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MASTER

^{238}Pu FUEL FORM PROCESSES BIMONTHLY REPORT

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DISCLAIMER

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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 by the DOE Division of Advanced Nuclear Systems and Projects (DANSP).

Goals of the Savannah River Laboratory (SRL) program are to provide technical support for the transfer of DANSP ^{238}Pu fuel form fabrication operations from Mound Laboratory to new facilities 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. The program includes:

Demonstration and adaptation of processes and techniques, developed by the Los Alamos Scientific Laboratory (LASL) and Monsanto Research Corporation (MRC), 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 of new processes 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 process efficiency and safety and product reliability, and to adapt plant facilities for new products.

GENERAL-PURPOSE HEAT SOURCE PROCESS DEMONSTRATION

The SRP Plutonium Fuel Form (PuFF) Facility will begin production of a new $^{238}PuO_2$ fuel form in 1980 for use in the General-Purpose Heat Source (GPHS), currently undergoing design, development, and testing at Los Alamos Scientific Laboratory (LASL). Savannah River Laboratory (SRL) will transfer technology from LASL on fuel form fabrication, develop the final process flowsheet, and assist Savannah River Plant (SRP) personnel in adapting the process for production of GPHS fuel in the PuFF Facility.

The 62.5-watt $^{238}PuO_2$ fuel form (an ~1.09-in. x 1.09-in. cylindrical pellet with rounded corners) and the preliminary flowsheet for fabricating the GPHS form were described previously.[1] The original flowsheet has been revised slightly to incorporate changes made in the LASL process. The revised flowsheet, summarized in Figure 1, contains several modifications to the original LASL process to accommodate equipment differences between LASL and Savannah River (SR). The modifications were similar to those that were made to the Multi-Hundred Watt (MHW) process.

To establish the final process flowsheet and to evaluate characteristics of the future GPHS product, two types of fabrication experiments are being performed. Small-scale fabrication tests (with ~6.5 g pellets) are being performed in the SRL Actinide Materials Facility (AMF) to test important non-scale-sensitive process variables and to evaluate the effects of these variables and fuel form density on microstructure and fracture behavior of GPHS fuel pellets. Full-scale testing recently began in the SRL Plutonium Experimental Facility (PEF) to 1) demonstrate and verify the LASL process, 2) adjust process variables for differences in equipment between LASL and the PuFF production facility, and 3) establish limits on key process variables. Results of these tests will be reported in this and future issues of this report series.

EFFECT OF SHARD SINTERING-TEMPERATURE ON FUEL FORM MICROSTRUCTURE

Microstructural studies of small-scale GPHS pellets indicate that a reduction in the sintering temperature of the high-fired shards from 1600°C to 1400°C has 1) only small effects on fuel microstructure and 2) apparently no effect on dimensional stability or fracture resistance of the pellets. Characterization of shards sintered at 1400 and at 1600°C revealed that both shard types have similar densities. This study is part of a

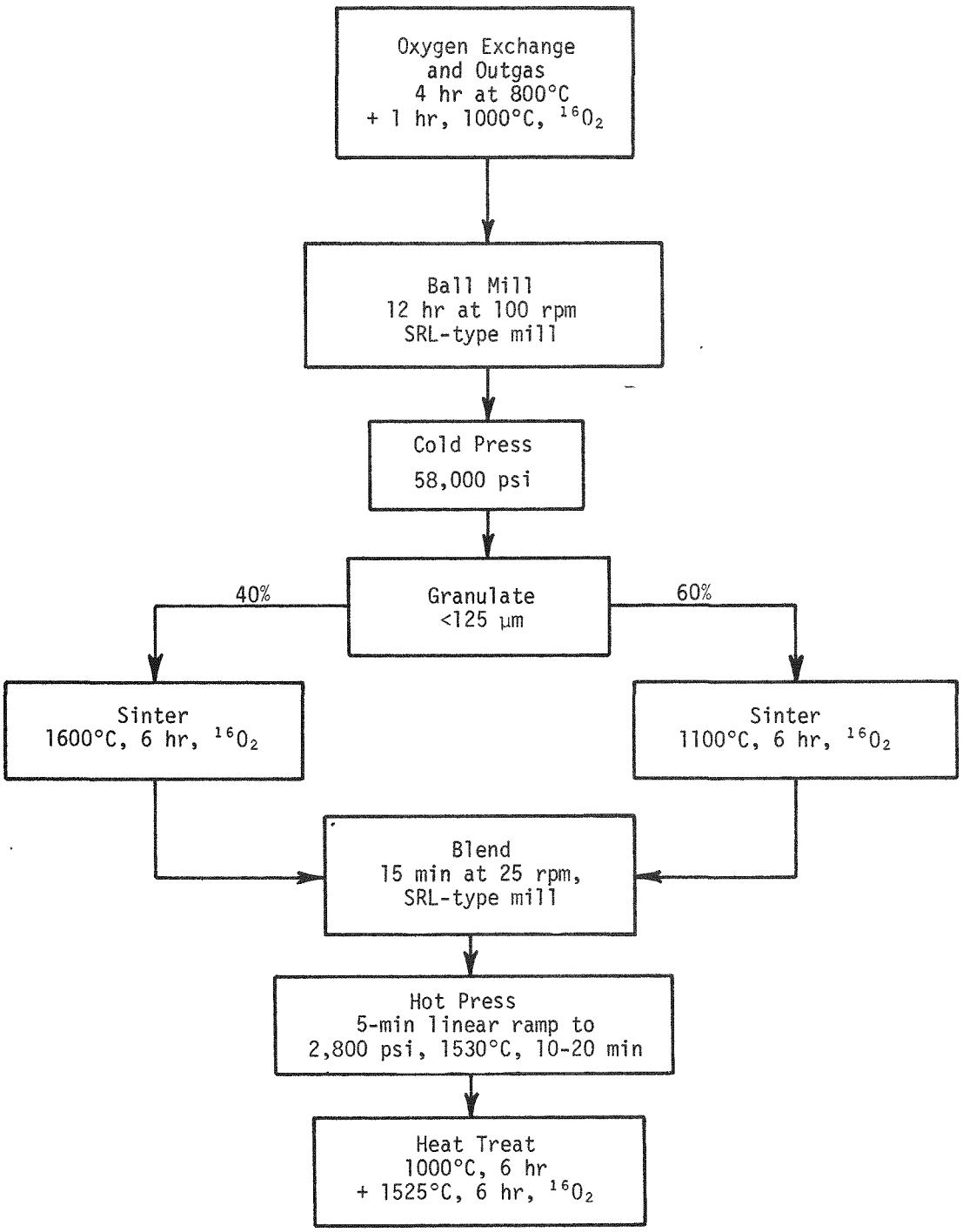


FIGURE 1. GPHS Process Flowsheet

series of small-scale experiments in the AMF to evaluate certain non-scale-sensitive process conditions for GPHS fuel forms.

