis.
2. Photomicrographs of isolated typical radioactive particles were prepared. These particles have x,y (projected) dimensions ranging from $1\ \mu \times 1\ \mu$ to $30\ \mu \times 30\ \mu$.
3. Radiochemical analysis was performed to determine the total amount of plutonium in the portion of sample 4 used by Tracerlab personnel for particle study. This result was interpreted as indicating that no significant portion of the mass of the radioactive particles in the $<20\ \mu$ fraction has equivalent $^{238}\text{PuO}_2$ spherical diameters $<0.19\ \mu$, the smallest measured by the hollow star technique.

It will be noted that the particle measurements used for the Tracerlab results are expressed as equivalent spherical $^{238}\text{PuO}_2$ diameters. These values were obtained by the hollow star technique (1,2) or electronic counting of individual particles. The Mound measurements of the radioactive particles are listed as equivalent circular diameters which were obtained from photomicrographs using a Zeiss counter. This instrument has a circle of light with a variable diameter which is coupled with instrumentation which records the diameters of the circles equivalent to the projected (x,y) areas for each particle shown on a photomicrograph.

In order to convert equivalent spherical $^{238}\text{PuO}_2$ diameters to equivalent circular diameters, both types of measurements were obtained from radioactive particles which Tracerlab personnel isolated. These measurements are shown in Table V. A plot of these values is shown in Figure 3. For particles smaller than $2\ \mu$ in diameter, the equivalent circular and spherical diameters are approximately identical. The ratio of circular to spherical diameters increases to 2.1 for a circular diameter of $17\ \mu$ and 2.5 for a circular diameter of $28\ \mu$.

Several points along the curve of Figure 3 were selected for calculation of the average thickness for the radioactive fragments. Such a calculation is performed by dividing the equivalent $^{238}\text{PuO}_2$ volumes by the corresponding equivalent circular areas.* These values for the

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- 1 SC-SR-68-3470, Analysis of Debris from Ablation of $^{238}\text{PuO}_2$ Microspheres in a Hyperthermal Arc Tunnel, W. Holland, Tracerlab for Sandia Corporation (in preparation).
 - 2 Third Quarterly Report of SNAP-9A Particle Studies during 15 April 1966 - 15 July 1966, W. Holland, Tracerlab, a division of LFE, HASL Contract No. AT(30-1)-3547 Mod. 3, September 14, 1966.
- * It is pointed out that the thickness calculated in this manner for spheres would be thickness = $2/3$ diameter.

selected points from Figure 3 are listed in Table VI. Note that the particle thicknesses are remarkably close to $1\ \mu$ indicating the particles are flakes.

Figure 3 was used to obtain the corresponding equivalent circular diameters for the size ranges of the Tracerlab results shown in Table IV. These new values are shown in Table VII. In addition, this table contains the percentages of the mass of the microspheres impacted for each of these size ranges. These values were obtained by multiplying each of the percents of the mass of the particles measured in each size range by 0.727%, the percent of the mass of all the impacted particles which are in the range from 0 - $20\ \mu$. See Table III.

In a similar manner, equivalent $^{238}\text{PuO}_2$ diameters were obtained for the corresponding circular diameter size ranges for the Mound results. These values are shown in Table VIII. The mass of the particles measured in each size range was obtained by the product of the mass of the average size particle in each size range and the number of particles in each range. The percents of the mass of all microspheres were determined by the product of each percent mass for particles measured in range and 0.72%, the percent of the mass of all particles in the range from 10 - $20\ \mu$. See Table III.

It is point out that the four size ranges for the Mound results of Table VIII overlap the four largest ranges of the Tracerlab results shown in Table VII.

In order to compare the overlapping of the two sets of results and attempt combining them in a meaningful manner, the values of cumulative percent mass of the impacted particles were plotted with their particle sizes as shown in Figure 4. The actual values plotted in Figure 4 are shown in Tables IX and X. The Tracerlab values which are circled were used to draw the solid line. The Mound values represented by "x's" were obtained by successively adding them to the Tracerlab cumulative percentage for $10\ \mu$, as shown in Table X. The nearness of the Mound values (x's) to the solid line curve of Figure 4 indicates the two sets of data may dovetail nicely. Such conservativeness is appropriate because additional results will be combined with the Figure 4 information in the next few days when Mound is expected to provide the author with the information required for determining the equivalent spherical and circular diameters for particles having circular diameters $>30\ \mu$. Specifically, this information consists of photomicrographs of particles in the 20 - $40\ \mu$ sieved fraction. It is hoped that the thicknesses of these particles can be estimated from the photomicrographs. If possible, an average thickness will be determined, the equivalent volumes estimated, and thus the equivalent spherical diameters obtained. The spherical diameters are required to obtain the mass of the average size particle in each size range. After these masses are obtained, one can calculate the percent mass in the 20 - $40\ \mu$ fraction and also the percent mass of all the impacted particles. If photomicrographs and percent total mass values are available for the larger sieved fractions, similar information will be obtained for them. Not only would we have a cumulative plot of the percent mass of total microspheres impacted versus equivalent circular diameter, but also the author is optimistic a cumulative plot of the

Mr. Burton Hill

-4-

March 7, 1968

total number of microspheres impacted versus equivalent circular diameter is obtainable.

At this time, estimating the accuracy and precision of the work shown in Figure 4 is quite difficult. However, the author considers $\pm 200\%$ as a reasonable estimate for the precision each of the Figure 4 percentages. Obviously, we will gain a better opinion for the precision when the results from several duplicate experiments are treated in the manner described here for sample 4. The reader is advised that estimating the accuracy, that is the comparison of size distributions of particles produced by actual and simulated intact reentry, will be much more difficult.

Please contact me regarding your questions and comments. (And I expect some.)

Very truly yours,

/S/ Walter Holland

/sfb

cc: Paul Bonnell

TABLE I MOUND DATA FOR RADIOACTIVE PARTICLES
IN A PORTION OF THE 0-20 μ SIEVED
FRACTION OF SAMPLE 4

<u>Size Ranges</u> <u>Equivalent Circular</u> <u>Diameters (μ)</u>	<u>No. of</u> <u>Particles</u> <u>Per Size Range</u>
10 - 14	150
14 - 18.5	1639
18.5 - 23	4695
23 - 27.5	1127
27.5 - 32	430
32 - 36	142
36 - 40.5	55
40.5 - 45	13
45 - 49	4
Total	8256

TABLE II MOUND DATA FOR RADIOACTIVE PARTICLES
IN A PORTION OF THE 20-40 μ SIEVED
FRACTION OF SAMPLE 4

<u>Size Ranges</u> <u>Equivalent Circular</u> <u>Diameters (μ)</u>	<u>No. of</u> <u>Particles</u> <u>Per Size Range</u>
10 - 14	1
14 - 18.5	24
18.5 - 23	261
23 - 27.5	571
27.5 - 32	731
32 - 36	864
36 - 40.5	666
40.5 - 45	652
45 - 49	458
49 - 54	390
54 - 58.5	188
58.5 - 63	111
63 - 67.5	50
67.5 - 72	15
72 - 76	5
Total	4987

TABLE III PERCENTS OF THE TOTAL MASS OF
IMPACTED MICROSPHERE FRAGMENTS
WHICH WERE COLLECTED ON THE
THREE SMALLEST SIEVES

Approximate Size Ranges (μ)	% Mass
20 - 40	4.5
10 - 20	0.72
0 - 10	0.007

TABLE IV TLW DATA FOR RADIOACTIVE PARTICLES IN A PORTION
OF THE 0-20 μ SIEVED FRACTION OF SAMPLE 4

Size Ranges Equivalent $^{238}\text{PuO}_2$ Spherical Diameters (μ)	No. of Particles Per Size Range	Numerical % Per Size Range	Mass of Particles Per Size Range ($\mu\mu\text{g}$)	% Mass Per Size Range
0.19 - 0.21	12	1.3	0.4	5.4×10^{-4}
0.21 - 0.30	248	26.2	23.5	3.2×10^{-2}
0.30 - 0.42	322	33.9	88.7	1.2×10^{-1}
0.42 - 0.60	182	19.2	146	2.0×10^{-1}
0.60 - 0.87	97	10.2	232	3.2×10^{-1}
0.87 - 1.2	30	3.2	208	2.8×10^{-1}
1.2 - 1.8	26	2.7	537	7.3×10^{-1}
1.8 - 2.6	11	1.2	666	9.1×10^{-1}
2.6 - 3.6	8	0.8	1,416	1.9
3.6 - 5.3	2	0.2	1,050	1.4
5.3 - 7.5	2	0.2	3,114	4.2
7.5 - 10.8	5	0.5	23,060	31.4
10.8 - 11.6	5	0.5	42,900	58.4
		Total	73,441.6	

TABLE V EQUIVALENT SPHERICAL $^{238}\text{PuO}_2$ AND
CIRCULAR DIAMETERS FOR RADIOACTIVE
PARTICLES ISOLATED FROM SAMPLE 4

Particle Designations	Equivalent Spherical $^{238}\text{PuO}_2$ Diameters (μ)	Equivalent Circular Diameters (μ)
A-1-4	10.6	30
B-1-25	3.14	5
C-1-10	1.4	2
A-1-1	11.2	26
A-1-3	11	26
A-1-6	11	28
A-1-5	10	26
A-1-7	10	28
A-1-8	4.8	8
A-1-9	5.4	12
A-1-11	3.6	5
A-1-12	3.9	6
A-1-14	6.6	13
B-1-3	1.5	3
B-1-4	3.5	5
B-1-9	1.4	2
B-1-30	2.5	3
C-1-1	1.1	1.1
C-1-7	1.1	1.1
A-1-10	10.7	28
A-1-13	11.5	28
A-1-15	8.6	21

TABLE VI VALUES FOR CALCULATING THICKNESSES
OF PARTICLE FRAGMENTS

Equivalent Spherical $^{238}\text{PuO}_2$ Measurements		Equivalent Circular Measurements		Calculated Thickness (μ)
Diameter (μ)	Volume (μ^3)	Diameter (μ)	Area (μ^2)	
1.9	3.4	2	3.1	1.1
5.3	78	10	78.5	1.0
8.8	357	20	314	1.1
11.2	736	28	615	1.2

TABLE VII TLW DATA COMBINED WITH MOUND DATA FOR
RADIOACTIVE PARTICLES IN A PORTION OF
THE 0-20 μ SIEVED FRACTION OF SAMPLE 4

Size Ranges		No. of Particles Per Size Range	Mass of Particles Per Size Range (μg)	% Mass Per Size Range	% Mass of Microspheres Impacted*
Equivalent Circular Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Spherical Diameters (μ)				
0.19 - 0.21	0.19 - 0.21	12	0.4	5.4×10^{-4}	4×10^{-6}
0.21 - 0.30	0.21 - 0.30	248	23.5	3.2×10^{-2}	2×10^{-4}
0.30 - 0.42	0.30 - 0.42	322	88.7	1.2×10^{-1}	9×10^{-4}
0.42 - 0.60	0.42 - 0.60	182	146	2.0×10^{-1}	1.5×10^{-3}
0.60 - 0.87	0.70 - 0.87	97	232	3.2×10^{-1}	2.3×10^{-3}
0.87 - 1.6	0.87 - 1.2	30	208	2.8×10^{-1}	2.0×10^{-3}
1.6 - 3.0	1.2 - 1.8	26	537	7.3×10^{-1}	5.3×10^{-3}
3.0 - 4.8	1.8 - 2.6	11	666	9.1×10^{-1}	6.6×10^{-3}
4.8 - 7.0	2.6 - 3.6	8	1,416	1.9	1.4×10^{-2}
7.0 - 11.2	3.6 - 5.3	2	1,050	1.4	1.0×10^{-2}
11.2 - 16.4	5.3 - 7.5	2	3,114	4.2	3.1×10^{-2}
16.4 - 27.0	7.5 - 10.8	5	23,060	31.4	2.3×10^{-1}
27.0 - 29.5	10.8 - 11.6	5	42,900	58.4	4.2×10^{-1}
		Total	73,441.6		

*These values were obtained by multiplying the corresponding values in the previous column by 0.727%, the percent of the mass of all the impacted particles which are in the range from 0 - 20 μ . See Table III.

