**SAVANNAH RIVER LABORATORY
MONTHLY REPORT**

^{238}Pu Fuel Form Processes

DECEMBER 1973

MASTER

**E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, S. C. 29801**

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(07-2)-1

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

SAVANNAH RIVER LABORATORY MONTHLY REPORT

²³⁸Pu Fuel Form Processes

DECEMBER 1973

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

This document is
PUBLICLY RELEASABLE
JoAnn Wingard (SRS)
Authorizing Official
Date 11/21/03

by Dorald Williann 11/24/03

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, S. C. 29801

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(07-2)-1

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

SUMMARY

Tap and pour densities ranged from 1.5 to 5.1 g/cc for calcined, reverse-strike feed and sintered, PMC shards, respectively. Densities of shards increased with increasing particle size and increasing sintering temperature; densities for milled powder are higher than for calcined, reverse-strike powder, indicating a strong effect of particle shape that is removed by milling. (p 6)

Vibratory milling calcined, reverse-strike oxide for 8 hr produced the same size distribution as a ball milling at LASL for 32 hr. (p 6)

Milling is postulated to reduce low-density aggregates to high-density platelets without changing specific surface area significantly. (p 14)

Post-hot-pressing heat treatment has a small effect on pellet dimensions and weight. The slight weight increase is probably due to oxidation of the suboxide. (p 16)

Air sampling stations between filter stages in exhausts from the AMF show ^{238}Pu concentrations that are only 10^{-2} to 10^{-3} times the expected values. (p 24)

DISCLAIMER

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

DISCLAIMER

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

CONTENTS

	<u>Page</u>
List of Figures and Tables	4
Introduction	5
Process Development and Demonstration	6
Pour and Tap Densities of ^{238}Pu Powder and Shards . .	6
Milling of Calcined, Reverse-Strike Oxide	6
Morphology of As-Precipitated and Milled Powder . . .	14
PPO Pellet Fabrication	15
Hot Pressing	16
Post-Hot-Pressing Heat Treatment	16
Documentation	23
Milliwatt Thermoelectric Generator	23
Equipment and Facilities	24
Alpha Materials Facility	24
Scanning Electron Microscope	24
Scrap Recovery	24
References	25
Meetings and Visits	26

LIST OF FIGURES AND TABLES

Figure	<u>Page</u>
1. Pour and Tap Density of $^{238}\text{PuO}_2$ Powders	8
2. Pour and Tap Density of $^{238}\text{PuO}_2$ Shards	9
3. Size Distribution of LASL Ball-Milled $^{238}\text{PuO}_2$	10
4. Size Distribution of SRL <300- μ Shards	11
5. Size Distribution of SRL <355- μ Shards	12
6. SRL Small-Scale Mill Chamber	14
7. Center of MHP-2 Pellet As-Pressed	22

Table	<u>Page</u>
I Pour and Tap Densities of $^{238}\text{PuO}_2$ Powder and Shards.	7
II Particle Size Statistics of Milled $^{238}\text{PuO}_2$	12
III Summary of PPO Pellets Hot-Pressed at SRL	17
IV Hot Pressing Experiment MHP-3	18
V Hot Pressing Experiment MHP-4	19
VI Hot Pressing Experiment MHP-5	20
VII Dimensions and Density of Hot-Pressed Pellets Before and After Heat Treatment at 1540°C for 12 hr in Air.	21
VIII Post-Hot-Pressing Heat-Treatment Temperature and Time in Air	21

INTRODUCTION

This report is one of a series to summarize progress in the Savannah River ^{238}Pu Fuel Form Program; this program is supported by the AEC Divisions of Space Nuclear Systems (SNS) and Military Applications (DMA).

Goals of the Savannah River Laboratory (SRL) program for SNS are: to provide technical support for the transfer for SNS ^{238}Pu fuel form fabrication operations from Mound Laboratory to a new facility to be built at the Savannah River Plant (SRP), to provide the technical basis for ^{238}Pu scrap recovery at SRP, and to assist in sustaining plant operations. This part of the program includes:

Demonstration of processes and techniques, currently in use at Los Alamos Scientific Laboratory (LASL) and Mound, for production at SRP, thereby providing design information and the data upon which technical standards and operating procedures will be based.