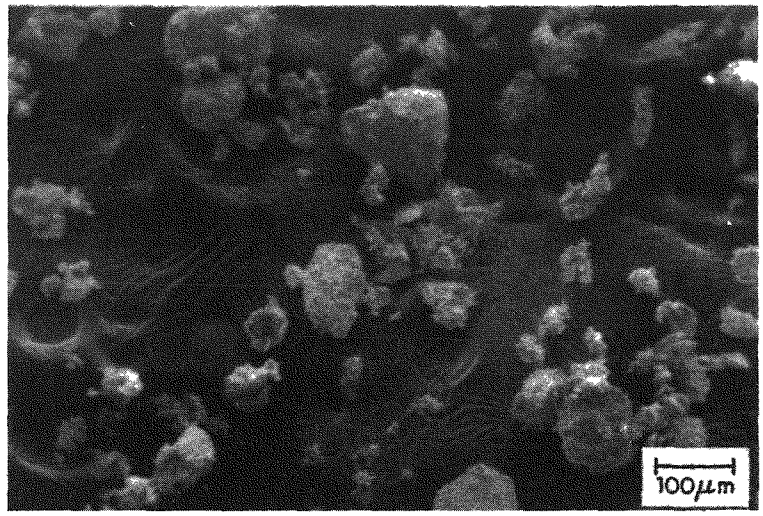
A temperature of 1600°C is required by the present LASL GPHS fuel fabrication flowsheet[1] to presinter the high-fired component of the "grog" granule feed before hot pressing. At this high temperature, the existing plutonium-rhodium furnace racks in the PuFF facility would have an unacceptably short lifetime. Furnace racks designed to accommodate higher operating temperatures will be procured for the GPHS production campaign (see below); however, the dispersion-hardened alloy to be used in the rack assemblies (ZGS Pt-10% Rh) may or may not survive the 1600°C presintering without creeping significantly. A reduction of the presintering temperature of high-fired shards to about 1500°C is being considered in case the planned creep tests, described below, indicate the need for operating the new racks at a lower temperature.

Another purpose of the shard-sintering temperature study is to investigate the sensitivity of the product to temperature variations. Further small-scale experiments in the AMF and full-scale experiments in the PEF will be required to completely evaluate the effect of shard-sintering temperature on the structure and fracture resistance of GPHS fuel.

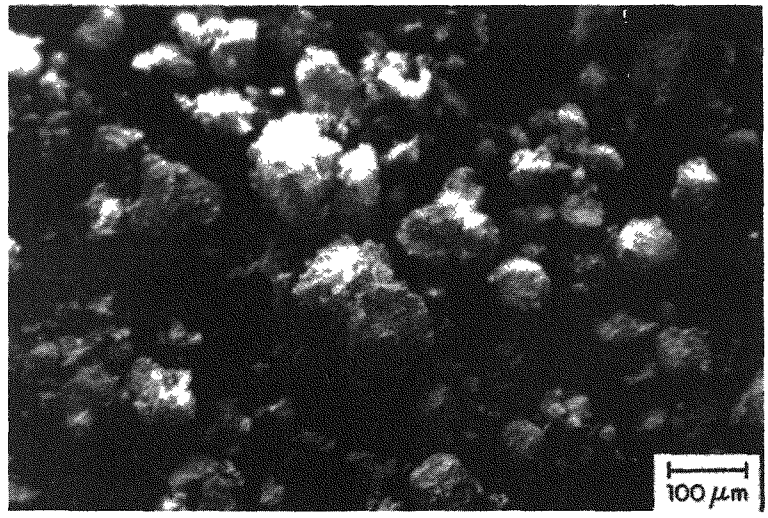
Shard Characterization

For characterizing the shards used in the sintering temperature study, two batches of feed were prepared for hot pressing, one batch according to the GPHS flowsheet[1] and the other batch from a 40%-60% mixture of shards presintered at 1400°C and 1100°C, respectively. The standard grog feed for GPHS fuel consists of 40% shards presintered at 1600°C and 60% shards presintered at 1100°C (Figure 1). Samples from each batch of high-fired shards were characterized by Scanning Electron Microscopy (SEM) and metallography.

The surface morphology of the shards was difficult to study because the shards appeared to be coated with a layer of fine, unsintered powder as shown in Figure 2. However, SEM photomicrographs of clean-fractured surfaces of shards (Figure 3) and metallography of polished surfaces of shards (Figure 4) revealed that the shards, after presintering at either 1400°C or 1600°C, were approximately 95 to 96% of theoretical density (TD). The pores in shards sintered at 1400°C were generally intragranular, and relatively stable to sintering. Therefore, shard density did not increase significantly at higher presintering temperature. However, a significant increase in the mean grain size (4.5 μm to 6.5 μm), attributed to the 200°C increase in shard presintering temperature, was observed (Figure 5).