TABLE VIII MOUND DATA COMBINED WITH TLW DATA FOR
RADIOACTIVE PARTICLES IN A PORTION OF
THE 0-20 μ SIEVED FRACTION OF SAMPLE 4

Size Ranges		No. of Particles Per Size Range	Mass of Particles Per Size Range (μg)	% Mass Per Size Range	% Mass of Microspheres Impacted*
Equivalent Circular Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Spherical Diameters (μ)				
10 - 14	5 - 6.6	150	0.17	0.5	3.6×10^{-3}
14 - 18.5	6.6 - 8.1	1639	3.9	11.4	8.2×10^{-2}
18.5 - 23	8.1 - 9.4	4695	19	55.6	4×10^{-1}
23 - 27.5	9.4 - 10.9	1127	7.1	20.8	1.5×10^{-1}
27.5 - 32	10.9 - 12.2	430	4.0	11.7	8.4×10^{-2}
		Total	34.2		

*These values were obtained by multiplying the corresponding values in the previous column by 0.72%, the percent of the mass of all the impacted particles which are in the range from 10-20 μ . See Table III.

TABLE IX TRACERLAB DATA FOR CUMULATIVE
% MASS VALUES

Size Ranges			
Equivalent Circular Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Spherical Diameters (μ)	% Mass of Microspheres Impacted	Cumulative % Mass
0.19 - 0.21	0.19 - 0.21	4×10^{-6}	4×10^{-6}
0.21 - 0.30	0.21 - 0.30	2×10^{-4}	2×10^{-4}
0.30 - 0.42	0.30 - 0.42	9×10^{-4}	1.1×10^{-3}
0.42 - 0.60	0.42 - 0.60	1.5×10^{-3}	2.6×10^{-3}
0.60 - 0.87	0.60 - 0.87	2.3×10^{-3}	4.9×10^{-3}
0.87 - 1.6	0.87 - 1.2	2.0×10^{-3}	6.9×10^{-3}
1.6 - 3.0	1.2 - 1.8	5.3×10^{-3}	1.2×10^{-2}
3.0 - 4.8	1.8 - 2.6	6.6×10^{-3}	1.9×10^{-2}
4.8 - 7.0	2.6 - 3.6	1.4×10^{-2}	3.3×10^{-2}
7.0 - 11.2	3.6 - 5.3	1.0×10^{-2}	4.3×10^{-2}
11.2 - 16.4	5.3 - 7.5	3.1×10^{-2}	7.4×10^{-2}
16.4 - 27.0	7.5 - 10.8	2.3×10^{-1}	3.0×10^{-1}
27.0 - 29.5	10.8 - 11.6	4.2×10^{-1}	7.24×10^{-1}

TABLE X MOUND DATA COMBINED WITH TRACERLAB DATA
FOR CUMULATIVE % MASS VALUES

Size Ranges			
Equivalent Circular Diameters (μ)	Equivalent $^{238}\text{PuO}_2$ Spherical Diameters (μ)	% Mass of Microspheres Impacted	Cumulative % Mass
0 - 10		4×10^{-2}	$4 \times 10^{-2*}$
10 - 14	5.0 - 6.6	3.6×10^{-3}	4.4×10^{-2}
14 - 18.5	6.6 - 8.1	8.2×10^{-2}	1.3×10^{-1}
18.5 - 23	8.1 - 9.4	4×10^{-1}	5.3×10^{-1}
23 - 27.5	9.4 - 10.9	1.5×10^{-1}	6.8×10^{-1}
27.5 - 32	10.9 - 12.2	8.4×10^{-2}	7.6×10^{-1}
*This is the Tracerlab value for 10 μ . See Figure 5.

Figure 1 Numerical size distribution histogram of equivalent $^{238}\text{PuO}_2$ particle measurements obtained from sample 4

Figure 2 Plot of equivalent $^{238}\text{PuO}_2$ diameters versus percent less than number count and mass for sample 4. The total mass for the 950 particle measurements is 7.3×10^{-8} gm.

Figure 3 Plot of equivalent spherical $^{238}\text{PuO}_2$ and circular diameters for radioactive particles isolated from sample 4

Figure 4 Plot of equivalent diameter versus percent less than for the mass of the total microspheres impacted as sample 4. The solid line is determined by the circled points representing the Tracerlab results. The "x" points are Mound results successively added to the Tracerlab value at 10 μ.

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TRACER LAB

A DIVISION OF LABORATORY FOR ELECTRONICS, INC.

2030 WRIGHT AVENUE • RICHMOND, CALIFORNIA

March 11, 1968

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Mr. Burton Hill
Division 9312
Sandia
P. O. Box 5800
Albuquerque, New Mexico 87115

Dear Mr. Hill:

I wish to update the information concerning Sample 4 which was included in my letter of March 7. The results of radiochemical analysis and the particle studies have been corrected and are listed in Table I.

TABLE I Determination of $^{238}\text{PuO}_2$ in Sample 4 by
Radiochemical and Particle Analyses

<u>Analysis</u>	<u>Total $^{238}\text{PuO}_2$ (μg)</u>
Radiochemical	0.063
Hollow Star	0.073

The conclusion is still valid that comparison of the radiochemical and particle analysis results indicates no significant portion of the total mass of plutonium impacted is in particles having equivalent $^{238}\text{PuO}_2$ diameters smaller than 0.19μ .

The corrected values for the mass percentages for particles in each size range are listed here in Table II. This table also contains the cumulative numerical and mass percentages for particles in each size range.

Should you have any questions, please contact me.

Very truly yours,

/S/ Walter Holland, Supervisor
Physical Measurements

/sfb

TABLE II Mass and Numerical Percentages of
Particles in Each Size Range

Size Range (μ)	Numerical (%)	Cumulative Numerical (%)	Mass (%)	Cumulative Mass (%)
0.19 - 0.21	1.3	1.3	5.4×10^{-4}	5.4×10^{-4}
0.21 - 0.30	26.2	27.5	3.2×10^{-2}	3.3×10^{-2}
0.30 - 0.42	33.9	61.4	1.2×10^{-1}	1.5×10^{-1}
0.42 - 0.60	19.2	80.6	2.0×10^{-1}	3.5×10^{-1}
0.60 - 0.87	10.2	90.8	3.2×10^{-1}	6.7×10^{-1}
0.87 - 1.2	3.2	94.0	2.8×10^{-1}	9.5×10^{-1}
1.2 - 1.8	2.7	96.7	7.3×10^{-1}	1.7
1.8 - 2.6	1.2	97.9	9.1×10^{-1}	2.6
2.6 - 3.6	8.0×10^{-1}	98.7	1.9	4.5
3.6 - 5.3	2.0×10^{-1}	98.9	1.4	5.9
5.3 - 7.5	2.0×10^{-1}	99.1	4.2	10.1
7.5 - 10.8	5.0×10^{-1}	99.6	31.4	41.5
10.8 - 11.6	5.0×10^{-1}	100.1	58.4	99.9

APPENDIX D

Walter D. Holland, TLW 6004, Particle
Analysis of Radioactive Fragments with
Diameters $<10\ \mu$ From Impacted Samples 18 and 20,
May 28, 1968.

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C O P Y

TLW-6004

PARTICLE ANALYSIS OF RADIOACTIVE FRAGMENTS WITH
DIAMETERS $< 10 \mu$ FROM IMPACTED SAMPLES 18 and 20

May 28, 1968

MONSANTO RESEARCH CORPORATION
A Subsidiary of Monsanto Company
Mound Laboratory
Miamisburg, Ohio 45342

Contract No. 31966-TT

Reported by

/S/ Walter D. Holland
Project Leader

Approved by

/S/ Dr. Marcel Nathans
Manager, Research
and Development Dept.

TRACERLAB
A Division of Laboratory for Electronics, Inc.
2030 Wright Avenue
Richmond, California 94804

ABSTRACT

Representative portions of $^{238}\text{PuO}_2$ particles in sieved fractions of less than $10\ \mu$ from two experiments simulating intact atmospheric re-entry of a SNAP device were analyzed. Equivalent $^{238}\text{PuO}_2$ spherical diameters from $0.2\ \mu$ to $10.6\ \mu$ were determined with the hollow-star and electronic-alpha-counting techniques. The size distributions of both samples were plotted as numerical and mass percentages. The total equivalent $^{238}\text{PuO}_2$ masses in the two samples, as determined by the hollow-star and electronic-counting techniques agreed within 10% with those determined radiochemically. These results indicated that particles with equivalent $^{238}\text{PuO}_2$ diameters smaller than $0.2\ \mu$ did not significantly contribute to the total particulate mass for either sample. The results of optical microscopy and electron microprobe analyses of single particles were used with electronic counting data to determine that the radioactive particles with equivalent $^{238}\text{PuO}_2$ diameters greater than $4\ \mu$ were flakes as thin as $0.4\ \mu$.

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1.0 INTRODUCTION

The purpose of these studies was to determine the size distribution of fragments from SNAP fuel particles in capsules 18 and 20, which were impacted under conditions simulating intact re-entry from the atmosphere at different velocities. These capsules, each containing 5.4 grams of $^{238}\text{PuO}_2$ microspheres, were impacted head-on into granite at velocities of 105.9 ft/sec (capsule 18) and 101.7 ft/sec (capsule 20).