Technical Support to assist plant startup, and to assure continuation of safe and efficient production of high quality heat source fuel.

Technical Assistance after startup to accommodate changes in product and product specifications, to assist user agencies in improving product performance, to assist the plant in making process improvements that increase efficiency and product reliability, and adapting plant facilities for new products.

The goal of the SRL program for DMA is to provide characterization for the $^{238}\text{PuO}_2$ feed manufactured at Savannah River and sent to Mound Laboratory to be fabricated there into fuel forms for the Milliwatt Radioisotope Thermoelectric Generator.

PROCESS DEVELOPMENT AND DEMONSTRATION

POUR AND TAP DENSITIES ^{238}Pu POWDER AND SHARDS

Measurements of pour and tap density have been completed for SRP reverse-strike powder, LASL ball-milled powder, SRL ball-milled powder, SRL vibratory-milled powder, LASL shards, and SRL shards; data are summarized in Table I and Figures 1-5. These data are an extension of results initially described in the October report, p. 8. Pour and tap densities were measured to provide bases for measurements of thermal conductivity and for design of process containers in which the maximum temperature experienced by the various forms of $^{238}\text{PuO}_2$ during fuel processing is controlled. Values of tap density also provide an indication of particle morphologies and particle densities.

Tap and pour densities ranged from 1.5 to 5.1 g/cc for calcined, reverse-strike feed and sintered, plutonia-molybdenum cermet (PMC) shards, respectively. This large variation is expected to affect design of process equipment significantly, because of its effect on temperature distributions for the various kinds of powder. As expected, tap and pour densities of shards increase with both increasing particle size and increasing sintering temperature (higher particle density), Figure 2. However, contrary to expectations, pour and tap densities of milled powder are higher than for calcined, reverse-strike powder, although the median particle size for milled powder is smaller. This observation, along with the relative difference between pour and tap density for reverse-strike powder and the large number of taps required to achieve final tap density, indicates a strong effect of particle shape that is removed by milling. The rosette structure of $^{238}\text{PuO}_2$ particles and the impact on milling rates is discussed below.

MILLING OF CALCINED, REVERSE-STRIKE OXIDE

The characteristic particle size distribution of LASL ball-milled $^{238}\text{PuO}_2$ has been measured. Vibratory milling has been shown to produce the same size distribution in 8 hr compared to the 32-hr LASL ball-milling time. Vibratory and ball milling are being evaluated with $^{238}\text{PuO}_2$ on a 10-g scale with objectives of characterizing milled product, shortening milling times to obtain a uniform and reproducible powder, and reducing impurities arising from wear of the mill. Vibratory milling is expected to be faster and produce less wear than ball milling because the balls spend more of their time impacting than being dragged up the side of the rotating mill. Mill performance described below and in Table 2 is an extension of the data initially presented in the October report.

TABLE I
POUR AND TAP DENSITIES OF $^{239}\text{PuO}_2$ POWDER AND SHARDS

Material and Treatment	Median Particle Size, μ	Pour Density g/cc	Final Tap Density, g/cc	Number of Taps to Final Density
<u>Reverse-Strike Powder</u> SRP reverse-strike No. 209HA220, As-calcined	5.5 (October report, Fig.3)	1.45	2.24	7,500
<u>SRL Ball-Milled Powder (SRL-29A)</u> SRP reverse-strike No. 209HA220, Ball-milled 32 hr	1.78 (October report, Fig.4)	2.80	3.39	4,000
<u>LASL Ball-Milled Powder (LASL-BND 203)</u> SRP reverse-strike No. 204904, Ball-milled 32 hr by LASL	2.3 (Figure 3)	2.53	3.51	4,000
<u>Vibratory-Milled Powder (SRL-5-B)</u> SRP reverse-strike No. 209HA220, Vibratory-milled 32 hr	~1.5 (October report, Table II & Figs. 5-7)	2.91	3.34	7,500
<u><125-μ Heat-Treated Shards (LASL-BND 199)</u> SRP reverse-strike No. 20490, Ball-milled 32 hr, cold-pressed, passed through 125- μ screen, heat- treated at 1075°C for 3 hr	<125	2.83	4.11	5,000
<u><300-μ Heat-Treated Shards (SRL-19-B)</u> SRP reverse-strike No. 209HA220, Ball-milled 32 hr, cold-pressed, passed through 300- μ screen, heat- treated at 1075°C for 3 hr	115.5 (Fig. 4)	3.77	4.32	10,000
<u><355-μ Heat-Treated Shards (SRL-11-B)</u> SRP reverse strike No. 209HA220, Vibratory-milled 32 hr, cold-pressed, passed through 355- μ screen, heat- treated at 1075°C for 3 hr	157.6 (Fig. 5)	3.96	5.27	10,000
<u><300-μ Sintered Shards (SRL-19-B/2)</u> SRP reverse-strike No. 209HA220, Ball-milled 32 hr, cold-pressed, passed through 300- μ screen, heat- treated at 1075°C for 3 hr, sintered at 1625°C for 6 hr	<115	5.11	5.46	5,000