1400°C, 6 hr



1600°C, 6 hr

FIGURE 2. Surface Morphology of Sintered Shards

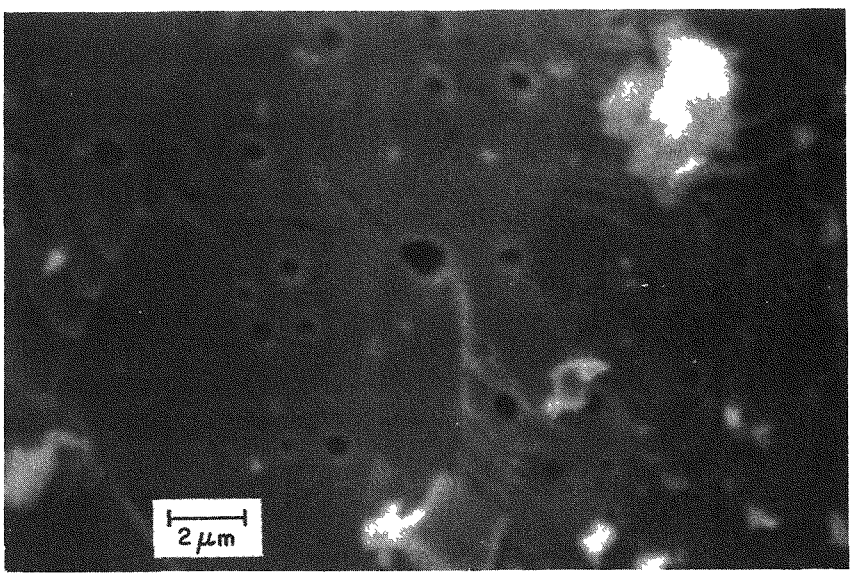
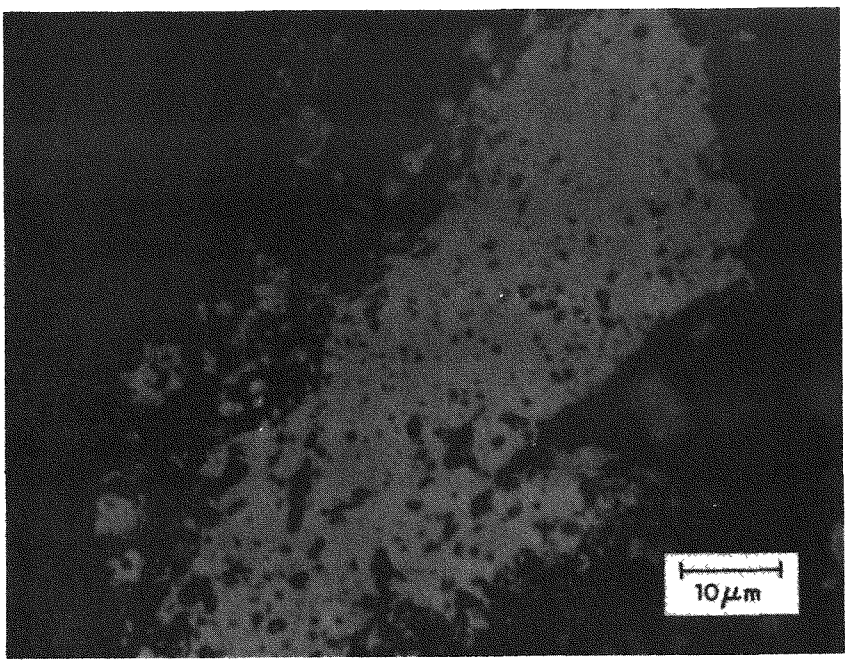
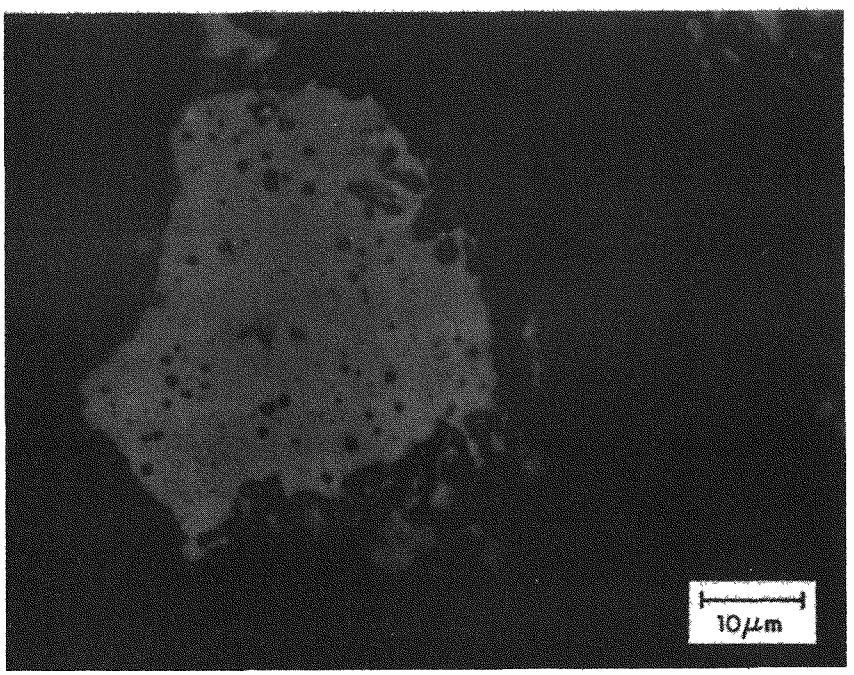


