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^{238}Pu FUEL FORM PROCESSES BIMONTHLY REPORT

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DISCLAIMER

CONTENTS

FOREWORD

This report is one of a series to summarize progress in the Savannah River Laboratory ^{238}Pu Fuel Form Program. This program is supported primarily by the DOE Advanced Nuclear Systems and Projects Division (ANSPD).

Goals of the Savannah River Laboratory (SRL) program are to provide technical support for the production of $^{238}PuO_2$ fuel forms in the Savannah River Plant's (SRP) Plutonium Fuel Form (PuFF) Facility. This part of the program includes:

Demonstration of processes and techniques, developed by the Los Alamos Scientific Laboratory (LASL) for production at SRP. Information from the demonstration will provide the technical data for technical standards and operating procedures.

Technical Support to assist plant startup and to ensure 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 SRP in making process improvements that increase efficiency and product reliability, and to adapt plant facilities for new products.

GENERAL PURPOSE HEAT SOURCE (GPHS) PROCESS DEMONSTRATION

FABRICATION TESTS OF GPHS FUEL FORMS

Full-scale fabrication tests continued in the Plutonium Experimental Facility (PEF) with the successful fabrication of seven additional GPHS pellets (Tables 1 and 2). Three pellets (GPHS Pellets 14, 15, and 16) were fabricated at off-centerline conditions to help define process limits for production of GPHS fuel pellets in the Plutonium Fuel Fabrication (PuFF) Facility. Two additional limit-test pellets (GPHS Pellets 12 and 13) previously[1] hot pressed underwent final heat treatment. Two pellets (GPHS Pellets 17 and 18) were fabricated at centerline conditions as part of the effort to have Savannah River Laboratory (SRL) GPHS pellets impact tested at LASL. All seven pellets remained integral and demonstrated excellent dimensional stability during final heat treatment (Tables 3 and 4). However, the quality of those pellets fabricated at centerline conditions was superior to those that were fabricated as part of the limit tests (compare Figure 1 with Figures 2, 3, and 4).

GPHS Pellets 12 and 13 were fabricated to test the effect of varying the temperature at which the hot pressing force is initiated. For centerline conditions, the hot pressing force is initiated at 1350°C. The hot pressing force was initiated at 1100°C and 1500°C for GPHS Pellets 12 and 13, respectively. All other process conditions were centerline except for the use of a fast preload.

GPHS Pellet 14 was hot pressed at a final temperature of 1575°C. Centerline final temperature specified by SRL is 1525°C.

GPHS Pellet 15 was fabricated in an attempt to produce a high-density [about 86%-theoretical density (TD)] pellet. In this test, the charge to the hot press die was increased by ~3 g, and the final load was increased from 2600 to 2800 lb. This final force was increased to bring about die closure in about the same time as with a nominal die charge.

GPHS Pellet 16 was hot pressed to test the effect of using shards sintered at 1050°C instead of 1100°C. During hot pressing, the final temperature was held at 1475°C, instead of 1525°C, because a pellet fabricated under these conditions should undergo maximum shrinkage during final heat treatment.

GPHS pellets 17 and 18 were fabricated using centerline conditions.

TABLE 1

Process Conditions Used to Fabricate GPHS Pellets 12-18

^{16}O Exchange (simulated)	4 hr @ 800°C
Outgas	1 hr @ 1000°C
Ball Mill	12 hr @ 100 rpm
Compact	58,000 psi
Granulate	<125 μm
Sinter Shard	60%, 6 hr @ 1100°C* 40%, 6 hr @ 1600°C
Hot Press	See Table 2
Heat Treatment	6 hr @ 1525°C

* For GPHS Pellet 16, temperature was 1050°C.

TABLE 2

Hot Pressing Conditions for GPHS Pellets

GPHS Pellet No.	12	13	14	15	16	17	18
Preload, lb	300	250	225	200	200	200	200
Rate	Fast	Fast	Fast	Slow	Slow	Slow	Slow
Heating							
Time to 1100°C, min	3	3	3	3	3	3	3
Final Temp, °C	1530	1530	1575	1525	1475	1525	1530
Time to Final Temp, min	8	7	8	8	6	7	7
Force							
Temp of Initiation, °C	1100	1500	1350	1350	1350	1350	1350
Final Force, lb	2600	2600	2600	2800	2600	2600	2600
Ramp, min	5	5	5	5	5	5	5
Time Between Initiation of Heat and Force, min	3	5	5	4	3.5	3	4
Time to Die Closure after Final Force, Min.	1.5	1	1	1	2	3	3
Time at Final Force and Temp after Closure, Min.	5	5	5	5	5	5	5