FIGURE 1. POUR AND TAP DENSITY OF $^{238}\text{PuO}_2$ POWDERS

FIGURE 2. POUR AND TAP DENSITY OF $^{238}\text{PuO}_2$ SHARDS

FIGURE 3. SIZE DISTRIBUTION OF LASL BALL-MILLED $^{238}\text{PuO}_2$

FIGURE 4. SIZE DISTRIBUTION OF SRL <300- μ SHARDS

FIGURE 5. SIZE DISTRIBUTION OF SRL <355-μ SHARDS

TABLE II
PARTICLE SIZE STATISTICS OF MILLED $^{238}\text{PuO}_2$

Material	Milling Time, hr	Particle Size, μ		
		Median	Mean	Std. Dev.
Reverse-strike as-calcined (HB-Line)	--	5.5	8.4	8.2
Ball-milled (LASL)	32	2.3	3.6	6.0
Ball-milled (SRL)*	32	1.8	2.3	2.8
Vibratory-milled (SRL)*	8	1.4	2.2	4.8
Vibratory-milled (SRL)*	16	1.3	2.0	3.0
Vibratory-milled (SRL)*	24	1.5	2.2	3.7

*Graphical data in Figures 4-7 of October report.

The particle size distribution of the LASL ball-milled material was used as a basis for this study. LASL currently mills 75-g batches of SRP calcined, reverse-strike oxide that has been oxygen exchanged at 775°C and outgassed and normalized in thermal history at 1000°C. Their mill chambers and balls are made of hardened Type 440 stainless steel. The mill chamber is 3 in. OD (Rockwell C-58-60), and is partially filled with 0.5-in.-dia balls (Rockwell C 62-64). Milling time is 32 hr at a rotation of 50 rpm. Iron, chromium, and nickel are introduced at the rate of 2 ppm/hr.

The SRL mill chamber is shown in Figure 6 and was used both for ball milling and vibratory milling. The mill chamber was scaled to a nominal 10-g batch size. The Type 304L stainless steel mill chamber has a 1.750-in.-dia x 0.500-in. cavity, and contains 1/4-in.-dia hardened alloy steel balls. The same chamber is rotated at 50 rpm for ball milling or vibrated at 3360 cycles/min with ≈ 0.1 -in. amplitude for vibratory milling. When used for ball milling, efficiency was optimized by filling the chamber 50% with balls.¹ When used for vibratory milling, efficiency was optimized by filling the chamber 80% with balls.² For all experiments sufficient $^{238}\text{PuO}_2$ powder was charged to fill the interball voids.

Particle size distribution of standard LASL material, which had been ball-milled for 32 hr, is shown graphically in Figure 3 and in Table II. The 32-hr ball milling reduces the median particle size from the 3 to 9 μ found in typical HB-Line material to 2.3 μ . Ninety percent is less than 5 μ and about 60% is in the nominal range of 2 ± 1 μ generally required by LASL. As shown in Table II and in Figure 4 of the October report, the small ball mill described above reproduces the particle size distribution of LASL material fairly well for the same 32-hr ball milling time. In fact, the parameters describing the particle size distribution indicate that on this smaller scale, the milling time to achieve the desired 2 ± 1 μ size range could be considerable shorter than 32 hr.

Preliminary results from vibratory mill tests show that a similar particle size distribution can be obtained in less than 8 hr, Table II. As indicated by particle size distributions for vibratory milling for 16 and 24 hr, additional milling time has no effect on either median size or residual amount of larger particles.