2.0 SAMPLE PREPARATION

The content of each capsule was removed and placed on a 44- μ -mesh screen. Ethyl alcohol was added to wash particles smaller than 44 μ through the mesh. The suspensions were then passed successively through two electroformed sieves, one with 20- μ pores and one with 10- μ pores (sieves manufactured by Buckbee Mears Company, St. Paul, Minnesota). The fragments from the microspheres were separated into three fractions:

- < 44 μ > 20 μ
- < 20 μ > 10 μ
- < 10 μ

A few drops of the <10 μ fraction from both samples 18 and 20 were dispersed on microscope slides (ten slides for each sample). A few drops of 4% collodion in amyl acetate were added to each slide to attach the particles firmly to the slide.

3.0 EXPERIMENTAL WORK WITH <10 μ FRACTIONS

3.1 HOLLOW-STAR EXAMINATIONS

With a method developed for measuring plutonium particles produced in a hyperthermal tunnel,⁽¹⁾ each of the ten slides containing a drop representative of the suspension of the <10 μ fraction from samples 18 and 20 was exposed for 10 seconds to nuclear emulsion plates which were scanned for hollow stars with an optical microscope. (The hollow-star technique is described in detail in other reports.)^(1,2) Figure 1 shows a typical hollow star produced by an alpha-emitting particle. From the hollow stars, the equivalent $^{238}\text{PuO}_2$ diameters and the locations of individual radioactive particles were determined.

For each sample, all radioactive particles on one of the slides were isolated (removed to another slide) so that their hollow stars would not obscure the stars produced by particles with smaller equivalent $^{238}\text{PuO}_2$ diameters during the subsequent exposures. (These exposures were of increasing durations.) A drop of 50% collodion in amyl acetate was placed over each particle. After the collodion had dried, a segment of about 25 μ in diameter was scribed to include each radioactive particle. These segments ("eyes") were then transferred with a pointed 3-mm-diameter rod to another microscope slide. The radioactive particles remaining on the original slide were washed off with amyl acetate and the suspension from each slide was distributed evenly over four microscope slides.

To calculate the equivalent $^{238}\text{PuO}_2$ diameters of particles with diameters larger than 4 μ more accurately than is possible with the hollow-star technique, the particles were counted electronically after isolation. The counting system consisted of a 1-1/2 in. diameter Nuclear Enterprises plastic scintillator, type NE 102, coupled to a

(1) SC-SR-68-3470, Analysis of Debris from Ablation of $^{238}\text{PuO}_2$ Microspheres in a Hyperthermal Arc Tunnel, W. Holland, Tracerlab for Sandia Corporation (in preparation).

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

Dumont 6292 photomultiplier tube attached to a Tracerlab Compu/Matic scaler, model SX-4. The background for this system is about 0.6 dpm.

The slides were treated in a Tracerlab LTA-600 Low-Temperature Asher to remove the 50% collodion, which, in this concentration, absorbs the alpha radiation, preventing accurate hollow-star measurements. A few drops of 4% collodion were added to the ash on each slide, and the slides were exposed to nuclear emulsions. After 2-minute exposures of the sample-18 slides (Fig. 2) and 1-minute exposures of the sample-20 slides (Fig. 3), these slides were scanned for hollow stars to determine the additional equivalent $^{238}\text{PuO}_2$ diameters and particle locations. For each sample, the radioactive particles located on one of the slides were isolated. The particles remaining on each of these two slides (one from sample 18 and one from sample 20) were transferred to four slides (a total of eight slides). The four slides from sample 18 and two slides from sample 20 were exposed to nuclear emulsions for 15 minutes; they were then scanned as before, and the equivalent $^{238}\text{PuO}_2$ diameters and particle locations were determined.

All the equivalent $^{238}\text{PuO}_2$ measurements for each exposure period were normalized to reflect the total particles on the ten slides for each sample (Figs. 2 and 3). The normalized values were 8,491 total particles for sample 18 and 9,165 total particles for sample 20.

3.2 PLOTS OF EQUIVALENT $^{238}\text{PuO}_2$ MEASUREMENTS

The number and size of the hollow stars were used to calculate the numerical and mass percentages of particles in several size ranges for each sample. These values are listed in Table I and plotted as numerical size distributions, cumulative number counts, and cumulative mass plots in Figures 4 to 7. The equivalent diameters range from 0.2 μ to 3.7 μ for sample 18 and from 0.2 μ to 10.9 μ for sample 20.

3.3 DETERMINATION OF TOTAL ^{238}Pu FOR EACH FRACTION OF SAMPLES 18 AND 20 WHICH WAS SUBJECTED TO PARTICLE ANALYSIS

To determine the total mass of ^{238}Pu for each fraction of samples 18 and 20 that was analyzed by the hollow-star technique and electronic counting, the slides with the plutonium for these portions were analyzed radiochemically. Specifically, the portions analyzed were the slides

subjected to exposure before hollow-star examination and the slides with the isolated radioactive particles. The radiochemical procedure is described in an earlier report.⁽¹⁾ The results of the analysis are shown in Table II.

Table I. Numerical and Mass Percentages of Particles in Each Size Range for Samples 18 and 20

Top of Range (μ)	Sample 18		Sample 20	
	Numerical %	Mass %	Numerical %	Mass %
0.21	--	--	--	--
0.30	2.9	0.07	1.3	0.02
0.42	26.3	1.8	18.4	0.8
0.61	28.7	5.8	37.6	5.1
0.87	17.5	10.5	21.7	8.7
1.3	17.8	31.5	13.1	15.5
1.8	5.5	28.9	6.3	22.0
2.6	1.1	16.6	1.1	11.1
3.7	0.1	4.8	0.4	11.6
5.3	--	--	0.1	10.9
7.6	--	--	0.02	5.8
10.9	--	--	0.01	8.5

Table II. $^{238}\text{PuO}_2$ Determined by Radiochemical and Particle Analyses

Sample Designation	$^{238}\text{PuO}_2$ Analyses	
	Radiochemical (10^{-8} g)	Hollow Star (10^{-8} g)
18	3.8	3.5
20	5.4	5.6

There is a difference of less than 10% between the total mass of $^{238}\text{PuO}_2$ determined radiochemically for each sample and the corresponding values determined by the hollow-star technique. This is interpreted as indicating that no significant portion of the total mass of either sample is in the form of particles with equivalent $^{238}\text{PuO}_2$ diameters smaller than 0.2μ (the smallest diameter calculated by the hollow-star technique for these samples).

3.4 ISOLATION OF TYPICAL RADIOACTIVE PARTICLES

Several of the radioactive particles isolated (Section 3.1) were alpha-counted electronically and examined for hollow stars both before and after isolation. Photomicrographs of typical particles are shown in Figures 8 to 10. The radioactive particles were generally transparent and pale yellow. In only a few cases, the calculation of the equivalent $^{238}\text{PuO}_2$ diameters of a particle revealed a decrease in value as a result of the isolation process. The photomicrographs of the particles before and after isolation also showed little variation in size.

3.5 MICROPROBE ANALYSIS OF TYPICAL RADIOACTIVE PARTICLES

Several radioactive particles from samples 18 and 20 were analyzed by electron microprobe to determine their composition. Each of these particles was isolated and attached to a one-inch diameter bakelite disc with a drop of 50% collodion. The discs were vacuum-coated with a conductive film of carbon and examined in a microprobe analyzer for plutonium, tungsten, silver, copper, nickel, iron, manganese, chromium, and silicon. The particles analyzed with the microprobe are shown in Figures 8 and 11 to 14.

The Applied Research Laboratories electron microprobe analyzer, model EMX, can scan an area as small as $12.5\ \mu$ by $12.5\ \mu$, at the accelerating voltage of 25 kV. The electron beam has a diameter as small as $1\ \mu$. The characteristic X-rays produced by interaction between the electrons and the material being scanned are diffracted and passed through a slit so that the X-rays from a single element can be counted in an electronic counter. After amplification, the output from the counter is used to modulate the brightness of the beam of a cathode ray tube. The scanning of this CRT beam is synchronized with the movement of the electron beam of the microprobe. Thus an "elemental" picture of a specimen can be obtained at magnifications from 180X to 6000X. In addition, backscattered electrons from the electron-beam interaction with the scanned surface can be collected, magnified, and similarly presented on a CRT, disclosing more general characteristics of the specimens.

The results of the microprobe examination of the selected particles are shown in the Appendix. The particles shown in Appendix Figures 1a,

2a, 3a, and 4a, oscillographs produced by backscattered electrons, and the photomicrographs of the same particles (Figs. 11, 8, 12, and 13) correspond in size and shape. That Appendix Figure 5a and Figure 14 (particle A5-82) do not correspond in overall size of the particle may be because the particle broke up early in the analysis.

The X-ray oscillographs show that the particles consist mainly of plutonium, probably in the form of $^{238}\text{PuO}_2$. The plutonium particles in A5-46 and A5-67 have become attached to particles composed of iron and chromium. Particle A5-74 consists of a plutonium particle associated with a low-density particle that may be collodion polymerized by alpha radiation. The plutonium flakes in the particles appear to be pure except for a small amount of iron, as shown in Appendix Figures 1c and 5d. The specifications for $^{238}\text{PuO}_2$ microspheres permit no more than 2% impurities by weight.⁽³⁾ Colorimetric analysis of a typical group of microspheres indicated 0.35% iron. The amount of iron in the particles as indicated in the oscillographs appears to be low and, therefore, does not disagree significantly with that determined colorimetrically.

As shown in Table III, division of the equivalent $^{238}\text{PuO}_2$ spherical volumes* of the particles by their corresponding equivalent circular areas** determines the average thicknesses of the particles, which vary from 0.4 μ to 2.7 μ . In particles as thin as these there is negligible self-absorption of alpha radiation.

(3) Plutonium-238 and Polonium-210 Data Sheets, Mound Laboratory for Monsanto Research Corporation, Report No. MLM-144, p. 27, November 9, 1967.

*Equivalent $^{238}\text{PuO}_2$ spherical volumes are calculated from equivalent $^{238}\text{PuO}_2$ diameters determined by electronic counting.

**Equivalent circular areas are calculated from equivalent diameters from the Pu oscillographs, Appendix Figures 1b, 2b, 3b, 4b, and 5b.

