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A FAILURE-RESISTANCE EVALUATION OF $^{238}\text{PuO}_2$
MICROSPHERES UNDER SIMULATED
ENVIRONMENTAL CONDITIONS

Prepared by
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SANDIA LABORATORIES

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UNDER SIMULATED ENVIRONMENTAL CONDITIONS

February 1968

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Sandia Corporation

under

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ABSTRACT

The resistance of $^{238}\text{PuO}_2$ microspheres to spallation and fragmentation under environmental conditions simulating isotopic-heat source catastrophic failure under reentry was evaluated. Small test samples of uncoated microspheres were subjected to a helium environment at 1300°F and 2500 psi and rapidly depressurized in time periods as short as 25 millisecc with no apparent loss of particle integrity.

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A FAILURE-RESISTANCE EVALUATION
OF $^{238}\text{PuO}_2$ MICROSPHERES UNDER
SIMULATED ENVIRONMENTAL CONDITIONS

Introduction and Summary

Some uncertainties exist regarding the use of uncoated $^{238}\text{PuO}_2$ microspheres as radioisotopic fuels in aerospace vehicles. If, as a result of rapidly changing environmental conditions, especially such as might be encountered as a result of capsule failure in orbit or during reentry, the particles fragment to produce a substantial quantity of "fines" less than $10\text{ }\mu\text{m}$ in size, these inhalable fines can represent a serious biological hazard upon reaching the earth's surface.

To evaluate the failure resistance of $^{238}\text{PuO}_2$ microspheres under simulated environmental conditions, test samples of microspheres were subjected to a temperature of 1300 F under gradually increasing pressure to a maximum pressure of 2500 psi in an autoclave under a helium atmosphere, exposed at constant temperature and pressure for several hours, and then rapidly depressurized. The conditions of the test were specified by Sandia personnel. It was calculated that these pressurization conditions would cause saturation of the microspheres with helium based upon an assumed diffusion coefficient of $D = 10^{-8}\text{ cm}^2$ per sec. Two tests were performed: one in which the depressurization to one atmosphere occurred in 4 to 5 sec, the second in which the depressurization to one atmosphere was accomplished in 25 millisec. Post-test evaluation of the microspheres by metallographic, radiographic, and physical screening techniques failed to reveal any evidence of gross particle failure.

Objective

Several SNAP devices considered for space application in the near future have as a reference fuel uncoated $^{238}\text{PuO}_2$ microspheres for the source of heat resulting from radioactive decay. For purposes of design, these particles were initially assumed to completely release the helium generated during the alpha decay process; therefore, the containing capsule was designed with sufficient internal void volume so that all of the helium generated could be accommodated without exerting undue internal stresses on the capsule.

The fuel particles are generally formed by spheroidizing irregular PuO_2 granules in a plasma torch. Although capable of producing a highly spherical particle, the plasma-torch process produces a particle which is quite often characterized by large central voids, especially when the particle diameter exceeds 100 μm . Should these voids become filled with helium as a result of alpha decay, it is possible that this internal pressure might cause the particles to fragment if the surrounding capsule pressure were suddenly decreased as a result of capsule failure. Such behavior requires that the release of gas from the particles is not rapid and that the mechanical strength of the microspheres is less than that required to withstand the sudden differential pressure. The objective of this research program is to simulate within reasonable limits such an event, and to evaluate the resistance of the microspheres to fragmentation under the test conditions.

To simulate such an event, small samples of uncoated $^{238}\text{PuO}_2$ microspheres were placed in an autoclave and were subjected to a temperature of 1300 F. The helium partial pressure was gradually increased to a maximum pressure of 2500 psi and the samples were exposed to the constant temperature and pressure for several hours, and then rapidly depressurized. These test conditions were selected on the basis of operating temperatures and a diffusion constant for helium in PuO_2 estimated by Sandia. Two tests were performed; one where the depressurization time was 4 to 5 sec, and another where depressurization was accelerated and accomplished in 25 millisec.

The 1300 F temperature is a typical operating condition for the encapsulated fuel, while 2500 psi is representative of the internal design pressure of the capsule after 20 years of operating life, if complete helium release is postulated. With the use of classical diffusion equations⁽¹⁾, it can be shown that a 125- μ m diameter microsphere would be fully saturated with helium in about 0.55 hr, assuming a diffusion coefficient of $D = 10^{-8}$ cm² per sec for the temperature of interest. Allowing for error in postulating a diffusion coefficient, it can also be shown that the mean helium concentration within the microsphere voids would be nearly 98 percent of the capsule concentration in 4 hr if the diffusion coefficient is decreased by an order of magnitude, i.e., $D = 10^{-9}$ cm² per sec. Therefore, the 4 hr exposure should allow particles with these characteristics to become fully saturated with helium provided the estimated diffusion coefficients are accurate.