TABLE 3

GPHS Pellet Characteristics

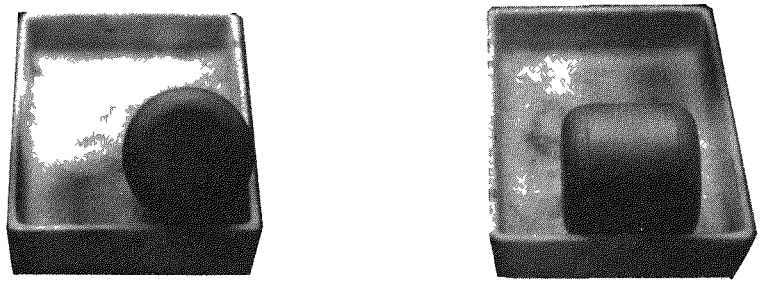
Pellet No.	Condition	Diameter, in.	Length, in.	Weight, g	Density, % TD	O/Pu
4	As Pressed	1.100	1.104	151.450	81.8	1.90
	Heat Treated	1.096	1.100	152.367	83.3	
	Difference	-0.4%	-0.4%	0.917	1.5	
5	As Pressed	1.095	1.097	151.707	83.3	1.93
	Heat Treated	1.092	1.093	152.351	84.3	
	Difference	-0.3%	-0.4%	0.644	1.0	
7	As Pressed	1.093	1.099	152.864	84.0	1.93
	Heat Treated	1.089	1.096	153.470	85.2	
	Difference	-0.4%	-0.3%	0.606	1.2	
8	As Pressed	1.098	1.112	155.582	83.7	1.92
	Heat Treated	1.095	1.108	156.300	84.9	
	Difference	-0.3%	-0.4%	0.418	1.2	
9	As Pressed	1.093	1.098	151.790	83.5	1.93
	Heat Treated	1.093	1.099	152.400	83.7	
	Difference	0	0.1	0.610	0.2	
10	As Pressed	1.094	1.100	151.582	83.0	1.91
	Heat Treated	1.090	1.095	152.365	84.5	
	Difference	-0.4%	-0.5%	0.783	1.5	
11	As Pressed	1.094	1.096	151.589	83.4	1.91
	Heat Treated	1.091	1.092	152.437	84.6	
	Difference	-0.3%	-0.4%	0.848	1.2	
12	As Pressed	1.092	1.096	151.740	83.7	1.93
	Heat Treated	1.088	1.092	152.418	85.1	
	Difference	-0.4%	-0.4%	0.678	1.4	
13	As Pressed	1.094	1.099	151.880	83.3	1.92
	Heat Treated	1.090	1.096	152.573	84.5	
	Difference	-0.4%	-0.3%	0.693	1.2	
14	As Pressed	1.096	1.098	151.560	82.9	1.89
	Heat Treated	1.093	1.094	152.516	84.2	
	Difference	-0.3%	-0.4%	0.956	1.3	
15	As Pressed	1.092	1.103	154.819	84.9	1.93
	Heat Treated	1.093	1.102	155.449	85.2	
	Difference	+0.1%	-0.1%	0.630	0.3	
16	As Pressed	1.091	1.095	151.407	83.8	1.96
	Heat Treated	1.092	1.096	151.774	83.8	
	Difference	+0.1%	-0.1%	0.367	0	
17	As Pressed	1.093	1.098	151.690	83.4	1.93
	Heat Treated	1.091	1.094	152.290	84.4	
	Difference	-0.2%	-0.4%	0.600	1.0	
18	As Pressed	1.091	1.095	151.664	83.9	1.93
	Heat Treated	1.086	1.088	152.316	85.6	
	Difference	-0.5%	-0.6%	0.652	1.7	

TABLE 4

GPHS Hot Press Tests

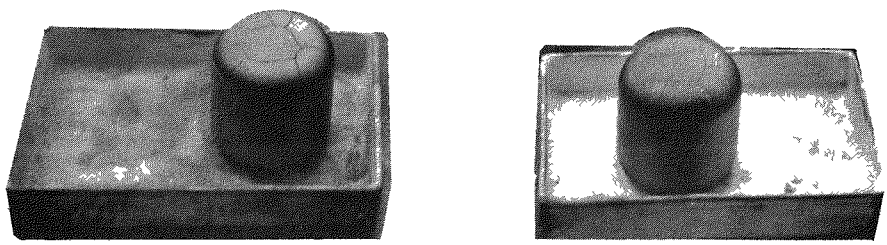
GPHS Pellet No.	Hot Press Test Condition	Centerline Condition	Final* Diameter, in.	Final* Length, in.	Shrinkage, %	Final Density, % TD	Microstructure	Comments
4	Slow heat to temp (18 min) Slow load ramp (10 min)	8 min 5 min	1.096	1.100	-0.4	83.3	Homogeneous with uniform density distribution	
5	Min time at max temp. and load (2 min)	5 min	1.092	1.093	-0.35	84.3	Homogeneous with uniform density distribution; high density aggregrates	
7	Centerline	-	1.089	1.096	-0.35	85.2	Homogeneous with uniform density distribution	Survived repeated thermal shock from 800 to 400°C
8	High Die Charge (156.7 g)	152.4 g	1.095	1.108	-0.35	84.9	No analysis	1/8-in. hole drilled from top to center. No surface cracks after final heat treatment
9	Centerline	-	1.093	1.099	0.05	83.7	Homogeneous with uniform density distribution	Thermally shocked 3 times from ~1500°C to 200°C in 1-1/2 hr because of furnace failure
10	Fast Preload (<30 Sec)	5-8 min	1.090	1.095	-0.45	84.5	Uniform with laminar cracks	No surface cracks after final heat treatment
11	Evacuate (16 hr) Fast Preload	~1 hr	1.091	1.092	-0.35	84.6	Uniform with laminar cracks	No surface cracks after final heat treatment
12	Initiate load at 1100°C Fast Preload	1350°C	1.088	1.092	-0.4	85.1	Uniform with laminar cracks	Fractured during sectioning
13	Initiate load at 1500°C Fast Preload	1350°C	1.090	1.096	-0.35	84.5	Uniform with surface cracks	Fractured after sectioning
14	High max. temp. (1575°C) Fast Preload	1525°C	1.093	1.094	-0.35	84.2	Uniform, cracked throughout cross section	Survived 14-in. drop tests
15	High Die Charge (155.4 g) High Max Load (2800 lb)	152.4g 2600 lb	1.093	1.102	0.0	85.2	Uniform, surface cracks	Fractured during sectioning
16	60% of 1050°C shards Low max temp (1475°C)	1100°C 1525°C	1.092	1.096	0.0	83.8	Unifform, some pieces badly cracked	
17	Centerline	-	1.091	1.094	-0.3	84.4	-	
18	Centerline	-	1.086	1.088	-0.55	85.6	-	

* Dies machined to production tolerances used from GPHS test 9-16.



Bottom Side

FIGURE 1. Bottom and Side Surfaces of GPHS Pellet 18 after Heat Treatment. Only one small hairline crack was observed on bottom surface.