Table III. Sizes of Radioactive Particles Analyzed by Microprobe

Particle Designation	Equivalent Spherical $^{238}\text{PuO}_2$ Measurements		Equivalent Circular Measurements		Calculated Thickness (μ)
	Diameter (μ)	Volume (μ^3)	Diameter (μ)	Area (μ^2)	
A5-45	5	65	12.0	113	0.6
A5-46	7.4	212	12.6	124	1.9
A5-67	4.8	58	8.4	55	1.1
A5-74	4.5	48	12.0	113	0.4
A5-82	8.5	322	12.6	120	2.7

4.0 CONCLUSIONS

- The radioactive particles in the $<10 \mu$ fraction of impacted samples 18 and 20 had equivalent $^{238}\text{PuO}_2$ diameters ranging from 0.2μ to 10.9μ for 18 and from 0.2μ to 3.7μ for 20.
- Radiochemical analyses of the sample fractions subjected to particle analysis indicated that particles with equivalent $^{238}\text{PuO}_2$ diameters smaller than 0.2μ did not significantly contribute to the total particulate mass for either sample.
- Optical microscopy of isolated radioactive particles showed that they were transparent and pale yellow.
- Electron microprobe analysis of isolated particles showed that they were mainly flakes of plutonium, probably PuO_2 . The oscillographs obtained by plutonium X-rays from the particles indicated equivalent circular diameters as large as 12.6μ .
- For the radioactive particles with equivalent $^{238}\text{PuO}_2$ diameters larger than 4μ , average thicknesses were calculated and found to vary from 0.4 to 2.7μ .

5.0 RECOMMENDATIONS

For future analyses similar to those described here, nuclear-emulsion exposure time should be extended so that radioactive particles with equivalent $^{238}\text{PuO}_2$ diameters as small as $0.1\ \mu$ can be counted by the hollow-star technique.

The flakes of radioactive material produced by the impaction of the microspheres would be expected to have different physical properties, such as solubility, from spherical particles with the same individual masses. Studies should be made to determine the solubility of the flakes in relation to the physiological hazard.

Figure 1. Photomicrograph of a typical hollow star produced by a radioactive particle exposed for 10 seconds to nuclear emulsion. The shim distance was $76\text{ }\mu$. It is estimated that 520 alpha tracks are in this star. This particle has an equivalent $^{238}\text{PuO}_2$ diameter of $3.7\text{ }\mu$. The magnification of the photomicrograph is 100X.

TEN ORIGINAL SLIDES FROM SAMPLE 18

Figure 2. Diagram showing microscope slides with sample 18 particles which were scanned for hollow stars. A total of 8,491 normalized particle measurements was obtained.

TEN ORIGINAL SLIDES FROM SAMPLE 20

Figure 3. Diagram showing microscope slides with sample 20 particles which were scanned for hollow stars. A total of 9,165 normalized particle measurements was obtained. The slides designated by an asterisk were not scanned.

Figure 4. Numerical size distribution of equivalent $^{238}\text{PuO}_2$ diameters of particles from sample 18. 8,491 particles were found.

Figure 5. Plot of particle diameter versus percent less than number count and mass* for equivalent $^{238}\text{PuO}_2$ particles from sample 18. The total mass for these 8,941 particles is 3.5×10^{-8} grams.

*Cumulative percent of particles (or mass in particles) smaller than the indicated size.

Figure 6. Numerical size distribution of equivalent $^{238}\text{PuO}_2$ diameters of particles from sample 20. 9,165 particles were found.

Figure 7. Plot of particle diameter versus percent less than number count and mass* for equivalent $^{238}\text{PuO}_2$ particles from sample 20. The total mass for these 9,165 particles is 5.6×10^{-8} grams.

*Cumulative percent of particles (or mass in particles) smaller than the indicated size.

Before

500X

After

500X

Figure 8. Photomicrographs of radioactive particle A5-46 from sample 20. The equivalent $^{238}\text{PuO}_2$ diameter for both before and after isolation is $7.4\ \mu$. The actual dimensions of the pale yellow, transparent portion of the particle, believed to be PuO_2 , are $8\ \mu \times 12\ \mu$.

Before

500X

After

500X

Figure 9. Photomicrographs of radioactive particle A5-D39 from sample 20. The equivalent $^{238}\text{PuO}_2$ diameters for before and after isolation are $1.1\ \mu$ and $0.9\ \mu$, respectively.

Before

500X

After

500X

Figure 10. Photomicrographs of radioactive particle A4-H43 from sample 18. The equivalent $^{238}\text{PuO}_2$ diameter of the particle both before and after isolation is $0.4\ \mu$.

200X

500X

Figure 11. Photomicrographs of isolated particle A5-45 from sample 20 as it was prepared for electron microprobe analysis. As shown in the 200X photomicrograph, an "eye" was scribed around the particle of interest to identify its location. The equivalent $^{238}\text{PuO}_2$ diameter of the particle is $5\ \mu$. The actual dimensions of the particle are $10\ \mu \times 14\ \mu$.

500X

Figure 12. Photomicrograph of isolated particle A5-67 from sample 20 as it was prepared for microprobe analysis. The equivalent $^{238}\text{PuO}_2$ diameter is $4.8\ \mu$. The actual dimensions of the particle are $16\ \mu \times 18\ \mu$.

500X

Figure 13.

Photomicrograph of isolated particle A5-74 from sample 20 as it was prepared for microprobe analysis. The equivalent $^{238}\text{PuO}_2$ diameter is $4.5\ \mu$. The actual dimensions of the particle are $8\ \mu \times 16\ \mu$.

Figure 14.

Photomicrograph of isolated particle A5-82 from sample 20 as it was prepared for microprobe analysis. The equivalent $^{238}\text{PuO}_2$ diameter is $8.5\ \mu$. The actual dimensions of this particle are $22\ \mu \times 20\ \mu$.

500X

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APPENDIX

REPORT OF ELECTRON MICROPROBE ANALYSIS OF RADIOACTIVE PARTICLES FROM SAMPLES 18 AND 20

Note: The "plastic" referred to in the appendix figures is the mounting medium, collodion, which is used to attach the particle to the bakelite disc.

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December 7, 1967

TO: Walter Holland, Tracerlab
Paul Bonnell, Mound
Ed Hansen, Sandia

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

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

SAMPLE DESIGNATION: CMB-1-CFM-Mound-PB-A-45, -46, -47, -74, -82

SAMPLE DESCRIPTION: The samples were described as plutonium-containing particles mounted with collodion on bakelite.

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

ANALYTICAL RESULTS AND DISCUSSION:

A-5-45: The particle is approximately 10 microns in diameter (Figure 1a) and contains predominantly plutonium (Figure 1b), with trace amounts of iron (Figure 1c). Silicon, chromium, manganese, nickel, copper, silver, and tungsten were not detected. Detection limits are estimated to be in the range between 0.1 and 1 percent.

A-5-46: The particle is approximately 15 microns in diameter, and there is a lower density material to the left of the particle (Figure 2a). The high density particle contains predominantly plutonium (Figure 2b) and trace amounts of silicon (Figure 2e). The low density material contains predominantly iron (Figure 2c) and chromium (Figure 2d), with some plutonium (Figure 2b), silicon (Figure 2e), and nickel (Figure 2f). Manganese, 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.

A-5-67: The sample consists of several high density particles; the largest is triangular and is approximately 10 microns wide. A low density material (Figure 3a) partially surrounds the particles. The high density particles are predominantly plutonium (Figure 3b) and contain trace amounts of silicon (Figure 3g). The low density material contains predominantly iron (Figure 3c), some plutonium (Figure 3b), chromium (Figure 3d), and nickel (Figure 3e), and trace amounts of

tungsten (Figure 3f) and silicon (Figure 3g). Manganese, copper, and silver were not detected in this sample. Detection limits are estimated to be in the range between 0.1 and 1 percent.

A-5-74: The particle is approximately 10 by 15 microns (Figure 4a) and contains only plutonium (Figure 4b). Silicon, manganese, chromium, iron, 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.

A-5-82: The sample consists of two large irregularly shaped particles and two small particles approximately 1 micron in diameter (Figure 5a). The large particles and the small particle near the center contain only plutonium (Figure 5b). The small particle in the lower right quadrant contains chromium (Figure 5c) and iron (Figure 5d). Silicon, 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.

Photomicrographs 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

NOTE: Figures 1 through 5 referred to above
are identical to Figures 1 through 5
of Appendix A of this report (pages 73
through 79) and are not reproduced here.

APPENDIX E

Walter D. Holland, TLW 6025, Particle
Analysis of Radioactive Fragments with
Diameters of Less Than $20\text{ }\mu$ From Impacted
Sample 3, Sept. 16, 1968

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PARTICLE ANALYSIS OF RADIOACTIVE FRAGMENTS
WITH DIAMETERS OF LESS THAN 20 μ FROM
IMPACTED SAMPLE 3

September 16, 1968

SANDIA CORPORATION
Box 5800
Albuquerque, New Mexico 87115

Contract No. 48-6716

Reported by

/S/ Walter D. Holland
Project Leader

Approved by

/S/ Dr. Marcel Nathans
Manager, Research and
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ABSTRACT

A representative portion of the radioactive fragments obtained by impacting 6 grams of SNAP fuel particles ($^{238}\text{PuO}_2$ microspheres) under conditions simulating atmospheric reentry and sieved to give particles $<20\ \mu$ was analyzed. Equivalent $^{238}\text{PuO}_2$ spherical diameters from $0.1\ \mu$ to $7.8\ \mu$ were determined by the hollow star and the isolation-radio-chemistry techniques. Size distribution and cumulative mass distribution are plotted for the $<20\ \mu$ sieved fraction, and a cumulative mass plot for all microspheres impacted in this experiment is presented with 13 size ranges from $<0.1\ \mu$ to $7.8\ \mu$.

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1.0 INTRODUCTION

The purpose of these studies was to determine the size distribution of fragments from SNAP fuel particles in capsules impacted under conditions simulating "intact" re-entry from the atmosphere at different velocities. This report concerns sample 3, which contained 6.0 grams of aged and thermally cycled $^{238}\text{PuO}_2$ microspheres and was impacted head-on into granite at a velocity of 104.3 ft/sec.

2.0 ANALYSIS PROCEDURE

The procedure used for this work was essentially the same as the one described in the letter report of May 28, 1968.⁽¹⁾ A representative drop of the $<20\ \mu$ sieved fraction of the impacted sample was placed on each of ten slides and sent to Tracerlab for analysis. These slides were exposed by the hollow-star technique to nuclear emulsion plates for a few seconds to locate the most active particles.⁽²⁾ These particles were enumerated, sized, and isolated into ten size ranges as shown in Table I. Each of the ten portions of particles was radiochemically analyzed to determine the total equivalent $^{238}\text{PuO}_2$ mass in each portion. Radiochemical analysis was also performed to determine the total $^{238}\text{PuO}_2$ remaining on nine of the original slides, representing the plutonium in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters of less than $2\ \mu$. This value was normalized to all ten slides. The results of the radiochemical analysis are presented in Table I.

Also included in Table I are the equivalent $^{238}\text{PuO}_2$ spherical diameters for size ranges and midranges of these measurements. These values were derived from the equivalent circular diameter size ranges. It was first

(1) Particle Analysis of Radioactive Fragments with Diameters $<10\ \mu$ from Impacted Samples 18 and 20, W. Holland, Tracerlab, A Division of Laboratory for Electronics, for Mound Laboratory, May 28, 1968.