At the time this experimental effort was performed, the estimated diffusion coefficient of $D = 10^{-8}$ cm² per sec suggested by Sandia was based on estimates made for Cm₂O₃⁽²⁾ and evidence that helium was released from the fuel as rapidly as it was generated.⁽³⁾ Based on more recent observations it would appear that the diffusion coefficient of helium in PuO₂ may be as low as, or even lower, than $D = 10^{-12}$ cm² per sec, although this value has not yet been experimentally confirmed. If this is indeed the case, it would require over four months exposure for the mean helium concentration within the microsphere to equal one-half the surface concentration, neglecting any internal helium accumulation from the radioactive decay process. The internal pressure increase within the microspheres from the decay process can be estimated provided several conditions are assumed. These are (1) that no helium is released from the microsphere by diffusion or other mechanisms, (2) that all helium bred into the ²³⁸PuO₂ prior to processing into microspheres was released during the plasma torch spheroidizing process, and (3) that the microspheres contain approximately 8% void volume (based on reported density values) to accumulate

(1) Darken, L. S., and Gurry, R. W., Physical Chemistry of Metals, McGraw-Hill Book Co., Inc., New York (1953), p 437 ff.

(2) Eugene Lamb, Isotope Power Fuels Monthly Status Report for July 1966, Oak Ridge National Laboratory, August 12, 1966.

(3) G. R. Grove, Division of Isotopes Development and Related Activities for February and March 1965, USAEC Report MLM-CF-65-2-452 (March 22, 1965).

the helium. Using helium release data for Pu-238 reported by Los Alamos,⁽⁴⁾ it can be calculated that the internal helium pressure from radioactive decay after 61 days (time from spheroidizing to time of initial test) would equal 37 atm (545 psi) at the testing temperature of 1300 F. The observed interconnected porosity within these microspheres casts some doubt on the validity of some of the above calculations since this porosity would accelerate the release of decay helium from the microspheres, and conversely, would accelerate the saturation rate of the microspheres from an external helium source.

After testing, the microspheres were evaluated by screening, microradiography, and metallography to determine if any degradation had occurred. Three possible reasons might be advanced should the particles not fail, i.e., (1) the microspheres are mechanically strong enough to withstand the sudden pressure differential, (2) the particles have sufficient internal interconnected porosity to allow them to depressurize as rapidly as the capsule without relying on diffusion as a pressure-release mechanism, or (3) error in the estimated diffusion constant results in less than equilibrium concentration of helium in the microsphere.

Experimental Results

Test Facility Design

The facility designed and constructed for the initial test is shown schematically in Figure 1. The autoclave consists of a section of thick-walled, high-pressure stainless steel tubing, welded at one end and coupled to a valving system on the other end. Spacers are incorporated into the tubing to decrease internal helium volume and thus minimize the time required to depressurize the system. The capsule which contains the PuO₂ consists of a section of thin-walled stainless steel tubing with a 1/16-in. thick sintered stainless steel filter having porosity not exceeding 1 μ m in diameter welded into one end. The microspheres are

(4) Matlack, George M. and Metz, Charles F., Radiation Characteristics of Plutonium-238, USAEC Report LA-3696 (October, 1967).

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loaded into the capsule and the open end is then closed by welding a solid end plug into place. A small tube furnace monitored by several thermocouples supplies heat to the specimen zone of the autoclave. Helium pressure is introduced to the system and exhausted through the valves shown in Figure 1. With this design it was found experimentally that the autoclave and specimen capsule could be depressurized in about 4 sec. For the second test in this investigation, it was decided by Sandia that a depressurization time of less than 1 sec was desirable as being more representative of the actual case. As a result, the test facility was redesigned, as shown in Figure 2. A 1500-psi bursting-strength rupture disk (10) separates the connection blocks (6 and 7). A helium pressure of 2500 psi is maintained on the sample side of the rupture disk, and a 1250 psi pressure is maintained on the exhaust side. By actuating the quick-opening valve (12), the rupture disk bursts when the pressure differential between the exhaust side and the sample side exceeds 1500 psi, thus permitting an extremely rapid depressurization of the autoclave. A tubular spacer (5) was substituted for the solid slotted spacer previously used to eliminate any restriction of gas flow which may have been caused by the original spacer. The internal volume of the autoclave system on the sample side of the rupture disk was maintained as small as practical to minimize the volume of gas to be released, but not restrict gas flow.