GPHS Pellet 12 GPHS Pellet 13

FIGURE 2. Cracks on Top Surfaces of GPHS Pellets 12 and 13 after Heat Treatment

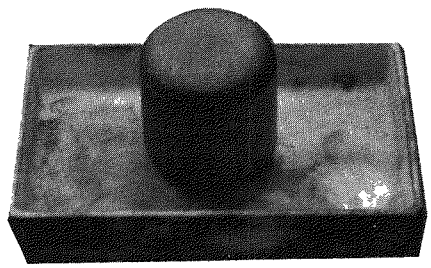


FIGURE 3. Small Hairline Cracks on Top Surface of GPHS Pellet 14 after Heat Treatment

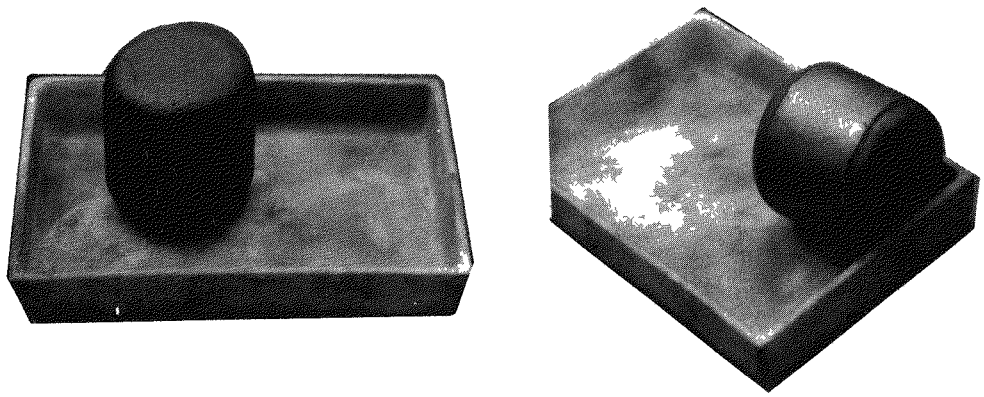


FIGURE 4. Cracks on Top and Sides of GPHS Pellet 16 after Heat Treatment

PELLET PHYSICAL CHARACTERISTICS

GPHS pellets of reasonably good quality continue to be fabricated. As with previous pellets made with the reference shard mixture, all pellets fabricated during this reporting period were integral and well formed as pressed with no apparent surface cracks. All pellets were integral after final heat treatment and demonstrated good dimensional stability (Tables 3 and 4). Surface cracking was much more evident in pellets fabricated under off-centerline conditions (GPHS Pellets 12-16) than in pellets fabricated under centerline conditions.

The characteristics of GPHS Pellets 12 and 13 after final heat treatment indicate that the temperature at which the hot pressing force is initiated affects pellet quality, especially surface cracking. Both of these pellets had about five large external cracks on the pellets' end surface after final heat treatment (Figure 2). These pellets also fractured during or after sectioning for microstructural analysis. Pellets made under SRL centerline conditions had only hairline surface cracks after final heat treatment (Figure 1) and can be sectioned without fracture for microstructural analysis. The primary cause of these differences between physical characteristics of GPHS Pellets 12 and 13 and pellets fabricated under centerline conditions is attributed to the temperature at which the hot pressing force was initiated. This temperature was 1100°C and 1500°C, respectively, for GPHS Pellets 12 and 13 compared to 1350°C for SRL centerline conditions.

The characteristics of GPHS Pellet 14 after final heat treatment indicate that increasing the final hot pressing temperature to 1575°C, 50°C above centerline temperature, has minimal effect on pellet quality. This pellet had a few hairline cracks on one end after final heat treatment, and the other end was crack free (Figure 3). GPHS Pellet 14 then survived a 14-inch drop test; no change in surface quality was observed. Finally, GPHS Pellet 14 did not fracture during sectioning.

The density of GPHS Pellet 15 after final heat treatment was lower than the expected 86% TD. Although the as-pressed density was 84.9% TD, the final density was only 85.2% TD. This lower density resulted because the pellet did not shrink during final heat treatment. If the pellet diameter and length had shrunk the expected 0.003 in. (0.3%), the final density would have been 86% TD. The absence of slight shrinkage during final heat treatment may indicate internal cracking. Both Los Alamos Scientific Laboratory (LASL) and SRL have identified a threshold for cracking at 85 to 86% TD. GPHS Pellet 15 was sectioned for microstructural analysis.

The poor surface characteristics of GPHS Pellet 16 after final heat treatment is another indication that pellet quality degrades as process conditions deviate from centerline conditions. For GPHS Pellet 16, shards sintered at 1050°C instead of 1100°C were used, and the final hot pressing temperature was maintained at 1475°C instead of 1525°C. After final heat treatment, the pellet was integral, but surface cracks were present over the entire pellet surface (Figure 4). Although essentially no shrinkage occurred during final heat treatment (Table 3), the cracking probably prevented the measurement of normal shrinkage, and the cracking may be a result of excessive shrinkage.

GPHS Pellets 17 and 18, fabricated using SRL centerline conditions, were integral and well formed both as pressed and after final heat treatment (Figure 2). No surface cracks were observed on the as-pressed pellets, and only one or two hairline cracks were visible on one end of each pellet after final heat treatment.

MICROSTRUCTURAL ANALYSES OF GPHS PELLETS 9 THROUGH 16

Microstructural analyses of GPHS Pellets 9 through 16 indicated that the degree of cracking in GPHS fuel pellets is sensitive to deviations from centerline process conditions, but that the microstructural uniformity is not sensitive to such deviations. All of the GPHS parametric pellets were integral after heat treatment and were suitable for encapsulation, but the deviations observed in the degree of cracking might affect their impact behavior.

The relationship between deviations from GPHS centerline fabrication conditions and pellet microstructure have previously been established.[2] A series of parametric experiments (fabrication of GPHS Pellets 10-16) was designed to provide data on which to base the Technical Standards for GPHS fuel fabrication in the PuFF Facility. The process conditions used to fabricate GPHS Pellets 9-16 and the physical properties of the pellets are described in this and in previous reports.[1,2] Longitudinal sections were cut from GPHS Pellets 9-16 and were prepared by standard metallographic techniques. The specimens were examined metallographically in the as-polished and acid-etched conditions.