FIGURE 6. SRL SMALL-SCALE MILL CHAMBER

MORPHOLOGY OF AS-PRECIPITATED AND MILLED POWDER

A basic shape or morphology factor apparently characterizes particles of calcined ^{238}Pu oxalate and may affect subsequent processing into fuel forms. This factor is suggested by the behavior of the powder in measurements of pour and tap densities (above), milling behavior (above), microscopy performed by Battelle on $^{239}\text{PuO}_2^{3,4}$, particle instability on ultrasonic dispersion for particle size analysis (January report, p. 9 and February report, p. 5), and unexpectedly low densities obtained by LASL on hot-pressing direct-strike oxide. Control of precipitation and calcination conditions, which determine shape and specific surface area, is probably more critical to control of $^{238}\text{PuO}_2$ powder sinterability than milling conditions. Milling is believed to produce negligible increases in specific surface area, but it degrades loosely bonded aggregates into their component particles for better packing during pressing. Planned scanning microscope and Brunauer-Emmett-Teller (BET) specific surface area measurements will establish a basis for understanding and control of morphology.

As-calcined $^{239}\text{PuO}_2$ has been shown microscopically to consist of nearly spherical low-density aggregates (rosettes), each of which is composed of numerous high-density platelets.^{3,4} Photomicrographs of as-calcined $^{238}\text{PuO}_2$ also indicate rosette aggregates (February report, p. 5). Decreases in median $^{238}\text{PuO}_2$ particle size with increasing sonic dispersion time (January report, p. 7) during particle size analysis indicate that calcined $^{238}\text{PuO}_2$ oxalate powder consists of loosely bonded aggregates. The large increase in tap

density after milling indicates degradation of these rosette aggregates. If the rosettes were fully dense, they would be expected to pack quite well to a pour density of about 30% of theoretical and a tap density of about 50% of theoretical.⁵ However, measured values for calcined, reverse-strike $^{238}\text{PuO}_2$ were 12.6% and 19.5% of theoretical density, respectively. Consequently, the rosette particle densities are $\approx 40\%$ of theoretical, indicating that as-calcined $^{238}\text{PuO}_2$ particles are skeletal and subject to degradation by milling. Upon milling the rosettes are postulated to be converted into individual, high-density platelets, which collectively comprised the rosette aggregate. The platelets appear not to be degraded significantly during further milling, as evidenced by negligible reduction of particle size after 8, 16, and 32 hr of vibratory milling and the fact that ball and vibratory milling produce the same ultimate size particles even with different sized balls.

During extensive milling, the ball-mill charge changes texture from a free flowing powder to agglomerates about 1/8 in. in diameter that are believed to consist of aligned platelets in contrast to the random edge-to-face and lower-density array of platelets in unmilled rosettes. The higher density of these platelets is indicated by the higher pour and tap densities for milled material reported above. Existence of loosely bonded agglomerates is also supported by the relative ease with which agglomerates are degraded with increasing times of exposure to ultrasound during particle size analyses.

These observations suggest that once rosettes are destroyed, further milling may not have a large effect on subsequent sintering because of the relatively small increase in specific surface area inferred by this hypothesis. However, milling influences the packing efficiency of powder and, hence, is expected to influence the kinetics of sintering.

PPO PELLET FABRICATION

Results of initial pellet fabrication experiments in the Alpha Materials Facility (AMF) duplicate, at least on a small scale (5 g $^{238}\text{PuO}_2$), the pure plutonium oxide (PPO) process technology developed by LASL. Small PPO pellets have been hot-pressed from sized and heat-treated feed material to densities demonstrated by LASL for the same feed and parameters. The small density increase in these pellets ($<3\%$), after post-hot-pressing heat treatments at temperatures up to 1540°C , 100°C above the expected fuel operating temperature, is typical of that observed at LASL under similar conditions. Full-scale demonstrations of PPO fabrication will not be possible until the Plutonium Experimental Facility (PEF) is operational or until hot pressing begins in the ^{238}Pu Fuel Form (PuFF) Facility. Details of initial hot-pressing and post-pressing heat-treatment studies are described below:

Hot Pressing

The results of the hot-pressing experiments with the miniature hot press in the AMF are summarized in Table III along with pertinent process variables. Higher density pellets were achieved with higher pressures, as can be observed by comparing experiments MHP-2 to MHP-1 and experiment MHP-4 to MHP-5. Higher density pellets are also obtained as the size of the feed material is decreased, as shown by MHP-3, MHP-4, and MHP-2. MHP-3, which was processed from <125- μ feed material, had the highest density (89.6%); MHP-2, hot-pressed from <355- μ feed material, had the lowest density (80.2%); and MHP-4, fabricated from an intermediate-size feed material (<297- μ), had a density between that of MHP-2 and MHP-3.

Pressing conditions and data obtained on MHP-3, MHP-4, and MHP-5 are reported in Tables IV, V, and VI, respectively. Experiments MHP-1 and MHP-2 were reported previously in the October and November reports, respectively.

Post-Hot-Pressing Heat Treatment

Similar densities, see Table VII, were achieved at LASL and at SRL for pellets processed in the same manner. In particular, pellet MHP-5, which is within PPO density specification, was fabricated following the recommended LASL flowsheet for PPO. Feed material for hot pressing was prepared by milling as-received HB-Line, reverse-strike $^{238}\text{PuO}_2$ powder, cold-pressing, and sizing according to LASL procedure. Maximum hot-pressing parameters of temperature, pressure, and time were also identical. Maximum heat-treatment temperature was 1540°C, which is approximately 100°C higher than either the temperature used at LASL or the expected fuel operating temperature; yet final density was still within PPO specifications, thus indicating a dimensional stability consistent with that observed by LASL.

The post-hot-pressing heat treatment is performed to evaluate dimensional stability, remove carbon, and adjust stoichiometry. The times at various temperatures used for this initial study are shown in Table VIII. Conditions are similar to those recommended by LASL except that the first three hold temperatures are each 100°C higher, and heating was done in air and not a $\text{H}_2^{16}\text{O}/\text{Ar}$ atmosphere. No attempt was made to control heating and cooling rates because small pellets are not as susceptible to thermal shock as large PPO spheres.

Little change in dimensions and weight of pellets was found after heat treatment, Table VII. Two pellets, MHP-4 and MHP-5, exhibited slight shrinkage apparently caused by residual sintering occurring during the heat treatment and/or contraction of the

lattice, which occurs during reoxidation of the suboxide to PuO_2 . Pellet MHP-3 had a slight increase in dimensions, but it also had a small radial crack on one end. Cracking could cause expansion of the pellet. Metallography is now being done to establish whether cracking was caused by: a chemical impurity, mechanical damage from thermal shock stress, stresses arising from lattice contractions during oxidation of the suboxide, or helium damage.

TABLE III
SUMMARY OF PPO PELLETS HOT-PRESSED AT SRL

Pellet	As-Pressed Density ^a , %	Feed Material ^b	Max.Shard Size, μ	Pressure, psi ^c	Max Temp, °C
MHP-1	62.0	SRL-S-1-11B	<355	<2400	1500
MHP-2	80.2	SRL-S-1-11B	<355	5000	1500
MHP-3	89.6	LASL-BND-199	<125	5000	1500
MHP-4	88.3	SRL-S-3-19B	<297	5000	1500
MHP-5	81.9	SRL-S-3-19B	<297	3000	1500

a. PPO requirement is $82.0 \pm 2.0\%$

b. All feed material heat-treated at 1075°C for 3 hr after sizing.

c. Maximum pressure held for 30 min.

TABLE IV

HOT PRESSING EXPERIMENT MHP-3

A. Feed Material - LASL-BND-199 <125- μ granules, heat-treated at 1075°C for 3 hr

B. Hot Pressing Parameters

(1) Heating Rate

1130°C in 7 min
1406°C in 22 min
1498°C in 36 min (accumulated)

(2) Maximum Temperature

1502°C

(3) Maximum Pressure

5000 psi

(4) Time at Maximum Pressure

30 min with pressure initially applied at 1406°C

(5) Cooling Rate

$\sim 10^\circ\text{C}/\text{min}$

(6) Vacuum

33-62 μ

C. Pellet Data

Powder Load	5.2055 g
Pellet Weight	5.0910 g
Diameter	0.380 in.
Thickness	0.267 in.
Density	10.26 g/cc
% of theoretical	89.6