In order to measure the actual transient pressure change as a function of time, a mockup assembly was fabricated in which the pressure transducer was mounted at the normally closed end of the thick-wall pressure tubing and a filter of 1/16-in. thick porous stainless steel of the type used on the specimen capsule was placed in the position of the capsule, completely isolating the pressure transducer from the valving system. The test was performed at room temperature at 2400 psi (gas-cylinder pressure). The pressure on the transducer side of the filter dropped to less than 100 psi in 25 millisecc (Figure 3). The porous filter was removed and examined macroscopically. No evidence of filter damage was observed.

LEGEND

1. Thick-wall pressure tubing (welded one end)
2. Spacer
3. Tube furnace
4. Specimen capsule
5. Slotted spacer
6. Pressure gage
7. Valves

FIGURE 1. SCHEMATIC DIAGRAM OF TEST FACILITY USED
FOR FIRST TEST CYCLE (Not to Scale)

1. Thick-wall pressure tubing (welded one end)
2. Solid spacer
3. Tube furnace
4. Specimen capsule
5. Tubular spacer
6. Connection block #1
7. Connection block #2
8. Pressure transducer
9. Pressure gage
10. Rupture disc assembly (1500 psi)
11. Helium inlet valves
12. Quick-opening valve

FIGURE 2. SCHEMATIC DIAGRAM OF TEST FACILITY USED
FOR SECOND TEST CYCLE (Not to Scale)

a. Depressurization of Test Apparatus at $R_{OC_{111}}$ Temperature with Sensing Transducer Located at Normally Sealed End of Autoclave (Chart Speed = 200 mm/sec)

b. Depressurization of Specimen Capsule Containing $^{238}\text{PuO}_2$ Microspheres from 2500 psi at 1300 F.

FIGURE 3. HIGH-SPEED RECORDER TRACES OF CAPSULE RESPONSE TO DEPRESSURIZATION

Characterization of Pretested Microspheres

Approximately 0.87 g of $^{238}\text{PuO}_2$ microspheres, identified as Mound Batch 101, was received from Mound Laboratory. Photomicrographs, photomacrographs, density, crushing strength, and impurity analyses were supplied by Mound and are given in Figures 4 through 7 and Table 1. Macroexamination of the as-received microspheres revealed a variety of surface conditions ranging from particles exhibiting a smooth, glass-like surface to those which were roughened and deeply pitted. This pitting was noted particularly in the particles of larger diameter. Figure 4 through 6 show microspheres exhibiting various internal structures. As noted previously, the frequency of internal voids increases as the particle diameter increases, and is generally attributed to a solidification phenomenon. The etched microstructure (Figure 5) reveals an apparent widespread network of continuous paths, or voids, throughout the particles from which internal helium might readily be released, even under conditions of rapid capsule depressurization. The as-received microspheres were screened and no particles with a diameter greater than 250 μm (60 mesh) or less than 44 μm (325 mesh) were observed. Some difficulty was noted in removing the particles from the inner aluminum shipping vial because of an apparent static electrical charge which caused the microspheres to adhere tightly to the inner vial surface.

Test Procedure (Test 1)

A weighed quantity (0.8422 g) of microspheres was loaded and sealed in the stainless steel capsule and inserted into the autoclave (Figure 1). The temperature and pressure were gradually increased to 1300 F and 2500 psi, respectively (Table 2). After a constant hold at the test conditions for 4 hr, the helium pressure was decreased to 1 atm within a time span of 4 to 5 sec. The specimen was then cooled to room temperature in approximately 3 hr. Visual examination of the capsule revealed no evidence of capsule or filter failure.

100X

PL3538

FIGURE 4. AS-POLISHED CROSS SECTION OF $^{238}\text{PuO}_2$ MICROSPHERES FROM THE 100 TO 150- μm -DIAMETER FRACTION OF MOUND BATCH 101

500X

PL3541

FIGURE 5. ETCHED CROSS SECTION OF $^{238}\text{PuO}_2$ MICROSPHERE FROM THE 100 TO 150- μm DIAMETER FRACTION OF MOUND BATCH 101
Note the continuous network of paths, or voids, which could enhance helium release.

100X

PL3539

FIGURE 6. $^{238}\text{PuO}_2$ MICROSPHERES FROM THE FRACTION OF MOUND BATCH 101 HAVING DIAMETERS GREATER THAN $150\ \mu\text{m}$

Note the increasing frequency of voids in these larger-size particles, as compared to Figure 5.