FIGURE 3. Clean-Fractured Surface of a Shard Sintered at 1400°C

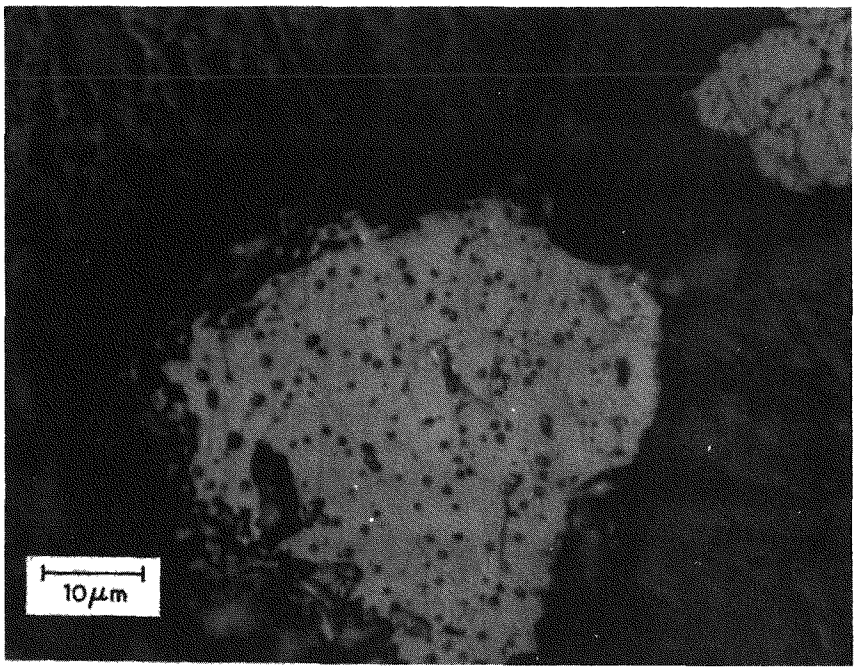


1400°C, 6 hr;
Avg Density = 95.9% TD

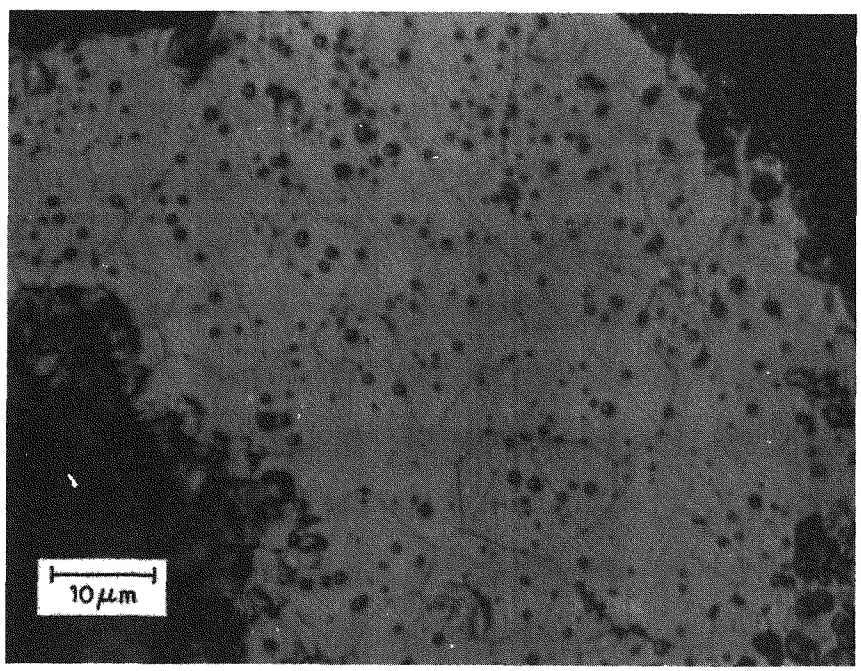


1600°C, 6 hr;
Avg Density = 96.1% TD

FIGURE 4. Effect of Sintering Temperature on Shard Microstructure (Polished Surface)



1400°C, 6 hr;
Mean Grain Size = 5 μm



1600°C, 6 hr;
Mean Grain Size = 6.5 μm

FIGURE 5. Effect of Sintering Temperature on Shard Grain Structure

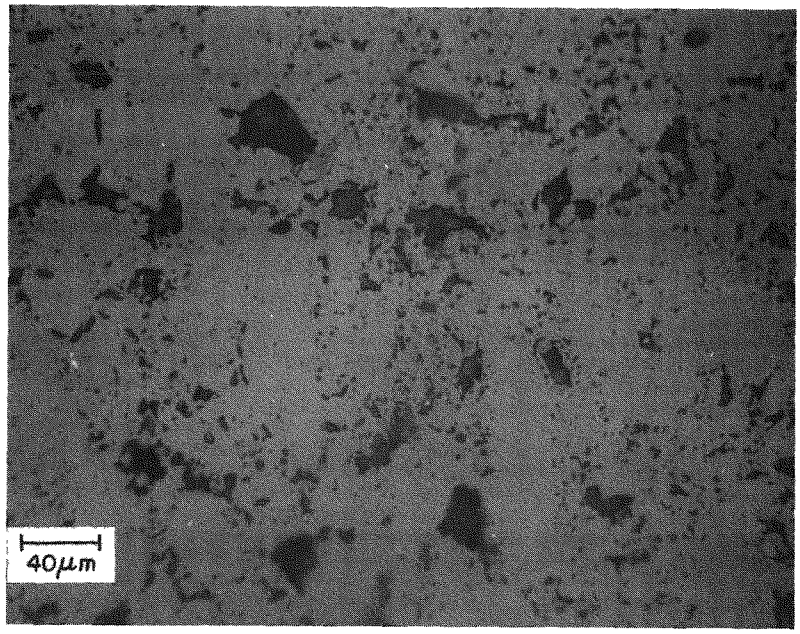
Pellet Characterization

To characterize the pellets, two pellets (3/8-in. dia., 3/8-in. high, ~6.5 g) were hot pressed in graphite dies at 5,000 psi. One was pressed from each of the two grog mixtures [MPH 103 (40% 1600°C + 60% 1100°C) and MHP 105 (40% 1400°C + 60% 1100°C)]. After pressing, the pellets were about 84% TD as determined from their dimensions and weights. The pellets were then bisected with one section of each pellet retained for microstructural analysis. The other section of each pellet was heat treated for 6 hr at 1000°C followed by 6 hr at 1350°C (original flowsheet conditions).[1] Both the as-pressed and heat-treated sections from the two pellets were examined metallographically.

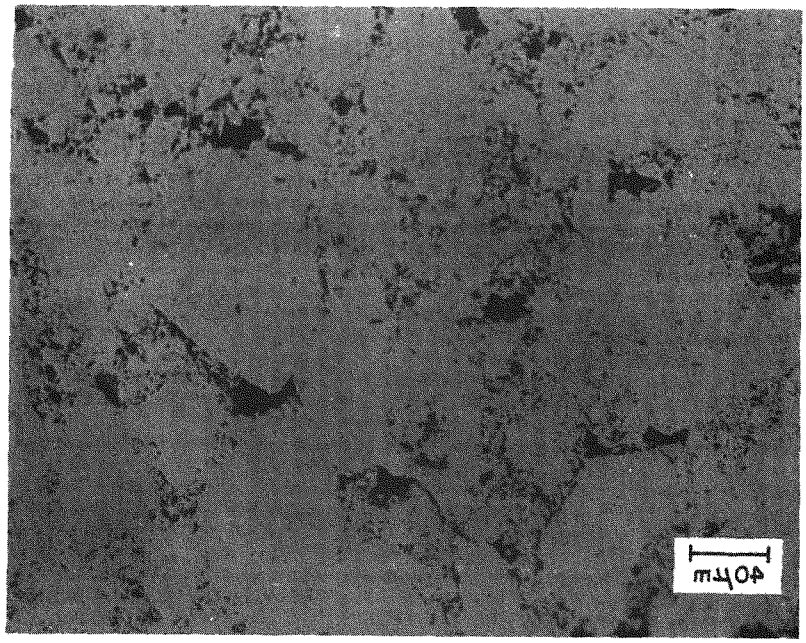
Slight, but possibly significant, microstructural effects of the reduced shard presintering temperature (1400°C versus 1600°C) were observed. The higher presintering temperature resulted in somewhat better-defined shards in the pellet microstructure for the hot pressing conditions used (Figure 6). Also, the intershard porosity differed in size and distribution for the two different shard compositions. However, the pellet made from shards presintered at 1400°C still possessed an acceptable microstructure with identifiable shards and had about the same density (determined from photomicrographs) as the pellet made from 1600°C shards. The retention of the coarse, stable intershard porosity, as shown in Figure 6, was demonstrated in microstructural studies of MHW fuel to be important to dimensional control and fracture resistance.[2]

Reoxidation cracking characteristics (extent and crack size) of both pellets did not differ significantly from each other. Induced cracking was observed after heat treatment in both pellets (Figure 7). As described in past reports and discussed in a later section of this report, reoxidation-induced cracking is caused by stresses induced by the phase change (or possibly the expansion of entrapped CO gas) that occurs when an as-pressed pellet (which has been reduced to PuO_{2-x} during hot pressing in graphite) is oxidized.