(2) The Determination of Sizes of SNAP-9A Abort Particles Collected at High Altitudes, W. Holland and H. Volchok, (submitted to Health Physics for publication).

necessary to calculate the midrange values of the equivalent $^{238}\text{PuO}_2$ spherical diameters. The calculation of the spherical diameters and the midranges is illustrated in the Appendix.

The remaining slide that contained radioactive particles with equivalent $^{238}\text{PuO}_2$ spherical diameters of less than $2\ \mu$ was analyzed by the hollow-star technique. The other nine slides of the original ten were analyzed radiochemically as described above. The results of the three hollow-star exposures are presented in Table II. These results have been normalized for all ten slides. The equivalent $^{238}\text{PuO}_2$ mass of particles with spherical diameters $<0.1\ \mu$ is calculated as follows:

$^{238}\text{PuO}_2$ in particles on the slides from which $>2\ \mu$ particles have been isolated (See Table I)	$1.54 \times 10^{-1}\ \mu\text{g}$
$^{238}\text{PuO}_2$ in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters from 0.1 to $2.6\ \mu$ (See Table II)	$-9.8 \times 10^{-2}\ \mu\text{g}$
$^{238}\text{PuO}_2$ in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters $<0.1\ \mu$	$5.6 \times 10^{-2}\ \mu\text{g}$

3.0 DISCUSSION OF RESULTS

In Tables II and IV the values are presented that were used to plot the numerical size distribution (Figure 1) and the cumulative mass curve (Figure 2). The data from Tables I and II have been combined in Tables III and IV and used to calculate the number and mass percentages per size range. These percentages for the various size ranges are plotted in Figures 1 and 2.

The mass percent and cumulative mass percent values listed in Table IV are used to calculate the percentages of the total impacted mass (including the cumulative percentages) which are listed in Table V and plotted in Figure 3. These results are based on the $<20\ \mu$ sieved fraction that contains 0.5% of the total impacted mass, as shown by Table VI. It is interesting to compare the 0.4% of the total impacted mass as listed in Table V for particles with equivalent circular diameters $<10\ \mu$ with 0.15%, which is listed in Table VI as the portion of the total mass

in the 10 μ sieved range. Such comparison shows that there is a factor of 2.5 between the two sets of data. This is particularly significant, since in one case 70% of the mass of the <20 μ fraction (from Table VI) is in the 10 - 20 μ range, while in the other case (from Table V), only 19% of the mass is in this range.

The difference between these two values is suggested as due to (1) plutonium dioxide particles that remain on the 10 μ sieve after the residue is removed, and/or (2) an error in obtaining a representative sample of the <20 μ fraction on the ten original slides.

These errors can be seen more clearly in a more complete description of the procedure used for removing portions of the radioactive particles in the <20 μ fraction and placing them on the original ten slides. Approximately one cc of the stirred suspension of the 20 μ fraction was removed with a clean pipette, and one drop was placed on one of the ten slides. The material remaining in the pipette was put back with the original liquid. This procedure was repeated with clean pipettes to prepare the other nine slides. After the alcohol in which the particles were suspended had evaporated, a few drops of 50% collodion were added to each slide. The suspension of the <20 μ fraction was now filtered through a 10- μ filter. The weight of the PuO_2 particles which passed through the filter was determined by radiochemical analysis. The PuO_2 that remained on the 10- μ filter was washed off and its weight, representing PuO_2 in the 10 - 20 μ range, was determined by radiochemical analysis. The PuO_2 which remained on the 10 μ filter after this washing was not determined.

4.0 CONCLUSIONS

- The isolation-radiochemistry and hollow-star techniques were used successfully to obtain the numerical and mass distribution plots of PuO_2 particles with equivalent $^{238}\text{PuO}_2$ spherical diameters from $\sim 8 \mu$ to $<0.1 \mu$ for the <20 μ sieved fraction of sample 3.
- The percentages of the total mass of microspheres impacted in sample 3 are calculated for 13 size ranges from 7.8 to $<0.1 \mu$ (equivalent $^{238}\text{PuO}_2$ spherical diameters).

5.0 RECOMMENDATION

An attempt to resolve the difference between considering the results on a $<20\ \mu$ sieve basis and on a $<10\ \mu$ sieve basis should be made. (See the last paragraph of Section 3.0.)

TABLE I RESULTS OF RADIOCHEMICAL ANALYSIS

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Equivalent Circular Diameter Size Range (μ)	Number of Particles per Size Range	Equivalent $^{238}\text{PuO}_2$ Mass per Size Range (μg)
Size Ranges (μ)	Midranges (μ)			
--	--	< 2.0	--	1.54×10^{-1}
0.70 - 2.14	1.42	2 - 4	223	3.83×10^{-3}
2.14 - 3.63	2.39 } 2.86	4 - 6	58	4.70×10^{-3}
	3.33 }	6 - 8	28	6.17×10^{-3}
3.63 - 5.31	4.01 } 4.40	8 - 10	21	8.11×10^{-3}
	4.78 }	10 - 12	14	9.18×10^{-3}
5.31 - 6.75	6.21 } 6.21	12 - 14	7	1.00×10^{-2}
	6.25 }	14 - 16	4	5.87×10^{-3}
	6.17 }	16 - 18	2	2.83×10^{-3}
6.75 - 7.83	7.50 } 7.29	18 - 23	7	1.77×10^{-2}
	7.08 }	23 - 26	3	6.40×10^{-3}
		Totals	367	2.28×10^{-1}

TABLE II NORMALIZED RESULTS OF MEASURING SAMPLE 3
PARTICLES BY THE HOLLOW STAR TECHNIQUE

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Number Per Size Range	Equivalent $^{238}\text{PuO}_2$ Mass Per Size Range (10^{-10} g)
Size Range (μ)	Midrange (μ)		
0.10 - 0.14	0.12	116,480	12.11
0.14 - 0.21	0.175	148,480	47.81
0.21 - 0.30	0.255	57,600	57.31
0.30 - 0.42	0.36	27,084	75.84
0.42 - 0.61	0.515	32,707	268.20
0.61 - 0.88	0.745	11,720	290.66
0.88 - 1.3	1.09	2,380	184.93
1.3 - 1.8	1.55	120	26.81
1.8 - 2.6	2.20	20	12.78
Totals		396,591	976.45

TABLE III NUMERICAL PERCENTAGES PER SIZE RANGE
FOR RADIOACTIVE PARTICLES IN THE LESS
THAN 20 μ SIEVED FRACTION OF SAMPLE 3

Equivalent ²³⁸ PuO ₂ Spherical Diameters		Equiv. Circular Diameter Size Ranges* (μ)	No. of Particles Per Size Range	Frequency Percent Per Size Range	Cumulative Numerical Percent Per Size Range
Size Ranges (μ)	Midranges (μ)				
0.1 - 0.14	0.12		116,480	29.34	29.34
0.14 - 0.21	0.175		148,480	37.40	66.74
0.21 - 0.30	0.255		57,600	14.51	81.25
0.30 - 0.42	0.36		27,084	6.82	88.07
0.42 - 0.61	0.515		32,707	8.24	96.31
0.61 - 0.88	0.745		11,720	2.95	99.26
0.88 - 1.30	1.09		2,380	6.0x10 ⁻¹	99.86
1.30 - 2.34	1.55 } 2.20 } 1.72 1.42 }	2 - 4	120) 20) 363 223)	9.1x10 ⁻²	99.89
2.34 - 3.63	2.39 } 3.33 } 2.86	4 - 6 6 - 8	58) 86 28)	2.2x10 ⁻²	99.97
3.63 - 5.31	4.01 } 4.78 } 4.40	8 - 10 10 - 12	21) 35 14)	8.8x10 ⁻³	99.979
5.31 - 6.75	6.21 } 6.25 } 6.21 6.17 }	12 - 14 14 - 16 16 - 18	7) 4) 13 2)	3.3x10 ⁻³	99.982
6.75 - 7.83	7.50 } 7.08 } 7.29	18 - 21 23 - 26	7) 10 3)	2.5x10 ⁻³	99.984
		Total 396,958			
* It was not possible to determine without extreme financial expense the equivalent circular diameters of particles measured by the hollow star technique.					

TABLE IV MASS PERCENTAGES PER SIZE RANGE FOR
RADIOACTIVE PARTICLES IN THE LESS
THAN 20 μ SIEVED FRACTION OF SAMPLE 3

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Equiv. Circular Diameter Size Ranges* (μ)	Mass per Size Range (10^{-10} g)	Mass Percent Per Size Range	Cumulative Mass % Per Size Range
Size Ranges (μ)	Midranges (μ)				
<0.1	<0.1		560	24.5	24.5
0.1 - 0.14	0.12		12.11	0.5	25.0
0.14 - 0.21	0.175		47.81	2.1	27.1
0.21 - 0.30	0.255		57.31	2.5	29.6
0.30 - 0.42	0.36		75.84	3.3	32.9
0.42 - 0.61	0.515		268.20	11.7	44.6
0.61 - 0.88	0.745		290.66	12.7	57.3
0.88 - 1.30	1.09		184.93	8.1	65.4
1.3 - 2.34	1.55) 2.20 } 1.72 1.42)	2 - 4	26.81) 12.78) 77.8 38.3)	3.2	68.8
2.34 - 3.63	2.39) 3.33 } 2.86	4 - 6 6 - 8	47.0) 108.7 61.7)	4.8	73.6
3.63 - 5.31	4.01) 4.78 } 4.40	8 - 10 10 - 12	81.1) 173 91.8)	7.6	81.2
5.31 - 6.75	6.21) 6.25 } 6.21 6.17)	12 - 14 14 - 16 16 - 18	100.0) 58.7) 186 28.3)	8.1	89.3
6.75 - 7.83	7.50) 7.08 } 7.29	18 - 23 23 - 26	177.0) 240 64.0)	10.5	99.8
		Total	2,284.05		