General Microstructural Observations

The density and microstructure of GPHS Pellets 9-16 were generally fairly uniform throughout the pellet cross sections, however, high-density regions (as in GPHS Pellet 5)[3] were observed throughout GPHS Pellet 15 (Figure 5). These high-density regions

most likely resulted from self-sintering of the shards during storage. The shard structure and large intershard pores were retained in all of the parametric pellets. Slightly lower densities (~2% TD lower) were observed near the surface of the pellets. These density and microstructure results indicate that deviations from process centerline conditions do not significantly affect the microstructural properties of GPHS pellets.

Degree of Fracture

The fracture characteristics of GPHS parametric pellets varied considerably. The fracture pattern and the degree of fracture were found to be dependent upon process conditions. Pellets fabricated using off-centerline process conditions were more severely fractured than were centerline pellets. A brief summary of the degree of fracture and the fracture patterns for GPHS Pellets 9-16 appears below:

GPHS Pellet 9 [(Figure 6) Centerline conditions, but was thermal shocked three times (see Reference 2)]

Some surface cracks (~0.2 in. long) and some internal cracking, primarily near the center of the pellet, were observed. Internal cracking was not previously observed in SRL "centerline" pellets.

GPHS Pellet 10 [Fast Preload (see Reference 2) (no micrographs available, specimen was lost during grinding)]

Numerous cracks were observed originating at the surface of the pellet and running perpendicular to the surface tangent. Some of these cracks traversed the entire cross section. No crack branching was apparent. Apparently the fast preload creates high surface tensile stresses in the pellet because the material has less time for rearrangement and/or gases are more readily entrapped during compaction. The microstructure of this pellet was similar to that of GPHS Pellet 12 (see Figure 7).

GPHS Pellet 11 [Fast preload, then evacuated 16 hr prior to hot pressing (see Reference 2); no micrographs available, specimen was lost during grinding)]

The crack pattern of GPHS Pellet 11 was very similar to that observed in GPHS Pellet 10 and GPHS Pellet 12 (see Figure 7). This pattern appears to confirm the belief that (1) fast preload is detrimental to pellet integrity and (2) the 16-hr evacuation prior to hot pressing has no effect on pellet integrity.

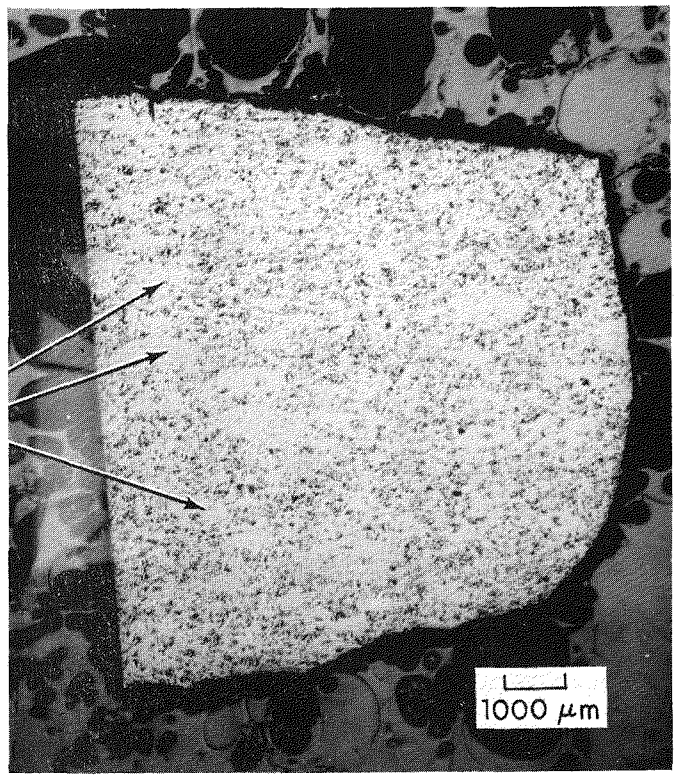


FIGURE 5. GPHS Pellet 15 with High-Density Regions

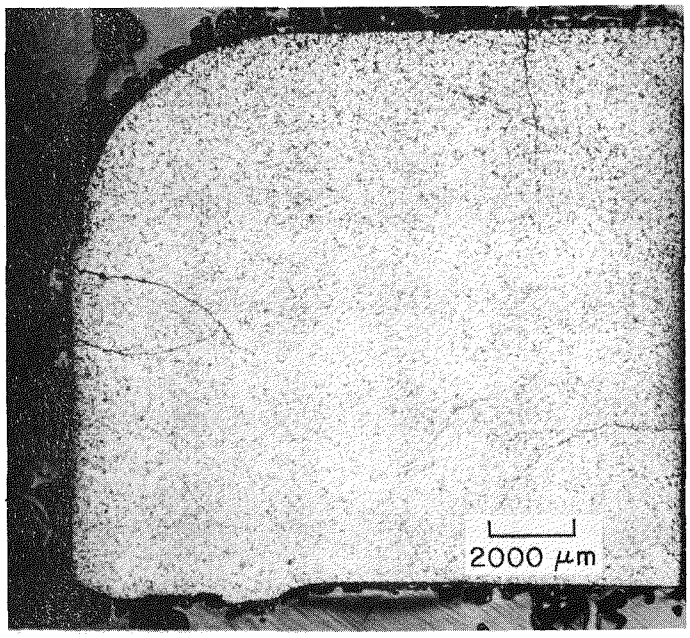


FIGURE 6. Longitudinal Section of GPHS Pellet 9

FIGURE 7. Longitudinal Section of GPHS Pellet 12

GPHS Pellet 12 [(Figure 7) Fast preload, initiated load at 1100°C]

Numerous fine cracks were observed throughout the cross section of GPHS Pellet 12. Most, but not all, of these cracks originated at the surface of the pellet.