Grains in the pellets fabricated from the two shard compositions (1400°C and 1600°C) showed similar bimodal size distributions after heat treatment, as shown in Figure 8. Typical grain sizes in the heat-treated pellets were ~10 μm for shards presintered at 1100°C, ~15 μm for shards presintered at 1400°C, and ~20 μm for 1600°C shards. On the basis of grain sizes, the low- and high-fired shards were uniformly distributed in the fuel structure.

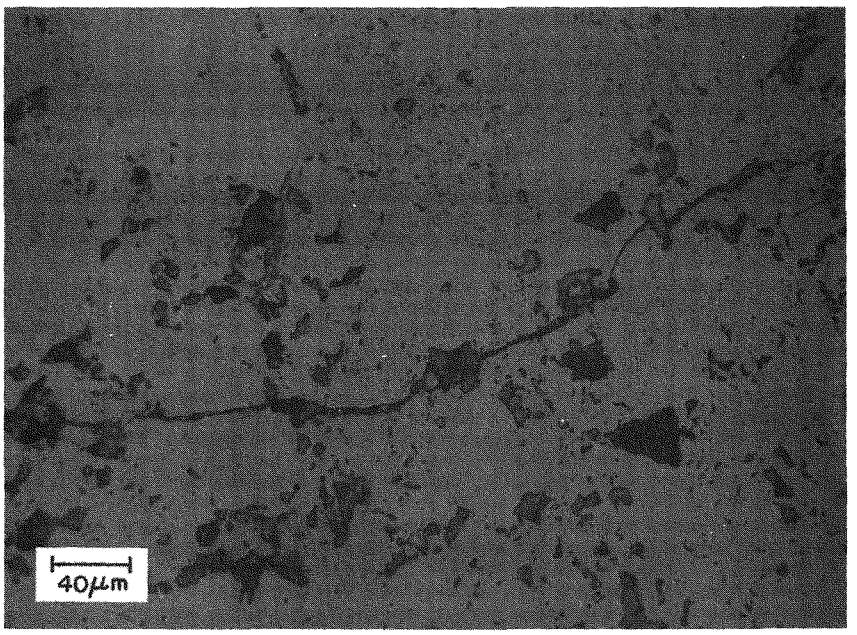


Shard Composition:
40% 1400°C + 60% 1100°C
(MHP 105)

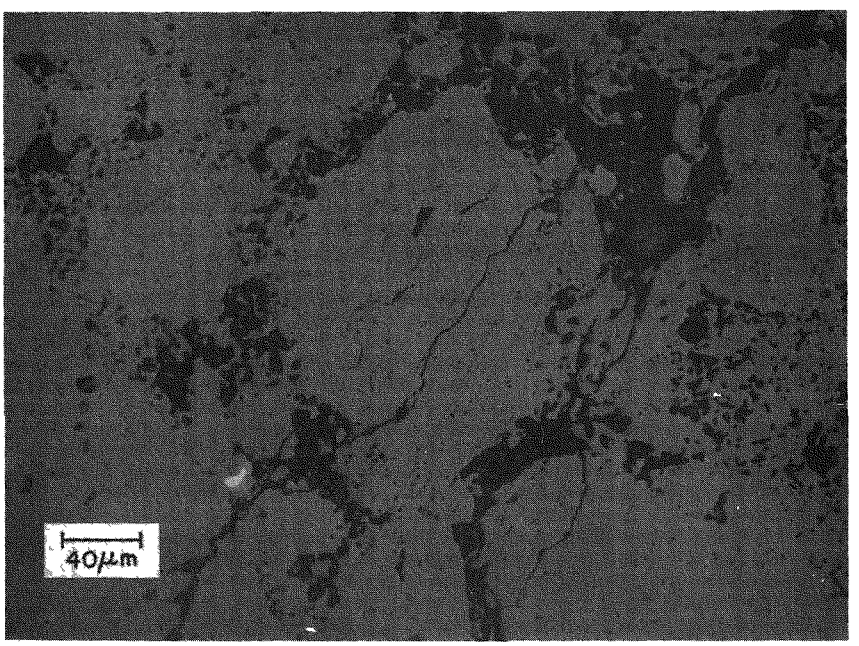


Shard Composition:
40% 1600°C + 60% 1100°C
(MHP 103)

FIGURE 6. Effect of Shard Composition on the Microstructure of GPHS Pellets, As-Pressed and As-Polished

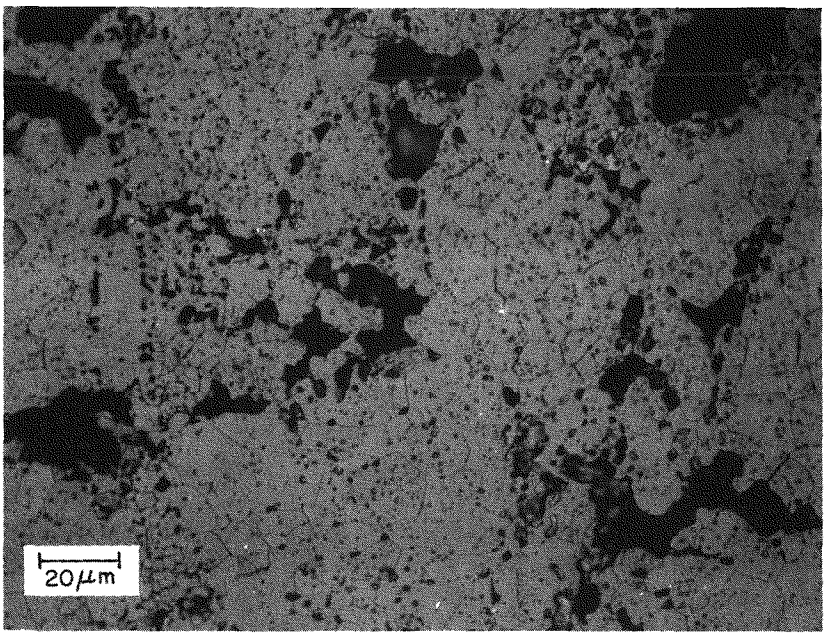


Shard Composition
40% 1400°C + 60% 1100°C
(MHP 105)