TABLE V MASS PERCENTAGES OF TOTAL IMPACTED MICROSPHERES
IN THE <20 μ SIEVED SIZE RANGES

Equivalent $^{238}\text{PuO}_2$ Spherical Diameter Size Ranges (μ)	Equivalent Circular Diameter Size Ranges (μ)	Mass % Per Size Range	Cumulative Mass % Per Size Range	Basis - 0.5% of Microspheres Impacted are in < 20 μ Sieved Fraction	
				Mass % Per Size Range	Cumulative % Mass
< 0.1		24.5	24.5	1.2×10^{-1}	1.2×10^{-1}
0.1 - 0.14		0.5	25.0	2.5×10^{-3}	1.2×10^{-1}
0.14 - 0.21		2.1	27.1	1.0×10^{-2}	1.3×10^{-1}
0.21 - 0.30		2.5	29.6	1.2×10^{-2}	1.4×10^{-1}
0.30 - 0.42		3.3	32.9	1.6×10^{-2}	1.6×10^{-1}
0.42 - 0.61		11.7	44.6	5.8×10^{-2}	2.2×10^{-1}
0.61 - 0.88		12.7	57.3	6.4×10^{-2}	2.8×10^{-1}
0.88 - 1.30		8.1	65.4	4.0×10^{-2}	3.2×10^{-1}
1.30 - 2.34	2 - 4	3.2	68.8	1.7×10^{-2}	3.4×10^{-1}
2.34 - 3.63	4 - 6 6 - 8	4.8	73.6	2.4×10^{-2}	3.6×10^{-1}
3.63 - 5.31	8 - 10 10 - 12	7.6	81.2	3.8×10^{-2}	4.0×10^{-1}
5.31 - 6.75	12 - 14 14 - 16 16 - 18	8.1	89.3	4.0×10^{-2}	4.4×10^{-1}
6.75 - 7.83	18 - 23 23.0 - 26	10.5	99.8	5.2×10^{-2}	4.9×10^{-1}
<p>Note: The total equivalent $^{238}\text{PuO}_2$ mass for particles having equivalent circular diameters < 12 μ is 0.4%. This is a factor of 2.5 greater than the 0.15% found by Mound Laboratory.</p>					

TABLE VI RESULTS OF SIEVING THE MICROSPHERES
IN SAMPLE 3 BEFORE AND AFTER IMPACTION

Sieved Size Range (μ)	Before Impaction		After Impaction	
	Mass %	Cumulative %	Mass %	Cumulative
177-210	16.7	100.1	13.0	96.5
149-177	16.7	83.4	16.5	83.5
125-149	20	66.7	17.8	67.0
105-125	16.7	46.7	15.5	49.2
63-105	25	30	23.2	33.7
44-63	5	5	7.5	10.5
20-44	---	----	2.5	3.0
10-20	---	----	0.35	5.0×10^{-1}
<10	---	----	0.15	1.5×10^{-1}

Figure 1 Size distribution histogram of the 396,958 particles found from sample 3. The arithmetic mean diameter is $\sim 0.2 \mu$.

Figure 2 Cumulative plot of the mass percentages of radioactive particles from sample 3. A total equivalent $^{238}\text{PuO}_2$ mass was 0.228 μg . The mass median diameter is $\sim 0.7 \mu$.

Figure 3 Cumulative plot of mass percentages of all microspheres impacted in sample 3. The mass median diameter is 120 μ .

APPENDIX

CALCULATION OF EQUIVALENT $^{238}\text{PuO}_2$ SPHERICAL DIAMETER SIZE RANGES AND MIDRANGES

The calculation of equivalent $^{238}\text{PuO}_2$ spherical diameter size ranges and midranges is illustrated by the following example, which determines these values for the equivalent circular diameter size range of 23 - 26 μ . (See Table I.)

The mass of $^{238}\text{PuO}_2$ in this size range is divided by its density to obtain the volume of PuO_2 particles in this range.

$$\frac{6.40 \times 10^{-9} \text{ g}}{11.46 \text{ g/cc}} = 5.6 \times 10^{-10} \text{ cc}$$

To calculate the average volume for particles in this size range, 5.6×10^{-10} cc is divided by the number of particles in this range.

$$\frac{5.6 \times 10^{-10} \text{ cc}}{3} = 1.9 \times 10^{-10} \text{ cc/particle}$$

For the Volume of Sphere,

$$\text{Volume} = 0.524 d^3, \text{ where } d \text{ is the diameter}$$

$$d = \left(\frac{1.9 \times 10^{-10}}{0.524} \right)^{1/3} = (3.60 \times 10^{-12})^{1/3}$$

$$= 7.08 \times 10^{-4} \text{ cm or } 7.08 \mu, \text{ which is midrange, corresponding to the } 23 - 26 \mu \text{ equivalent circular diameter size range}$$

Calculation of the equivalent $^{238}\text{PuO}_2$ spherical diameter size ranges for this midrange is now shown. The differences

between each of the midpoints were determined as illustrated by:

$$\begin{array}{r} 7.29 \\ - \underline{6.21} \\ 1.08 \div 2 = 0.54 \end{array}$$

The lower limit of this range was calculated by:

$$\begin{array}{r} 7.29 \\ - \underline{0.54} \\ 6.75 \end{array}$$

The upper limit was calculated by:

$$\begin{array}{r} 7.29 \\ + \underline{0.54} \\ 7.83 \end{array}$$

APPENDIX F

Walter D. Holland, TLW 6026, Particle
Analysis of Radioactive Fragments with
Diameters of Less Than 20 μ From Impacted
Sample 5, Sept. 17, 1968

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PARTICLE ANALYSIS OF RADIOACTIVE FRAGMENTS
WITH DIAMETERS OF LESS THAN 20 μ FROM
IMPACTED SAMPLE 5

September 17, 1968

SANDIA CORPORATION
Box 5800
Albuquerque, New Mexico 87115

Contract No. 48-6716

Reported by

/S/ Walter D. Holland
Project Leader

Approved by

/S/ Dr. Marcel Nathans
Manager, Research and
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A Division of Laboratory for Electronics, Inc.
2030 Wright Avenue
Richmond, California 94804

ABSTRACT

A representative portion of the radioactive fragments obtained by impacting 6 grams of SNAP fuel particles ($^{238}\text{PuO}_2$ microspheres) under conditions simulating intact atmospheric reentry and sieved to give particles $<20\text{ }\mu$ was analyzed. Equivalent $^{238}\text{PuO}_2$ spherical diameters from $0.1\text{ }\mu$ to $2.3\text{ }\mu$ were determined by the hollow-star and the isolation-radiochemistry techniques. Size distribution and cumulative mass distribution are plotted for the $<20\text{ }\mu$ sieved fraction, and a cumulative mass plot for all microspheres impacted in this experiment is presented with nine ranges from $<0.1\text{ }\mu$ to $2.3\text{ }\mu$.

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1.0 INTRODUCTION

The purpose of these studies was to determine the size distribution of fragments from SNAP fuel particles in capsules impacted under conditions simulating "intact" reentry from the atmosphere at different velocities. This report concerns sample 5, which contained 6.0 grams of aged and thermally cycled $^{238}\text{PuO}_2$ microspheres and was impacted head-on into granite at a velocity of 102.5 ft/sec.

2.0 ANALYSIS PROCEDURE

The procedure used for this work was essentially the same as the one already described.^(1,2) A representative drop of the $<20\ \mu$ sieved fraction of the impacted sample was placed on each of ten slides and sent to Tracerlab for analysis. These slides were exposed by the hollow-star technique to nuclear emulsion plates for a few seconds to locate the most active particles.⁽³⁾ These particles were enumerated, sized, and isolated into ten size ranges as shown in Table I. Each of the ten portions of particles was radiochemically analyzed to determine the total equivalent $^{238}\text{PuO}_2$ mass in each portion. Radiochemical analysis was also performed to determine the total $^{238}\text{PuO}_2$ remaining on nine of the original slides, representing the plutonium in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters of less than $2\ \mu$. This value was normalized to all ten slides. The results of this radiochemical analysis are presented in Table I.

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- (1) Particle Analysis of Radioactive Fragments with Diameters $<10\ \mu$ from Impacted Samples 18 and 20, W. Holland, Tracerlab, A Division of Laboratory for Electronics, for Mound Laboratory, May 28, 1968.
 - (2) Particle Analysis of Radioactive Fragments with Diameters $<20\ \mu$ from Impacted Sample 3, W. Holland, Tracerlab, A Division of Laboratory for Electronics, for Sandia Corporation, September 16, 1968.
 - (3) The Determination of Sizes of SNAP-9A Abort Particles Collected at High Altitudes, W. Holland and H. Volchok (submitted to Health Physics for publication).

Also included in Table I are the equivalent $^{238}\text{PuO}_2$ spherical diameters for size ranges and midranges of these measurements. These values were derived from the equivalent circular diameter size ranges. It was first necessary to calculate the midrange values of the equivalent $^{238}\text{PuO}_2$ spherical diameters. The calculation of the spherical diameters and the midranges is illustrated in the Appendix.

The remaining slide which contained radioactive particles with equivalent $^{238}\text{PuO}_2$ spherical diameters of less than $2\ \mu$ was analyzed by the hollow star technique. The other nine slides of the original ten were analyzed radiochemically as described above. The results of the three hollow-star exposures are presented in Table II. These results have been normalized for all ten slides. The equivalent $^{238}\text{PuO}_2$ mass of particles with spherical diameters $<0.1\ \mu$ is calculated as follows:

$^{238}\text{PuO}_2$ in particles on the slides from which $>2\ \mu$ particles have been isolated (see Table I)	$1.638 \times 10^{-1}\ \mu\text{g}$
$^{238}\text{PuO}_2$ in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters from 0.1 to $2.6\ \mu$ (see Table II)	$- 0.971 \times 10^{-1}\ \mu\text{g}$
$^{238}\text{PuO}_2$ in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters $<0.1\ \mu$	$0.667 \times 10^{-1}\ \mu\text{g}$

3.0 DISCUSSION OF RESULTS

In Tables III and IV the values are presented that were used to plot the numerical size distribution (Figure 1) and the cumulative mass curve (Figure 2). The data from Tables I and II have been combined in Tables III and IV and used to calculate the number and mass percentages per size range. These percentages for the various size ranges are plotted in Figures 1 and 2. Figure 1 shows that the radioactive particles in the $<20\ \mu$ sieved fraction have equivalent $^{238}\text{PuO}_2$ spherical diameters from $<0.1\ \mu$ to $2.3\ \mu$. Figure 2 shows that 40% of the equivalent $^{238}\text{PuO}_2$ mass is in particles with equivalent spherical diameters of $<0.1\ \mu$. The mass median diameter of radioactive particles in the $<20\ \mu$ sieved fraction is $\sim 0.34\ \mu$.