GPHS Pellet 13 [(Figure 8) Fast preload, initiated load at 1500°C]

Only a few surface cracks (<0.2 in. long) were apparent in the two pellet sections that were examined. The presence of these cracks indicates that early (at 1100°C) load initiation is much more detrimental to pellet integrity than is late (at 1500°C) load initiation. Moreover, one may conclude that late initiation of the preload seems to help compensate for the undesirable effects of fast preload. This conclusion appears to have a theoretical basis in that at 1500°C (versus 1100°C), the sintering kinetics of PuO_2 are much greater and the high surface tensile stresses, which apparently result from a fast preload, are more rapidly relieved by diffusional mechanisms (sintering).

GPHS Pellet 14 [(Figure 9) Fast preload, high maximum temperature]

GPHS Pellet 14 was cracked throughout the cross section that was analyzed. Fewer cracks were observed in GPHS Pellet 14 than in GPHS Pellets 10 to 12; however, many of the cracks in GPHS Pellet 14 traversed the entire quadrant. Unlike previous GPHS pellets, GPHS Pellet 14 showed considerable crack branching (bifurcation). The presence of bifurcation indicates a more rapid rate of crack propagation in GPHS Pellet 14 than rates observed in previously studied GPHS pellets. Some of the crack branching could have resulted from the rapid propagation of cracks when the pellet was dropped 14 inches. However, the pattern shown in Figure 9 indicates that the cracks originated at the top, bottom, and side of the pellet. This crack pattern is not characteristic of what would be expected to result from a single impact. More likely, the higher hot pressing temperature probably allowed more reduction and higher reoxidation stresses. These reoxidation stresses, in conjunction with surface tensile stresses related to the fast preload, probably increased the energy available for crack propagation.

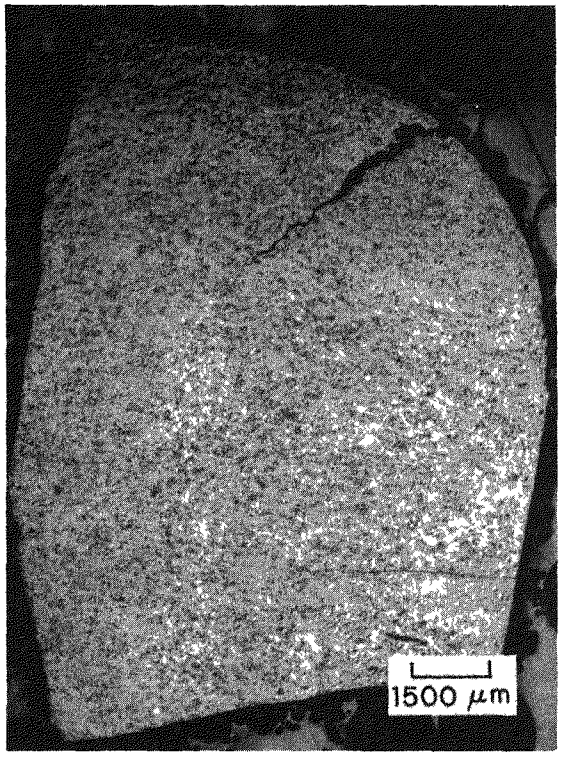


FIGURE 8. Longitudinal Section of GPHS Pellet 13

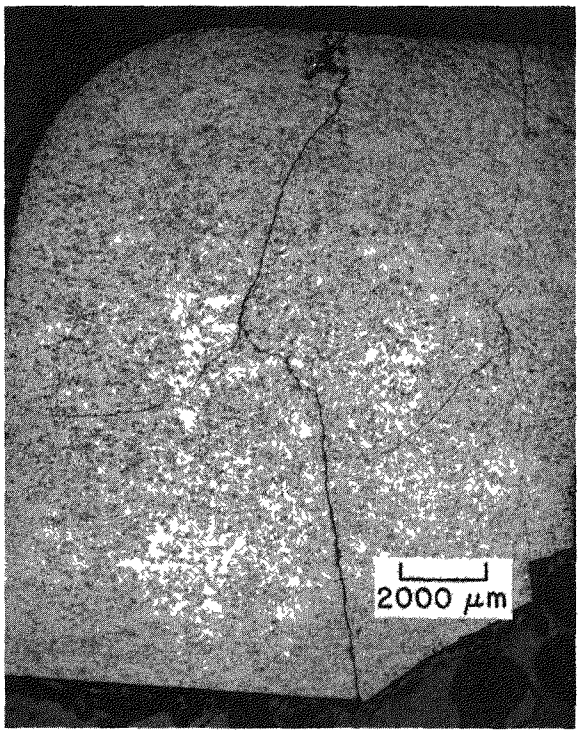


FIGURE 9. Longitudinal Section of GPHS Pellet 14

GPHS Pellet 15 [(Figure 10) High die charge (+3 g), high maximum load (2800 lb)]

GPHS Pellet 15 was characterized by numerous surface cracks (<0.2 in. long). Most of the observed cracks originated at the surface and ran normal to the surface tangent. The degree of cracking was less severe than in GPHS Pellets 10 to 12. This observation indicates that the off-centerline process conditions of high die charge and high maximum load cause surface tensile stresses of slightly lower magnitude than the off-centerline condition of fast preload rate.

GPHS Pellet 16 [(Figure 11) 60% 1050°C shards, low maximum hot press temperature (1475°C)]

The off-centerline conditions for GPHS Pellet 16 were chosen to maximize pellet shrinkage during final heat treatment. Although the percent shrinkage during heat treatment was very small, the additional stresses created by these off-centerline parameters were sufficient to degrade the integrity of the pellet. GPHS Pellet 16 fragmented during sectioning. The four largest pieces were prepared for metallography. Whereas the radiused corner shown in Figure 11a was nearly crack free, one of the fragments which was examined was severely cracked (Figure 11b).