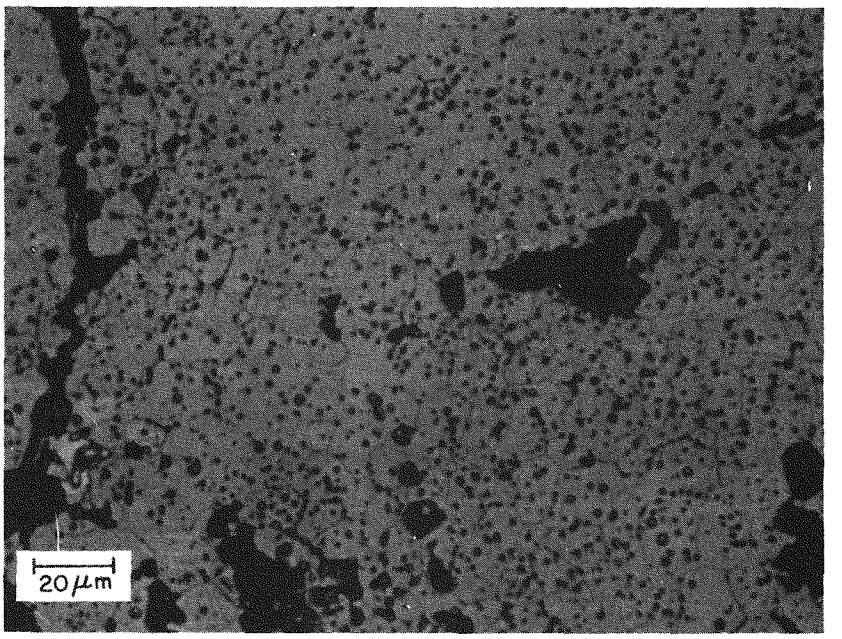


Shard Composition
40% 1600°C + 60% 1100°C
(MHP 103)

FIGURE 7. Reoxidation-Induced Fracture in Heat-Treated and As-Polished GPHS Pellets



Shard Composition
40% 1400°C + 60% 1100°C
(MHP 105)



Shard Composition
40% 1600°C + 60% 1100°C
(MHP 103)

FIGURE 8. Grain Size of Heat-Treated GPHS Pellets (Grain-Boundary Etched)

Conclusions

On the basis of the preliminary test results, substitution of shards presintered at 1400°C for high-fired (1600°C) shards in the fabrication of GPHS fuel pellets alters the microstructure slightly, but does not appear to significantly affect fuel density or fracture resistance. These results indicate that GPHS fuel comparable to forms made with the LASL flowsheet can be fabricated using an altered flowsheet in which the presintering temperature of the high-fired shard component is decreased from 1600°C to 1500°C and the hot-pressing conditions are adjusted slightly to compensate for the small resultant difference in shard characteristics. Additional experiments, including full-scale testing in PEF, are planned to verify the preliminary test results that indicate that a 100°C decrease in presintering temperature of high-fired shards does not compromise GPHS fuel form quality.

EFFECTS OF DENSITY AND REOXIDATION TEMPERATURE ON GPHS FUEL MICROSTRUCTURE

Small $^{238}PuO_2$ pellets hot pressed to different densities cracked during reoxidation in air at both low and high heat treatment temperatures. Cracking occurred in regions where microstructure exceeded about 85 to 85% TD and increased in severity with increasing density. These results indicate that cracking from reoxidation-induced stresses in GPHS fuel forms is to be expected; the severity of such cracking will depend on the homogeneity of the GPHS microstructure.

A study was undertaken as part of a series of small-scale tests that supplement full-scale GPHS fabrication tests now underway in the PEF. The objectives of these particular tests were 1) to determine the density threshold for reoxidation-induced cracking* in GPHS fuel and 2) to evaluate the effects of reoxidation temperature on this type of cracking. Previous results showed that cracking from reoxidation-induced stresses in MHW fuel occur primarily in regions of the microstructure which exceed 84 to 86% TD.[3] Because the nominal density of GPHS fuel will be 84 to 85% TD, small density gradients will cause regions of the microstructure to exceed this cracking threshold. Density gradients in MHW fuel have been measured at 10 to 15% TD.[2] GPHS fuel should be more homogeneous than MHW fuel because of the fuel form shape

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* PuO_2 is reduced by interaction with the graphite die during hot pressing to an O/Pu ratio of about 1.85. The resulting two-phase structure undergoes a phase (and volume) change on subsequent heat treatment in an oxygen atmosphere and cracks from the tensile streses induced by the volume change.

(cylindrical pellet versus sphere) and possibly because of improved compaction properties of the grog shard mixture; however, density variations of the order of 3% TD are expected. Control of processing conditions will therefore be critical to minimize density gradients and reoxidation-induced cracking in GPHS fuel.

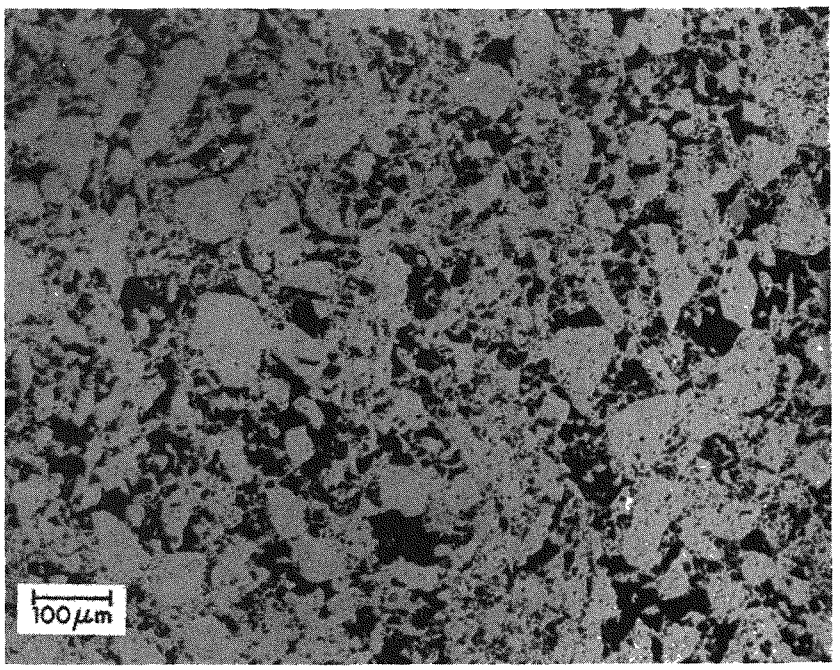
Also investigated in these tests was the effect of reoxidation temperature on cracking. LASL recommends that the as-pressed GPHS fuel form be stored in a vented graphite container for reoxidation at low temperature before heat treatment. Reoxidation at low temperature may decrease reoxidation-induced stresses and may reduce cracking during reoxidation. The reoxidation temperatures (180°C and 275°C) investigated in these tests were in the range of temperatures expected during storage of full-size GPHS fuel pellets in graphite.