The mass percent and cumulative mass percent values listed in Table IV are used to calculate the percentages of the total impacted mass (including the cumulative percentages) which are listed in Table V and plotted in Figure 3. These results are based on the $<10\ \mu$ sieved fraction that contains 0.15% of the total impacted mass, as shown by Table VI. Figure 3 indicates that the mass mean diameter of the sample 5 microspheres after impactation (including the fragments produced) is $\sim 120\ \mu$ and that $6 \times 10^{-2}\%$ of the mass is in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters $<0.1\ \mu$. Even though the sample analyzed was a portion of the $<20\ \mu$ sieved fraction, the data were treated as from a $<10\ \mu$ sieved fraction because only two particles with equivalent circular diameters $>10\ \mu$ were found. It is evident from the data in Table VI that the equivalent mass of particles in the 10 to $20\ \mu$ fraction should be $\sim 77\%$ of the equivalent mass in the $<20\ \mu$ fraction. The two particles in the 10 to $20\ \mu$ equivalent circular diameter range represent only 0.1% of the equivalent mass of the $<20\ \mu$ fraction. This absence of particles with equivalent circular diameters between 10 and $20\ \mu$ is considered due to errors in aliquoting the $<20\ \mu$ sieved fraction and/or in weighing the residue on the $10\ \mu$ filter. Both these errors were discussed in an earlier report.⁽²⁾

4.0 CONCLUSIONS

- The isolation-radiochemistry and hollow-star techniques were successfully used to obtain the numerical and mass distribution plots of PuO_2 particles with equivalent $^{238}\text{PuO}_2$ spherical diameters from $\sim 2.29\ \mu$ to $<0.1\ \mu$ for the $<20\ \mu$ sieved fraction of sample 5.
- The percentages of the total mass of microspheres impacted in sample 5 are calculated for nine size ranges from $2.3\ \mu$ to $<0.1\ \mu$ (equivalent $^{238}\text{PuO}_2$ spherical diameters).

5.0 RECOMMENDATION

- An attempt to correct the problem caused by an apparent absence of particles in the 10 to $20\ \mu$ equivalent circular diameter range for the $<20\ \mu$ fraction should be made. (See last paragraph of Section 3.0.)

TABLE I Results of Radiochemical Analysis - Sample 5

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Equiv. Circular Diameter Size Range (μ)	Number of Particles per Size Range	Equiv. $^{238}\text{PuO}_2$ Mass per Size Range (μg)
Size Range (μ)	Midrange (μ)			
-	-	<2.0	-	1.64×10^{-1}
1.08 - 1.58	1.30 } 1.335 1.37 }	2.0 - 4.0 4.0 - 6.0	254 151	3.37×10^{-3} 2.34×10^{-3}
1.58 - 1.98	1.72 } 1.84 1.96 }	6.0 - 8.0 8.0 - 10.0	30 7	9.13×10^{-4} 3.15×10^{-4}
1.98 - 2.26	2.11 } 2.115 2.12 }	10.0 - 12.0 12.0 - 14.0	1 1	5.66×10^{-5} 5.72×10^{-5}
		Totals	444	1.71×10^{-1}

TABLE II Normalized Results of Measuring Sample 5
Particles by the Hollow Star Technique

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Number per Size Range	Equiv. $^{238}\text{PuO}_2$ Mass per Size Range (10^{-10} gms)
Size Range (μ)	Midrange (μ)		
0.1 - 0.14	0.12	20,480	2.09
0.14 - 0.21	0.175	75,520	24.47
0.21 - 0.30	0.255	78,080	77.77
0.30 - 0.42	0.36	45,440	127.30
0.42 - 0.61	0.515	27,200	222.90
0.61 - 0.88	0.745	11,760	291.80
0.88 - 1.3	1.09	2,000	155.40
1.3 - 1.8	1.55	280	62.55
1.8 - 2.6	2.20	10	6.39
		Totals	260,770
			970.7

TABLE III Numerical Percentages per Size Range
for Radioactive Particles in the Less
Than 20 μ Sieved Fraction of Sample 5

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Equiv. Circular Diameter Size Ranges (μ)	No. of Particles per Size Range	Frequency Percent per Size Range	Cumulative Numerical Percent per Size Range
Size Ranges (μ)	Midranges (μ)				
0.1 - 0.14	0.12		20,480	7.84	7.84
0.14 - 0.21	0.175		75,520	28.91	36.75
0.21 - 0.30	0.255		78,080	29.89	66.64
0.30 - 0.42	0.36		45,440	17.40	84.04
0.42 - 0.61	0.515		27,200	10.41	94.45
0.61 - 0.88	0.745		11,760	4.50	98.95
0.88 - 1.59	1.09	2.0 - 4.0 4.0 - 6.0	2,000	0.92	99.87
	1.30		254		
	1.37		151		
1.59 - 2.29	1.55	6.0 - 8.0 8.0 - 10.0 10.0 - 12.0 12.0 - 14.0	280	0.12	99.99
	1.72		30		
	1.96		7		
	2.11		1		
	2.12		1		
	2.20		10		
		Total		261,214	

TABLE IV Mass Percentages per Size Range for
Radioactive Particles in the Less
Than 20 μ Sieved Fraction of Sample 5

Equivalent ²³⁸ PuO ₂ Spherical Diameters		Equiv. Circular Diameter Size Ranges (μ)	Mass per Size Range (10 ⁻¹⁰ g)	Mass Percent per Size Range	Cumulative Mass Percent per Size Range		
Size Ranges (μ)	Midranges (μ)						
<0.1	<0.1		667	40.7	40.7		
0.1 - 0.14	0.12		2.09	0.1	40.8		
0.14 - 0.21	0.175		24.47	1.5	42.3		
0.21 - 0.30	0.255		77.77	4.7	47.0		
0.30 - 0.42	0.36		127.30	7.8	54.8		
0.42 - 0.61	0.515		222.90	13.6	68.4		
0.61 - 0.88	0.745		291.80	17.8	86.2		
0.88 - 1.59	1.09 } 1.30 } 1.37 }	2.0 - 4.0 4.0 - 6.0	155.40 3.37x10 ⁻³ 2.34x10 ⁻³ }	155.4 9.5	95.7		
	1.59 - 2.29		1.55 } 1.75 } 1.96 } 2.11 } 2.12 } 2.20 }			62.55 9.13x10 ⁻⁴ 3.15x10 ⁻⁴ 5.66x10 ⁻⁵ 5.72x10 ⁻⁵ 6.39 }	68.94 4.3
Total		1,638					

TABLE V Mass Percentages of Total Impacted
Microspheres in the <20 μ Sieved
Size Ranges

Equivalent $^{238}\text{PuO}_2$ Spherical Diameter Size Ranges (μ)	Equivalent Circular Diameter Size Ranges (μ)	Mass Percent per Size Range	Cumulative Mass Percent per Size Range	Basis - 0.15% of Micro- spheres Impacted are in <10 μ Sieved Fraction	
				Mass Percent per Size Range	Cumulative Percent Mass
<0.1		40.7	40.7	6.0×10^{-2}	0.06
0.1 - 0.14		0.1	40.8	1.9×10^{-4}	0.06
0.14 - 0.21		1.5	42.3	2.2×10^{-3}	0.06
0.21 - 0.30		4.7	47.0	7.1×10^{-3}	0.07
0.30 - 0.42		7.8	54.8	1.2×10^{-2}	0.08
0.42 - 0.61		13.6	68.4	2.0×10^{-2}	0.10
0.61 - 0.88		17.8	86.2	3.0×10^{-2}	0.13
0.88 - 1.59	2.0 - 4.0 4.0 - 6.0	9.5	95.7	1.4×10^{-2}	0.14
1.59 - 2.29	6.0 - 8.0 8.0 - 10.0 10.0 - 12.0 12.0 - 14.0	4.3	100	6.4×10^{-3}	0.15
<p>Note: Our findings indicated that essentially no particles were in the equivalent circular range from 10-20 μ. Mound found approximately 77% of the mass of the < 20μ sieved fraction was in the 10-20 μ range (see Table VI).</p>					

TABLE VI Results of Sieving the Microspheres in
Sample 5 Before and After Impaction

Sieved Size Range (μ)	Before Impaction		After Impaction	
	Mass Percent	Cumulative Percent	Mass Percent	Cumulative Percent
177 - 210	16.7	100.1	12.8	95.65
149 - 177	16.7	83.4	14.5	82.85
125 - 149	20	66.7	17.8	68.35
105 - 125	16.7	46.7	14.5	50.55
63 - 105	25	30	25.1	36.05
44 - 63	5	5	7.2	10.95
20 - 44	-	-	3.1	3.75
10 - 20	-	-	0.5	0.65
<10	-	-	0.15	0.15

Figure 1. Size Distribution Histogram of the 261,214 Particles Found From Sample 5. The Arithmetic Mean Diameter is $\sim 0.16 \mu$.

Figure 2. Cumulative Plot of the Mass Percentages of Radioactive Particles from Sample 5. The Total Equivalent $^{238}\text{PuO}_2$ Mass was $1.71 \times 10^{-1} \mu\text{g}$. The Mass Median Diameter is $\sim 0.34 \mu$.

Figure 3. Cumulative Plot of Mass Percentages of All Microspheres Impacted in Sample 5. The Mass Median Diameter is $\sim 120 \mu$.

APPENDIX

CALCULATION OF EQUIVALENT $^{238}\text{PuO}_2$ SPHERICAL DIAMETER SIZE RANGES AND MIDRANGES

The calculation of equivalent $^{238}\text{PuO}_2$ spherical diameter size ranges and midranges is illustrated by the following example, which determines these values for the equivalent circular diameter size range of 12 to 14 μ . (See Table I.)

The mass of $^{238}\text{PuO}_2$ in this size range is divided by its density to obtain the volume of PuO_2 particles in this range.

$$\frac{5.72 \times 10^{-11} \text{ g}}{11.46 \text{ g/cc}} = 4.99 \times 10^{-12} \text{ cc}$$

To calculate the average volume for particles in this size range, $4.99 \times 10^{-12} \text{ cc}$ is divided by the number of particles in this range.

$$\frac{4.99 \times 10^{-12}}{1} = 4.99 \times 10^{-12} \text{ cc/particle}$$

For the Volume of the Sphere

$$\text{Volume} = 0.524 d^3, \text{ where } d \text{ is the diameter}$$

$$d = \left(\frac{4.99 \times 10^{-12}}{0.524} \right)^{1/3} = (9.52 \times 10^{-12})^{1/3}$$

$$= 2.12 \times 10^{-4} \text{ cm or } 2.12 \mu, \text{ which is midrange, corresponding to the 12 to } 24 \mu \text{ equivalent circular diameter size range}$$

Calculation of the equivalent $^{238}\text{PuO}_2$ spherical diameter size ranges for this midrange is now shown. The differences between

each of the midpoints was determined as illustrated by:

$$\begin{array}{r} 2.115 \\ - 1.84 \\ \hline 0.275 \div 2 = 0.14 \end{array}$$

The lower limit of this range was calculated by:

$$\begin{array}{r} 2.115 \\ - 0.14 \\ \hline 1.975 \text{ or } 1.98 \text{ (See Table I)} \end{array}$$

The upper limit was calculated:

$$\begin{array}{r} 2.115 \\ + 0.14 \\ \hline 2.255 \text{ or } 2.26 \text{ (See Table I)} \end{array}$$

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APPENDIX G

Letter dated October 23, 1968, from
Walter D. Holland, Tracerlab, to Walter Dalby,
Sandia Corporation, reporting the results of
Tracerlab analysis of sample 15