PELLET DIMENSIONS VERSUS DIE CAVITY

Hot press die assemblies machined to production-grade tolerances were used beginning with the fabrication of GPHS Pellet 9. As shown in Table 5, good agreement was obtained between dimensions of the die cavity and those of the pellets even with the variations in process conditions used i the limit tests. The difference between the final pellet diameter and the original die diameter ranged from -0.0001 to -0.008 in. The difference between the final pellet length and the original length of the die cavity ranged between +0.006 to -0.006 in.

O/Pu RATIO IN AS-PRESSED GPHS PELLETS

Carbothermic reduction of the PuO_2 results in the formation of a suboxide during hot pressing (Table 3). This reduction reaction is a strong function of temperature. GPHS Pellet 14, which was pressed at 1575°C, was reduced to an O/Pu ratio of 1.89, whereas the O/Pu ratio of GPHS Pellet 16, which was hot pressed at 1475°C, was 1.96. All of the other pellets, which were hot

pressed at about 1525°C, had O/Pu ratios of 1.91-1.93 as pressed. GPHS Pellet 4 had an O/Pu ratio of 1.90, but this pellet remained at elevated temperatures about twice as long as did the other pellets hot pressed at 1525°C.

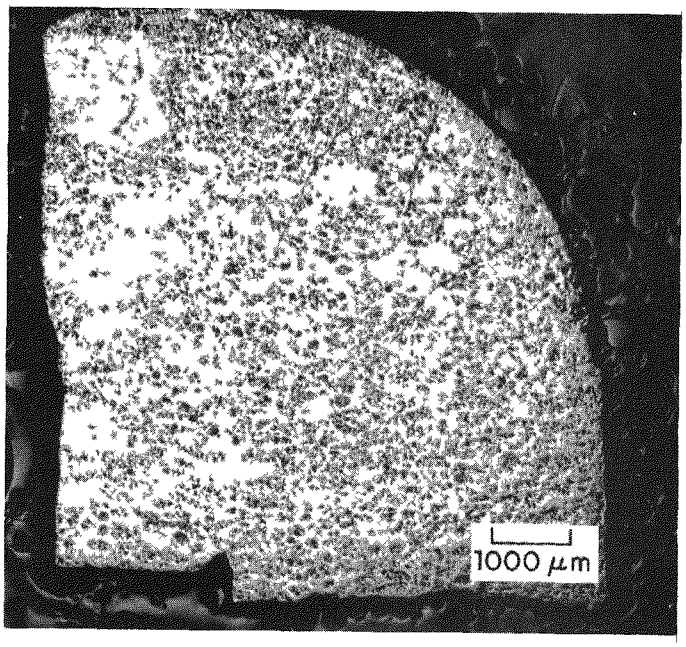
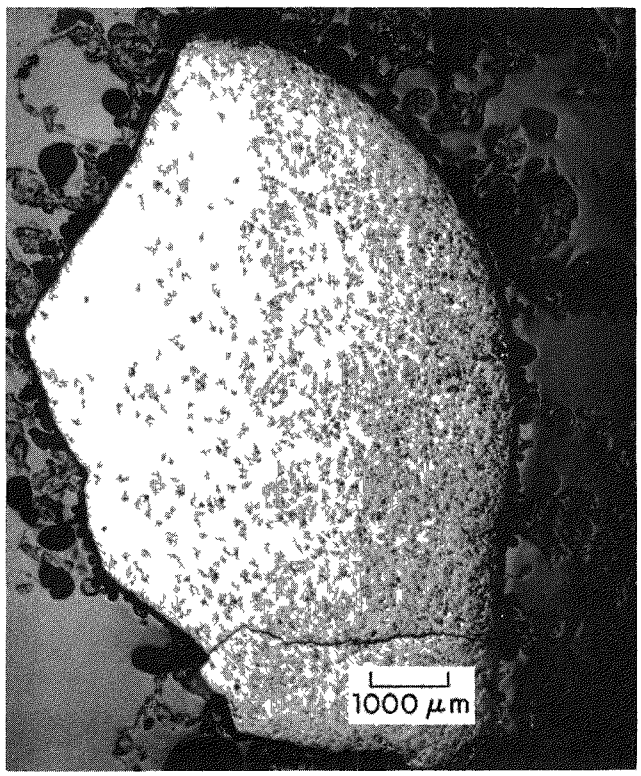
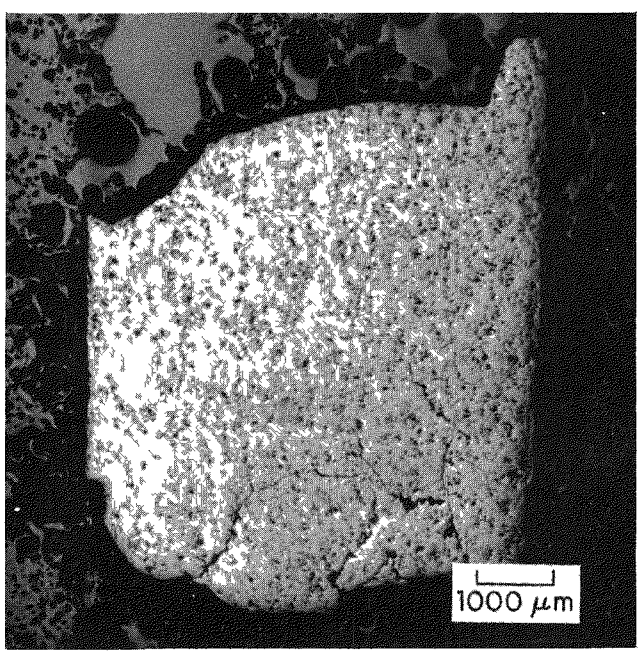