Fabrication Conditions

Shards were processed according to the GPHS flowsheet[1] and hot pressed to form cylindrical-shaped pellets (3/8-in. dia, 3/8-in. high, 6.5 g) of different densities. The shard feed was a 60%-40% mixture of shards presintered at 1100°C and 1600°C, respectively. The pellets were hot pressed to different densities by altering the pressing pressure. Pressures of 4,000 psi (MHP 102), 5,000 psi (MHP 103), and 20,000 psi (MHP 101) produced pellets with bulk densities of 80%, 84%, and 91% TD, respectively. These lower two densities represent the minimum and average densities anticipated in full-scale fuel forms. The maximum bulk density slightly exceeds the maximum density expected in full-size forms due to processing variations and normal density gradients. The pellets were sectioned into quarters; one section was retained for as-pressed metallography and the remaining sections were heat treated at 180°C, 275°C, or 1350°C. These sections were then characterized metallographically.

Microstructure

The microstructures of the as-pressed fuel sections reflect the variant bulk densities of the pellets. In the 80%-TD pellet, the individual shards were well defined and were separated by coarse intershard porosity (Figure 9). No macrocracks were found in the as-pressed structure. Pellets hot pressed to a bulk density of 84% TD were also essentially crack-free in the as-pressed condition. The intershard pores in the 84%-TD fuel were smaller than those in the 80%-TD pellet; however, the basic shard structure was fully retained, and the microstructure was generally homogeneous (Figure 10). In the 91%-TD as-pressed pellet, radial tensile cracks had formed at the surface, and the shard structure had been completely eliminated (Figure 11).



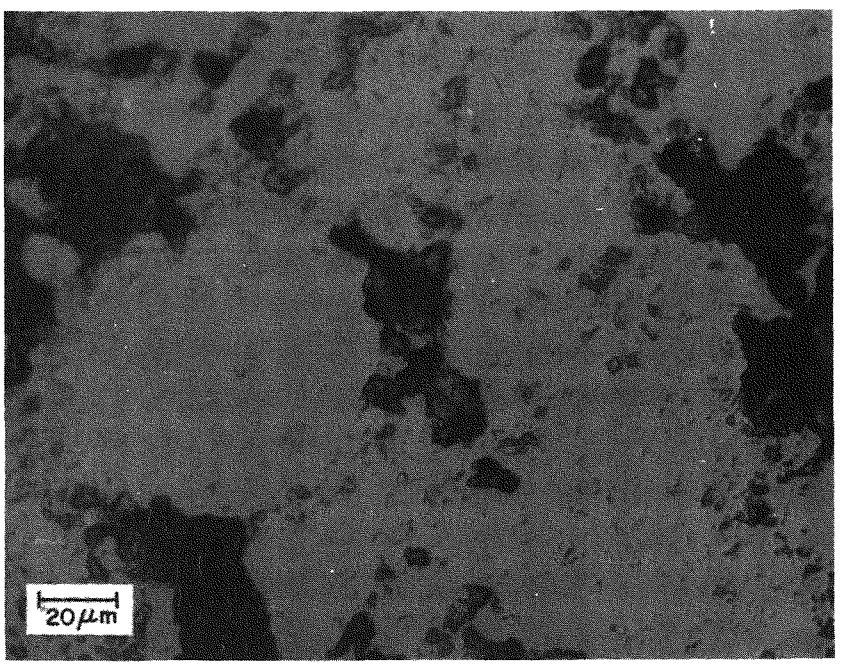
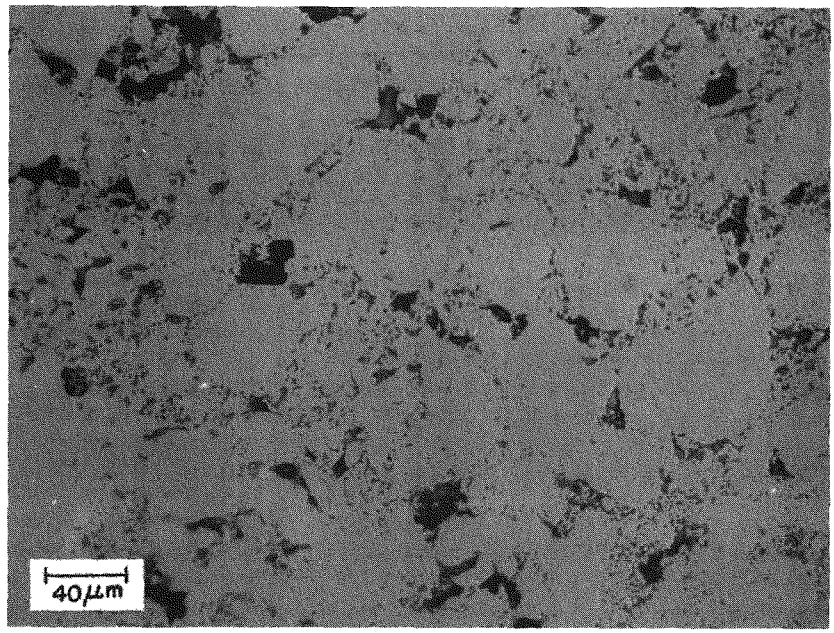


FIGURE 9. Coarse Intershard Porosity in As-Pressed, 80%-TD Pellet MHP 102 (Polished Surface)