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TRACERLAB

A DIVISION OF LABORATORY FOR ELECTRONICS INC

2030 WRIGHT AVENUE • RICHMOND CALIF 94804

October 23, 1968

235-2633

CABLE ADDRESS
LFE

Ref: Contract No. 48-6716
TLW #12762

Mr. Walter Dalby
Division 9312
Sandia Corporation
P.O. Box 5800
Albuquerque, New Mexico

Dear Mr. Dalby:

We have completed the analysis of the particulate fragments in the less than $10\ \mu$ sieved fraction of sample 15 which was impacted at 456.1 fps. Capsule 15 was impacted into a concrete target with a $3/4$ inch thick piece of Hitco graphite taped over the target area. (1)

The analysis procedure used was the same as described in earlier reports. (2)(3) The results of radiochemical analyses of the sized batches of radioactive particles having equivalent circular diameters larger than $2\ \mu$ are presented as Table I. Also included in Table I are the equivalent $^{238}\text{PuO}_2$ spherical diameters for the size ranges and mid-ranges of these measurements. The results of the hollow-star exposures for particles having equivalent $^{238}\text{PuO}_2$ spherical diameters between about 2 and $0.1\ \mu$ are included as Table II. The equivalent $^{238}\text{PuO}_2$ mass of particles having spherical diameters less than $0.1\ \mu$ is calculated as follows:

-
- (1) Analysis of Fines from Impact Capsule 15, P. H. Bonnell and R. E. Vallee, Monsanto Research Corporation, a Subsidiary of Monsanto Company, for Atomic Energy Commission, September 26, 1968.
 - (2) Particle Analysis of Radioactive Fragments with Diameters $<20\ \mu$ from Impacted Sample 3, W. Holland, Tracerlab, a Division of Laboratory for Electronics, for Sandia Corporation, Sept. 16, 1968.
 - (3) Particle Analysis of Radioactive Fragments with Diameters $<20\ \mu$ from Impacted Sample 5, W. Holland, Tracerlab, a Division of Laboratory for Electronics, for Sandia Corporation, Sept. 17, 1968.

Mr. Walter Dalby

-2-

October 23, 1968

$^{238}\text{PuO}_2$ in particles on the slides from which $>2\ \mu$ particles have been isolated (see Table I)	$2.96 \times 10^{-2}\ \mu\text{g}$
$^{238}\text{PuO}_2$ in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters from 0.1 to $2.6\ \mu$ (see Table II)	$- 1.46 \times 10^{-2}\ \mu\text{g}$
$^{238}\text{PuO}_2$ in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters $<0.1\ \mu$	$1.50 \times 10^{-2}\ \mu\text{g}$

In Table III the values are presented that were used to plot the numerical distribution (Figure 1). In Table IV the mass percentages per size range are presented for radioactive particles in the less than $10\ \mu$ sieved fraction of sample 15. Figure 1 shows that the radioactive particles in the less than $10\ \mu$ sieved fraction have equivalent $^{238}\text{PuO}_2$ spherical diameters from $<0.1\ \mu$ to $7.46\ \mu$.

The mass percent and cumulative mass percent values listed in Table IV are used to calculate the percentages of the total impacted mass (including the cumulative percentages) which are listed in Tables V and VI and plotted in Figure 2. These results are based on the less than $10\ \mu$ sieved fraction containing 12.72% of the total impacted mass, as shown by Table VI. Figure 2 indicates that the mass mean diameter of the sample 15 microspheres after impaction (including the fragments produced) is $\sim 44\ \mu$ and that 2.1% of the mass is in particles with equivalent $^{238}\text{PuO}_2$ spherical diameters less than $0.1\ \mu$.

Should you have any questions or comments, please contact me.

Very truly yours,

/S/ Walter Holland, Supervisor
Physical Measurements Laboratory

/sfb

cc: Paul Bonnell

TABLE I RESULTS OF RADIOCHEMICAL ANALYSIS

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Equivalent Circular Diam. Per Size Range (μ)	Number of Particles Per Size Range	Equivalent $^{238}\text{PuO}_2$ Mass Per Size Range (μg)
Size Ranges (μ)	Mid-Ranges (μ)			
-	-	<2	-	2.96×10^{-2}
0.70-2.08	1.39	2-4	500	8.12×10^{-3}
2.08-3.50	2.78	4-6	63	8.12×10^{-3}
3.50-4.81	4.21	6-8	24	10.81×10^{-3}
4.81-5.92	5.41	8-10	16	15.20×10^{-3}
5.92-6.78	6.44	10-12	7	11.19×10^{-3}
6.78-7.46	7.12	12-15	4	8.68×10^{-3}
		TOTALS	614	62.12×10^{-3}

TABLE II NORMALIZED RESULTS OF MEASURING SAMPLE 15
PARTICLES BY THE HOLLOW STAR TECHNIQUE

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Number Per Size Range	Equivalent $^{238}\text{PuO}_2$ Mass Per Size Range (10^{-10} g)
Size Ranges (μ)	Mid-Ranges (μ)		
0.10-0.14	0.12	6,656	0.6909
0.14-0.21	0.175	7,584	2.6529
0.21-0.30	0.255	4,640	4.8998
0.30-0.42	0.36	3,648	10.1998
0.42-0.61	0.515	3,448	29.0873
0.61-0.88	0.745	1,720	41.8166
0.88-1.3	1.09	644	51.5928
1.3 -1.8	1.55	0	0.0
1.8 -2.6	2.20	8	5.1110
TOTALS		28,368	146.0512

TABLE III NUMERICAL PERCENTAGES PER SIZE RANGE
FOR RADIOACTIVE PARTICLES IN THE LESS
THAN 10 μ SIEVED FRACTION OF SAMPLE 15

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Equivalent Circular Dia. Size Ranges* (μ)	No. of Particles Per Size Range	Frequency Percent Per Size Range	Cumulative Numerical Percent Per Size Range
Size Ranges (μ)	Mid-Ranges (μ)				
0.1-0.14	0.12		6,656	22.97	22.97
0.14-0.21	0.175		7,584	26.17	49.14
0.21-0.30	0.255		4,640	16.01	65.15
0.30-0.42	0.36		3,648	12.59	77.74
0.42-0.61	0.515		3,448	11.90	89.64
0.61-0.88	0.745		1,720	5.93	95.57
0.88-1.30	1.09		644	2.29	97.86
1.30-2.24	1.55 2.20 1.39	2-4	0 8 500	1.75	99.61
2.24-3.50	2.78	4-6	63	0.22	99.83
3.50-4.81	4.21	6-8	24	0.08	99.91
4.81-5.92	5.41	8-10	16	0.06	99.97
5.92-6.78	6.44	10-12	7	0.02	99.99
6.78-7.46	7.12	12-15	4	0.01	100.00
		TOTAL	28,982		

* It was not possible to determine without extreme financial expense the equivalent circular diameters of particles measured by hollow star technique.

TABLE IV MASS PERCENTAGES PER SIZE RANGE FOR
RADIOACTIVE PARTICLES IN THE LESS THAN
10 μ SIEVED FRACTION OF SAMPLE 15

Equivalent $^{238}\text{PuO}_2$ Spherical Diameters		Equivalent Circular Dia. Size Ranges* (μ)	Mass Per Size Range (10^{-10} g)	Mass Percent Per Size Range	Cumulative Mass Percent Per Size Range
Size Ranges (μ)	Mid-Ranges (μ)				
<0.1	<0.1		150	16.35	16.35
0.1-0.14	0.12		0.69	0.08	16.43
0.14-0.21	0.175		2.65	0.29	16.72
0.21-0.30	0.255		4.90	0.53	17.25
0.30-0.42	0.36		10.20	1.12	18.37
0.42-0.61	0.515		29.09	3.17	21.54
0.61-0.88	0.745		41.82	4.56	26.10
0.88-1.30	1.09		51.59	5.62	31.72
1.30-2.24	1.55) 2.20) 1.71 1.39)	2-4	0.00) 5.11) 86.31 81.20)	9.41	41.13
2.24-3.50	2.78	4-6	81.2	8.85	49.98
3.50-4.81	4.21	6-8	108.1	11.79	61.77
4.81-5.92	5.41	8-10	152.0	16.57	78.34
5.92-6.78	6.44	10-12	111.9	12.20	90.54
6.78-7.46	7.12	12-15	86.8	9.46	100.00
		TOTAL	917.25		

TABLE V MASS PERCENTAGES OF TOTAL IMPACTED MICROSPHERES
IN THE $<10\ \mu$ SIEVED SIZE RANGES

Equiv. $^{238}\text{PuO}_2$ Spherical Diam. Size Ranges (μ)	Equiv. Circular Diameter Size Ranges (μ)	Mass % Per Size Range	Cumulative Mass % Per Size Range	Basis - 12.7% of Micro- spheres Impacted are in <10 μ Sieved Fraction	
				Mass % Per Size Range	Cumulative % Mass
<0.1		16.35	16.35	2.08	2.08
0.1-0.14		0.08	16.43	1.02×10^{-2}	2.09
0.14-0.21		0.29	16.72	3.69×10^{-2}	2.12
0.21-0.30		0.53	17.25	6.74×10^{-2}	2.19
0.30-0.42		1.12	18.37	1.42×10^{-1}	2.34
0.42-0.61		3.17	21.54	4.03×10^{-1}	2.74
0.61-0.88		4.56	26.10	5.80×10^{-1}	3.32
0.88-1.30		5.62	31.72	7.15×10^{-1}	4.03
1.30-2.24	2-4	9.41	41.13	1.20	5.23
2.24-3.50	4-6	8.85	49.98	1.12	6.36
3.50-4.81	6-8	11.79	61.77	1.50	7.86
4.81-5.92	8-10	16.57	78.34	2.11	9.96
5.92-6.78	10-12	12.20	90.54	1.55	1.15×10^1
6.78-7.46	12-15	9.46	100	1.20	1.27×10^1

TABLE VI RESULTS OF SIEVING THE MICROSPHERES
IN SAMPLE 15 AFTER IMPACTION

Sieved Size Range (μ)	After Impaction	
	Mass %	Cumulative %
<10	12.72	12.72
10-20	15.60	28.32
20-44	21.41	49.73
44-63	12.52	62.25
63-105	9.95	72.20
105-125	5.79	77.99
125-149	9.03	87.02
149-177	6.63	93.65
177-210	6.35	100.00

FIGURE 1 SIZE DISTRIBUTION HISTOGRAM OF THE 28,982 PARTICLES FOUND FROM SAMPLE 15. THE ARITHMETIC MEAN DIAMETER IS $\sim 0.2 \mu$.

FIGURE 2 CUMULATIVE PLOT OF MASS PERCENTAGES OF ALL MICROSPHERES IMPACTED IN SAMPLE 15. THE MASS MEDIAN DIAMETER IS 44 μ .

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