FIGURE 10. Longitudinal Section of GPHS Pellet 15



a. Radiused Corner



b. Edge

FIGURE 11. Longitudinal Section of GPHS Pellet 16

TABLE 5

Pellet Dimensions Versus Die Cavity Dimensions*

GPHS Pellet No.	Diameter, in. Die	As-Pressed Pellet	Heat-Treated Pellet	Diametral Change, in.	Length, in. Die	As-Pressed Pellet	Heat-Treated Pellet	Linear Change, in.	Comments
9	1.094	1.093	1.093	-0.001	1.097	1.098	1.099	+0.002	Pellet thermally shocked three times
10	1.094	1.094	1.090	-0.004	1.096	1.100	1.095	-0.001	Limit test
11	1.094	1.094	1.091	-0.003	1.097	1.096	1.092	-0.005	Limit test
12	1.094	1.092	1.088	-0.006	1.097	1.096	1.092	-0.005	Limit test
13	1.094	1.094	1.090	-0.004	1.098	1.099	1.096	-0.002	Limit test
14	1.094	1.096	1.093	-0.001	1.095	1.098	1.094	-0.001	Limit test
15	1.094	1.092	1.093	-0.001	1.096	1.103	1.102	+0.006	High-density attempt
16	1.094	1.091	1.092	-0.002	1.096	1.095	1.096	0	Limit test
17	1.094	1.093	1.091	-0.003	1.098	1.098	1.094	-0.004	Centerline
18	1.094	1.091	1.086	-0.008	1.096	1.095	1.088	-0.006	Centerline
	Average Shrinkage Die Versus Pellet			-0.0033				-0.0016	
	Standard Deviation			±0.0023				±0.0037	

* Measurement error is estimated to be ±0.001 in.

Complete reoxidation of the hot-pressed pellets to PuO_2 occurs only during final heat treatment. Some reoxidation occurs if the GPHS pellet is exposed to the argon atmosphere of the glove box, which may contain several thousand ppm O_2. However, as shown in Table 6, the reoxidation rate is quite slow. The O/Pu ratio in GPHS Pellet 12 had increased from 1.925 to 1.948 after 162 hr of exposure to the glove box atmosphere. These O/Pu data confirm earlier data[3] which showed the same slow rate of reoxidation in GPHS pellets exposed to the glove box atmosphere.

TABLE 6

Reoxidation of GPHS Pellet 12

Weight, g	Elapsed Time, hr	O/Pu
151.740	As Pressed	1.925
151.801	20	1.932
151.927	48	1.946
151.902	118	1.943
151.912	140	1.944
151.948	162	1.948
152.418	Heat Treated	2.000

GPHS PELLET SHIPPING CONTAINER

Preliminary design was completed for a primary shipping container to contain an unencapsulated pellet. This shipping container will be used to ship SRL GPHS pellets to LASL for encapsulation and impact testing. A hollow graphite cylindrical insert will serve as an interface between the pellet sides and the stainless steel container. Graphite felt will serve as the interface at the ends of the pellet.

The design of the pellet shipping container is a modified version of the EP 60 container which is used to ship $^{238}PuO_2$ powder. The outside diameter of the shipping container is the same as that of the EP 60 container. Thus, the shipping container with pellet can be removed from the process line and loaded into the EP 61 secondary container using the breechlock loader/unloader that is normally used for $^{238}PuO_2$ powder. Prior to shipment, the cap of the EP 61 container will be welded to the secondary container body.

IMPACT TESTING OF SRL GPHS PELLETS

SRL GPHS pellets made under centerline conditions will be sent to LASL for encapsulation and impact testing. LASL recommends impact testing of SRL GPHS pellets because SRL found significant microstructural differences between GPHS pellets fabricated at SRL and those fabricated at LASL.[1] LASL believes that a number of impact tests of SRL GPHS centerline pellets are necessary to provide adequate data for a meaningful comparison of the impact behavior of LASL and SRL GPHS pellets.

SRL will supply additional pellets for encapsulation and impact testing. GPHS Pellet 18 will be the first pellet to be impact tested. GPHS Pellet 17, which was also fabricated under SRL centerline conditions, will be sectioned for microstructural analysis.

FUTURE WORK

Additional pellets will be fabricated at centerline conditions for encapsulation and impact testing at LASL. GPHS Pellet 19, the final pellet in the initial set of limit tests, will be fabricated to test the effect of using shards sintered at 1150°C instead of 1100°C. Microstructural analysis of GPHS Pellets 14, 15, 16, 17, and 19 will be completed. Additional full-scale fabrication tests will be based on the results of the characterization data obtained on GPHS Pellets 10-19.

MULTI-HUNDRED WATT (MHW) PROCESS SUPPORT

INCREASED THERMAL LOADING FOR MHW FUEL SPHERES

Background

In response to a DOE directive, thermal loading of MHW spheres was increased from a nominal 102.5 watts to 103.5 watts. DOE required this change to be implemented by increasing the sphere weight rather than increasing the ^{238}Pu isotopic content. Because dimensional specifications were unchanged, the increased weight increased the sphere density ~1-1.5% (from ~81 to ~82-82.5% TD). By itself this small increase in density was not expected to influence fracture resistance; however, since increased die charge affects die closure, fracture resistance could be significantly reduced if hot pressing conditions were not adjusted to compensate for increased die charge.

Based on the results of metallographic analyses of both parametric and quality assurance (QA) production spheres and on the statistical analysis of production spheres through MHW Sphere 58[3], a number of changes in the hot pressing procedure were recommended.

Two of the recommended changes were to (1) maintain pressure during cooldown and (2) increase time at temperature after die closure.

Since these two recommended process changes were adopted, ten of thirteen 103.5-watt, MHW spheres produced in the Plutonium Fuel Fabrication (PuFF) facility and adequate fracture resistance.

The following paragraphs summarize the recommended process changes that (1) helped to minimize adverse changes in sphere fracture resistance and (2) led to successful conversion to the higher-density spheres.

Recommended Process Changes

The following PuFF process changes to allow for increased die charges were recommended (Table 7):

1. Allow at least 7 minutes from indicated die closure until start of power ramp descent (10-15 min is probably the most desirable range). The total time at temperature should be increased to accommodate this recommendation if necessary.