50X

PL3540

FIGURE 7. REPRESENTATIVE EXTERNAL APPEARANCE OF SAMPLE OF $^{238}\text{PuO}_2$ MICROSPHERES FROM MOUND BATCH 101

Note the variety of surface conditions, ranging from a smooth glass-like surface to a roughened surface.

TABLE 1. CHARACTERIZATION OF AS-RECEIVED $^{238}\text{PuO}_2$
MICROSPHERES FROM MOUND BATCH 101

Density (a)	
G per cm ³	10.52
Percent of Theoretical	91.8
Crushing strength, g	
Average (b)	438
Maximum	1147
Minimum	77
Impurities, Percent as metal	
Nickel	0.06
Iron	0.35
Aluminum	0.09
Calcium	0.04
Chromium	0.10
Zinc	0.08
Copper	0.06

(a) Determined with mercury pycnometer.

(b) Average of 20 determinations.

TABLE 2. TEST CYCLE CONDITIONS (TEST 1)

Time, hr		Temperature of Indicated Thermocouple (a), F		Pressure, psi
Total	Cycle	T/C 3	T/C 4	
Start	--	--	--	300
1/3	--	437	455	600
1/2	--	642	650	600
1	--	908	912	1600
1-1/2	--	1121	1123	2400
2-1/2	Start	1280	1280	2500
3	1/2	1289	1289	2500
3-1/2	1	1291	1291	2500
4	1-1/2	1289	1289	2500
4-1/2	2	1293	1293	2500
5	2-1/2	1291	1291	2500
5-1/2	3	1293	1293	2500
6	3-1/2	1292	1292	2500
6-1/2	4	1291	1291	2500

(a) Thermocouples T/C 3 and T/C 4 were located on the outer surface of the autoclave opposite the specimen capsule.

3

The specimen capsule was opened using a tubing cutter, and the particles, all of which poured out freely, were screened on a 325-mesh (44 μm) screen. No particles less than 44 μm in diameter were observed. A screening system utilizing a series of electroformed "micromesh" sieves with nominal openings of 20, 10, and 5 μm had been constructed and evaluated for screening any particles less than 44 μm that might be produced. Screening is accomplished in this system by wet sieving in a water medium and applying a slight vacuum to the collection flask to minimize screen clogging. However, for this evaluation its use was not required. A small sample of particles was submitted for metallographic, macroscopic, and X-ray microradiograph evaluations. As shown in Figures 8 through 10, none of these evaluations revealed any evidence of particle fragmentation or failure. X-ray microradiography permits the evaluation of several hundred particles simultaneously. Figure 10 represents a portion of the particles evaluated in this manner. Neither irregular particles nor fragments were noted in the several microradiographs examined.

Test Procedure (Test 2)

A weighed quantity (0.7240 g) of microspheres recovered from the initial test was loaded and sealed in a new stainless steel capsule and inserted into the redesigned autoclave (Figure 2). The temperature and pressure were gradually increased to 1300 F and 2500 psi, respectively (Table 3). Temperature and pressure parameters were unchanged from the initial test, but the holding time at temperature and pressure was increased from 4 to 8 hr, as suggested by Sandia to further enhance the diffusion of helium into the microsphere void cavities. After the 8-hr hold, the helium pressure was decreased to 1 atm in approximately 25 millisecc. The trace of the high-speed-recorder input from the pressure-sensing transducer is shown in Figure 3b. After removal of the capsule from the test apparatus, a visual examination of the capsule revealed no evidence of capsule or filter failure. The capsule was

125X

PL3501

FIGURE 8. MICROSTRUCTURE OF MOUND BATCH 101 $^{238}\text{PuO}_2$ FOLLOWING A 4-HR EXPOSURE AT 1300 F AND 2500 PSI AND THEN RAPID DEPRESSURIZATION IN 4 TO 5 SEC (TEST 1)

50X

PL3496

FIGURE 9. PHOTOMICROGRAPH OF MOUND BATCH 101 $^{238}\text{PuO}_2$ AFTER A 4-HR EXPOSURE AT 1300 F AND 2500 PSI AND THEN RAPID DEPRESSURIZATION IN 4 TO 5 SEC (TEST 1)

150X

L20272

FIGURE 10. X-RAY MICRORADIOGRAPH OF MOUND BATCH 101 $^{238}\text{PuO}_2$ AFTER TEST EXPOSURE AT 1300 F AND 2500 PSI FOR 4-HR AND THEN RAPID DEPRESSURIZATION IN 4 TO 5 SEC (TEST 1)