TABLE V

HOT PRESSING EXPERIMENT MHP-4

A. Feed Material - SRL-S-3-19B granules <297- μ , heattreated at 1075°C for 3 hr

B. Hot Pressing Parameters

(1) Heating Rate

1173°C in 9 min
1402°C in 18 min
1496°C in 30 min (accumulated)

(2) Maximum Temperature

1502°C

(3) Maximum Pressure

5000 psi

(4) Time at Maximum Pressure

31 min with pressure initially applied at 1402°C

(5) Cooling Rate

$\sim 10^\circ\text{C}/\text{min}$

(6) Vacuum

22-50 μ

C. Pellet Data

Powder Load	5.0011 g
Pellet Weight	4.9525 g
Diameter	0.377 in.
Thickness	0.268 in.
Density	10.11 g/cc
% of theoretical	88.3

TABLE VI

HOT PRESSING EXPERIMENT MHP-5

A. Feed Material - SRL-S-3-19B granules <297- μ , heattreated at 1075°C for 3 hr

B. Hot Pressing Parameters

(1) Heating Rate

1149°C in 9 min
1402°C in 23 min
1497°C in 35 min (accumulated)

(2) Maximum Temperature

1506°C

(3) Maximum Pressure

3000 psi

(4) Time at Maximum Pressure

30 min with pressure initially applied at 1402°C

(5) Cooling Rate

$\sim 10^\circ\text{C}/\text{min}$

(6) Vacuum

24-48 μ

C. Pellet Data

Powder Load	5.0065 g
Pellet Weight	4.9711 g
Diameter	0.376 in.
Thickness	0.293 in.
Density	9.38 g/cc
% of theoretical	81.9

TABLE VII

DIMENSIONS AND DENSITY OF HOT-PRESSED PELLETS
BEFORE AND AFTER HEAT TREATMENT AS IN TABLE VIII

Pellet No.	Condition	Diameter, in.	Thickness, in.	Weight, g	Density, g/cc	Theo Density, %
MHP-3	As-Pressed	0.380	0.267	5.0910	10.26	89.6
	Heat-Treated ^a	0.382	0.268	5.1312	10.26	89.6
	% Change	+0.7	+0.4	+0.8	0	0
MHP-4	As-Pressed	0.377	0.268	4.9525	10.11	88.3
	Heat-Treated	0.374	0.266	4.9953	10.41	90.9
	% Change	-0.8	-0.7	+0.9	+3.0	+2.6
MHP-5	As-Pressed	0.376	0.293	4.9711	9.38	81.9
	Heat-Treated	0.374	0.290	4.9986	9.61	83.9
	% Change	-0.5	-1.0	+0.6	+2.5	+2.0

^a. Pellet had small radial crack.

TABLE VIII

POST-HOT-PRESSING HEAT-TREATMENT
TEMPERATURE AND TIME IN AIR

<u>Temperature, °C</u>	<u>Time, hr</u>
1100	6
1300	6
1540	12
1000	6
	<u>30 total</u>

Oxidation of the suboxide is indicated by the weight gain observed in all three pellets, Table VII. Removal of carbon alone will result in a weight loss. Analysis for carbon on a portion of the as-pressed MHP-2 pellet showed a carbon content of 1000 ppm ± 100 , higher than the ~ 200 ppm reported by LASL for as-pressed pellets. Metallography of pellet MHP-2 as-pressed showed that considerable substoichiometric oxide was formed, Figure 7. Because pellets MHP-3, MHP-4, and MHP-5 were hot-pressed under similar conditions, it is expected that the amounts of carbon and suboxide phase would be similar in these pellets. A portion of pellet MHP-2 was also heat-treated with the other three pellets. Carbon analysis and metallography will be done to determine the effectiveness of the heat treatment for carbon removal and oxidation of the suboxide.