2. Maintain the hot pressing load during cooldown to ambient temperature and then decrease load slowly (100-200 lb/min).

3. Increase the hot pressing load from 2500 to 2550 lb, in 25-lb increments, if necessary to ensure die closure.

TABLE 7

Recommendations for Hot Pressing 103.5-Watt MHW Spheres

1. Elapsed time from indicated die closure to start of power ramp down.	7 minutes
2. Maintain hot pressing load until cooldown to ambient temperature.	
3. Hot pressing load.	Up to 2550 lb if needed to close die.

Fabrication Results

Table 8 describes spheres that were made to test the effects of changes in hot pressing conditions on sphere properties. Spheres 135, 137, 140, and 142-145 were integral when Recommendations 1 and 2 were followed. Spheres 133 and 134 had a load of 2550 lb to compensate for the greater die charge and probably would have been integral if Recommendations 1 and 2 had been followed. However, the results of these tests through Sphere 145 suggest that pressures >2500 lb are not needed to close the die even with thermal loads up to or slightly greater than 104 watts. The fracture of Sphere 136 probably reflects the short elapsed time after die closure before start of cooldown. Spheres 138 and 139 were made using Recommendations 1 and 2 and 2600 lb load (versus the 2500 lb normally used). The fracture of these spheres can be attributed to too high a hot pressing load. Twenty-six hundred pounds, therefore, represents an upper load limit. A limit lower than 2470 lb (as used in pressing Spheres 144 and 145) has not been established, but statistical analysis predicted that lowering the load led to increased fracture tendency. The hot pressing temperature was held constant at the present level (~1550°C) for all spheres. Again, statistical analysis predicted that increasing sphere density by increasing hot press temperature led to greater fracture tendency.

TABLE 8

Fabrication Conditions for 103.5-Watt Test Spheres

Sphere	Nominal Watts	Die Charge, g	Elapsed Time After Die Closure, min	Hot Press Load, lb	Temperature When Load Was Removed, °C	Condition of Sphere
133	104.2*	249.4	3-4	2550	~1550	Intact after heat treatment, fractured when dropped.
134	104.0*	248.8	3-4	2550	~1550	Intact after heat treatment but broke into 3 pieces before gaging.
135	103.6*	247.8	7	2500	~1180	Integral. Hairline pole-to-bellyband crack.
136	103.8*	248.3	2-3	2450	~1200	Broken into 2 halves as pressed.
137	103.6*	247.8	7	2500	Ambient	Integral. Pole-to-bellyband crack wider than hairline.
138	103.4	248.2	11	2600	<1100	Cleaved after heat treatment. Three pieces when welded.
139	103.5	247.9	24	2600	<1100	Three pieces after heat treatment. Recycled due to fracture.
140	103.5	248.0	10	2500	<1100	Integral when welded.
141	103.5	248.0	14	2500	<1100	Two pieces after heat treatment.
142	103.6	248.1	12	2500	<1100	Integral when welded.
143	103.5	248.8	7	2500	<1100	Integral when welded. No observable cracks.
144	103.4	248.0	52	2470	<1100	Integral when welded; pole-to-pole crack after heat treatment.
145	103.4	248.0	34	2470	<1100	Integral when welded.

* Specific power = 0.418 W/g.

Bases for Recommendations

Recommended process changes were based on results of both the statistical analysis of MHW production data through Sphere 583 and metallographic analyses of several parametric and production spheres. The statistical analysis showed that 83%-TD spheres having the same fracture resistance as 81%-TD spheres could be expected. The analysis further showed that for the sphere density range being considered (80 to 83% TD) and for changes in shard sintering temperature, hot pressing load, and hot pressing temperature of only a few decades or less, fracture resistance increased with increasing shard sintering temperature and hot pressing load and decreased with increasing hot pressing temperature. These density-fracture resistance relationships served as guidelines during the rapid development of the proper hot pressing conditions for the 83%-TD sources.

Metallographic analyses of spheres support the statistical analysis of the MHW production data. Cracking appears to be due to rebound effects of removing the load from spheres still under compression. Spheres that were pressed in dies that were known not to close showed much more extensive cracking than did spheres that were pressed in dies that had closed 10 to 15 minutes before the end of the hot pressing run. During this time, these latter spheres had sintered away from the die wall. Spheres that indicated die closure less than 4 minutes from the end of the run also showed considerable cracking, suggesting that the spheres had still been in contact with the die wall and punches. The mechanism thought to be responsible for this cracking is expansion of compressed CO/CO_2 gas within the pores of a sphere when the load is removed, if the sphere is still under compression. If the sphere sinters away from the die wall, this pressure is relieved slowly, thus minimizing cracking. A few minutes residence time at temperature after die closure (Recommendation 1) is, therefore, necessary to permit the sphere to sinter away from the die wall. Maintaining the hot pressing load during cooldown (Recommendation 2) is a precaution for those spheres which may still be under compression at the end of a run. Internal gas pressure within the sphere decreases to less than half during cooling from 1550°C to ambient (~450-500°C for spheres). Also, the tensile strength of PuO_2 increases rapidly with decreasing temperatures. Increasing the load (Recommendation 3) decreases the intershard pore size slightly, which in turn raises the sinterability of the spheres and aids in sintering away from the die walls. Clearly this step has an upper limit of applicability since too much pressure can crush the intershard porosity and cause excessive shrinkage, density gradients, and more, not less, cracking.

Recommendation 3 was not needed to make the higher-density spheres because (1) the die closed quickly enough and the sphere sintered away from the die wall satisfactorily without requiring increased pressure, and (2) the greater pressure caused more cracking, probably by decreasing the pore size too much. However, had it been necessary to increase density of the 102-watt spheres by allowing sphere dimensions to change and keeping die load constant, an increase in hot press pressure of 50 to 100 lb would probably have been necessary.

If gas expansion is the principal cause of the extensive fracture observed on some spheres as suggested, then clearly increasing the hot pressing temperature would enhance cracking as predicted by the statistical analysis, since it leads to increases in fuel reduction and gas formation. In connection with the proposed gas mechanism, it should be remembered that as the hot press load is applied, the punch knife edges expand against the die wall, effectively sealing the die cavity and preventing escape of gas.

REFERENCES

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