TABLE 3. TEST CYCLE CONDITIONS (TEST 2)

Time, hr		Temperature of Indicated Thermocouple ^(a) , F		Pressure, psi
Total	Cycle	T/C 3	T/C 4	
Start	--	R.T.	R.T.	15
2/3	--	897	901	15
1	--	1171	1171	15
1-1/3	--	1278	1276	300
2-1/3	--	1295	1295	1350
3	Start	1299	1297	2450
3-1/2	1/2	1297	1297	2450
4	1	1297	1297	2500
4-1/2	1-1/2	1301	1301	2500
5	2	1302	1302	2500
5-1/2	2-1/2	1301	1301	2500
6	3	1303	1305	2500
6-1/2	3-1/2	1297	1297	2500
7	4	1304	1304	2500
7-1/2	4-1/2	1297	1297	2500
8	5	1297	1297	2500
8-1/2	5-1/2	1299	1299	2500
9	6	1299	1299	2500
9-1/2	6-1/2	1302	1302	2500
10	7	1299	1299	2500
10-1/2	7-1/2	1300	1300	2500
11	8	1300	1300	2500

(a) Thermocouples T/C 3 and T/C 4 were located on the outer surface of the autoclave opposite the specimen capsule.

opened and particles recovered in the manner previously described for the initial test. Macroscopic, metallographic, and screening evaluations of the collected microspheres failed to reveal any particle fragments less than 325 mesh (44 μ m). Photographic confirmation is shown in Figures 11 and 12. One X-ray microradiograph (from a series of five exposures) did reveal one or more fragmented particles (Figure 13), but this would not necessarily be a result of the test conditions since it represents an isolated occurrence.

500X

PL4382

FIGURE 11. MICROSTRUCTURE OF MOUND BATCH 101 $^{238}\text{PuO}_2$ FOLLOWING AN 8-HR EXPOSURE AT 1300 F AND 2500 PSI AND THEN RAPID DEPRESSURIZATION IN 0.025 SEC (TEST 2)

50X

PL4174

FIGURE 12. PHOTOMICROGRAPH OF MOUND BATCH 101 $^{238}\text{PuO}_2$ AFTER AN 8-HR EXPOSURE AT 1300 F AND 2500 PSI AND THEN RAPID DEPRESSURIZATION IN 0.025 SEC (TEST 2)

150X

L20468

FIGURE 13. X-RAY MICRORADIOGRAPH OF MOUND BATCH 101 $^{238}\text{PuO}_2$ AFTER AN 8-HR EXPOSURE AT 1300 F AND 2500 PSI AND THEN RAPID DEPRESSURIZATION IN 0.025 SEC (TEST 2)

CONCLUSIONS

On the basis of these two experimental tests at the specified test parameters, it would appear that this lot of $^{238}\text{PuO}_2$ microspheres would resist fragmentation if depressurized from 2500 psi to 1 atm at 1300 F in time periods as short as 0.025 sec. This conclusion is based on the assumption that the experiments performed are a good simulation of actual service conditions which could be encountered if a catastrophic failure of the microsphere containment capsule were to occur. It should be pointed out that when the helium is bred into the PuO_2 structure by a natural decay process, the probability of the internal pores containing a gas pressure equivalent to the external pressure is greater than it is in the experiments performed. The accuracy of the estimated diffusion coefficient, surface activation processes which affect the rate at which helium enters and leaves a surface, and the sensitivity of helium solubility in PuO_2 to external helium pressure will each have a bearing on whether an equilibrium gas pressure was established between isolated pores and the external pressure. As previously observed, these microspheres exhibited a continuous network of paths in the as-fabricated condition when metallographically etched (Figure 5). Even under the conditions of extremely rapid depressurization experienced in the second experimental test, it is conceivable that the internal helium may have been released through this network. Or perhaps, as postulated previously, the microspheres have sufficient mechanical strength to withstand the sudden pressure differential until the internal helium can escape through the network of interconnected pores, or by diffusion from the particle. It should be noted that this particular lot of microspheres performed satisfactorily during these experiments even though the relatively low reported density and the wide variations observed for crushing strength of individual particles might indicate that a significant portion of the microspheres had considerably less than the optimum density that might be desired for this radioisotopic fuel form. It would seem desirable to expand this series of experiments to include the evaluation of other plasma-torch

processing batches, and also to evaluate the resistance to fragmentation of microspheres formed by the Sol-Gel process. Additional experiments might also be conducted at higher temperatures and pressures to determine the threshold where particle failure might be initiated.

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