The stoichiometry of pellets MHP-3, MHP-4, and MHP-5 as-pressed was calculated to be $\text{PuO}_{1.84}$, $\text{PuO}_{1.83}$, and $\text{PuO}_{1.89}$, respectively, from the weight gain of each pellet on heat treatment in air. These calculations were done assuming that the plutonia was fully oxidized to $\text{PuO}_{2.00}$ after heat treatment, and that the plutonium isotopic content was 80% ^{238}Pu and 20% ^{239}Pu . Credit was taken for a carbon content of 0.1 wt % in the as-pressed pellets. All other impurities were ignored. If carbon is also ignored, the calculated stoichiometry is $\text{PuO}_{1.86}$, $\text{PuO}_{1.84}$, and $\text{PuO}_{1.91}$ for MHP-3, MHP-4, and MHP-5, respectively. These calculated values of stoichiometry support observed metallographic results on MHP-2, Figure 7.

FIGURE 7. CENTER OF MHP-2 PELLETT AS-PRESSED

DOCUMENTATION

The following documents were issued this month:

- DPST-73-127-19 Process Data for PuFF Facility: Comments on Molybdenum Coating of $^{238}\text{PuO}_2$ Shards
- DPST-73-127-20 Process Data for PuFF Facility: Comments on Revised SRP Scope of Work
- DPST-73-127-21 Process Data for PuFF Facility: Trip Report - Heat Transfer in ^{238}Pu Storage
- DPST-73-127-22 Process Data for PuFF Facility: Comments on Metallography Facility and Operations
- DPST-73-497 Unit Operations Equipment for ^{238}Pu Hot Press Facility, Building 235-F Preliminary Scope
- DPST-73-532 Experimental Facilities for ^{238}Pu Fuel Form Process Development

MILLIWATT THERMOELECTRIC GENERATOR

No work this month.

EQUIPMENT AND FACILITIES

ALPHA MATERIALS FACILITY (AMF)

Fabrication was begun on the containment glove box for the replacement metallograph in the AMF. Performance of the existing metallograph and glove box has deteriorated with age and now is inadequate for acceptable ceramographic analysis or interfacing with image analysis equipment. Installation of the new equipment is expected within three months.

A new stainless steel bagout hood was installed in place of a wooden hood to provide increased fire resistance and extended operational capability.

Air sampling stations have been added between filter stages on exhausts from the AMF. Preliminary analyses show ^{238}Pu concentrations downstream of the first filter stage that are 10^{-2} to 10^{-3} times the expected values even with operations on finely ball-milled powder.

SCANNING ELECTRON MICROSCOPE (SEM)

Setup and checkout of the microscope and vacuum coater at SRL began by the vendor representative. Quality of the image was found deficient, necessitating a change of printed circuits and additional checkout by the vendor in January. Additional power supply equipment will also be required to increase and regulate the supply voltage and isolate it from interference signals. Containment boxes are 50% complete.

SCRAP RECOVERY

176.6 grams of scrap ^{238}Pu from LASL were charged to the recovery line. There were no process problems. Recovery rate was limited, primarily because canyon frames could accept only a small quantity of ^{238}Pu because of equipment failures. After filling the main recovery tanks, operations were severely curtailed.

REFERENCES

1. *Handbook of Ball and Pebble Mill Operation*, Paul O. Abbé, Inc. Little Falls, New Jersey.
2. H. E. Rose and R. M. S. Sullivan, *Vibration Mills and Vibration Milling*, Constable and Co., Ltd., London 1961.
3. A. L. Bement, G. J. Dau, E. A. Evans, and R. E. Nightingale, *Quarterly Progress Report, January, February, March 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology*, USAEC Report BNWL-1058, Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Washington.
4. A. L. Bement, G. J. Dau, E. A. Evans, and R. E. Nightingale, *Quarterly Progress Report, April, May, June 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology*, USAEC Report BNWL-1144, Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Washington.
5. R. L. Brown and J. C. Richards, *Principles of Powder Mechanics*, Pergamon Press, New York 1970.

MEETINGS AND VISITORS

J. D. Watrous of DWDL, N. B. Elsner of GEES, and F. J. Jones of Bechtel visited SRL December 4 for discussions related to ^{244}Cm and ^{237}Np recovery.

T. K. Keenan of LASL visited SR December 5 and 6 for a review of the PuFF Facility and discussions of PPO processing.

J. E. Sheehan, C. W. Zeh, and D. R. Boyd visited MRC on December 18 for discussions of primary encapsulation of PPO.