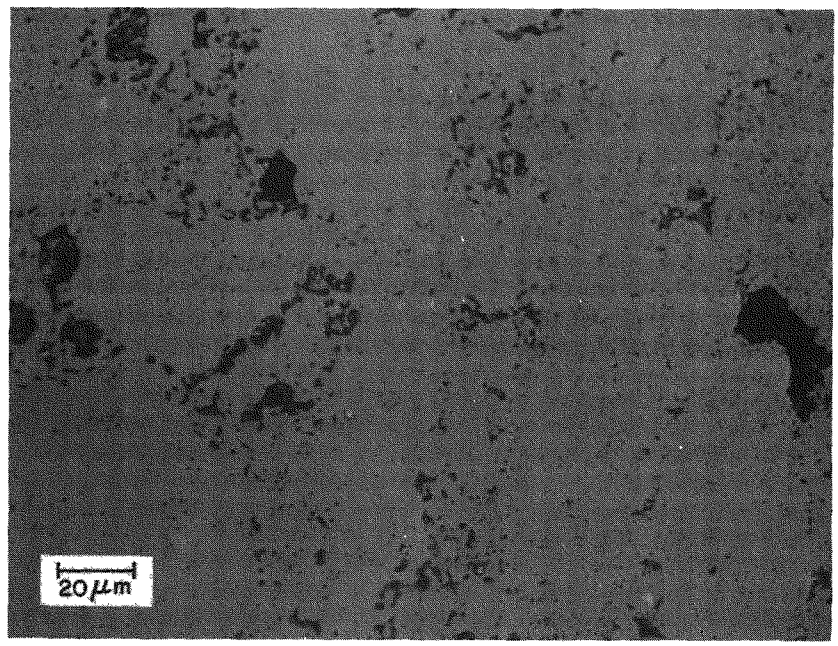


FIGURE 10. Microstructure of As-Pressed, 84%-TD Pellet MHP 103 (Polished Surface)

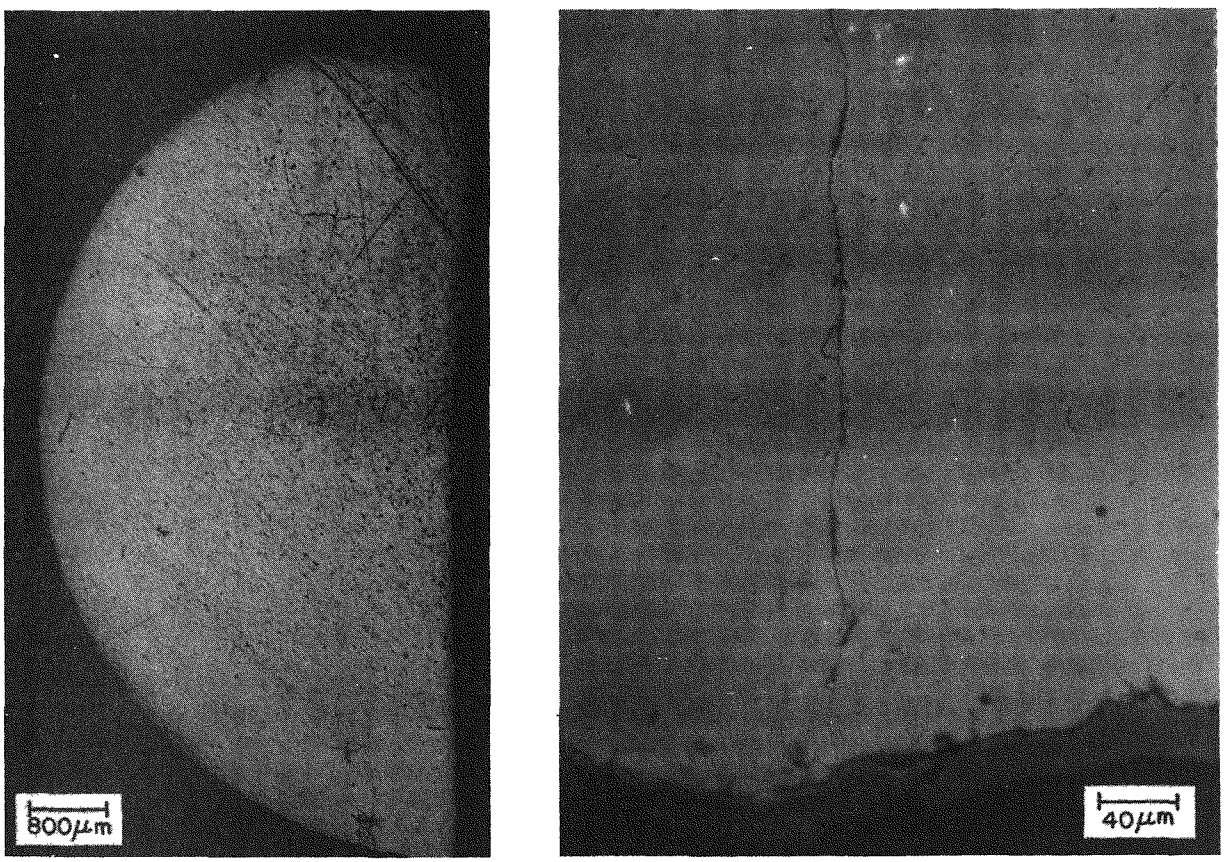
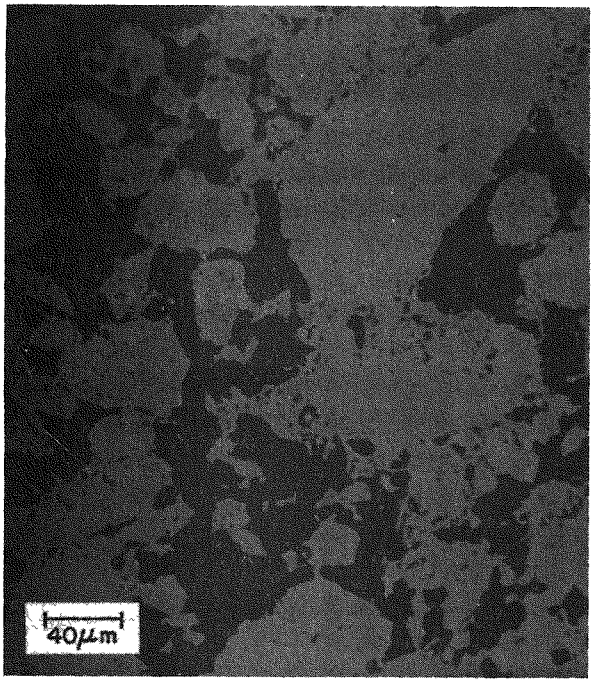


FIGURE 11. Radial Tensile Cracks at the Surface of 91%-TD Pellet MHP 101, As Pressed (Polished Surface)

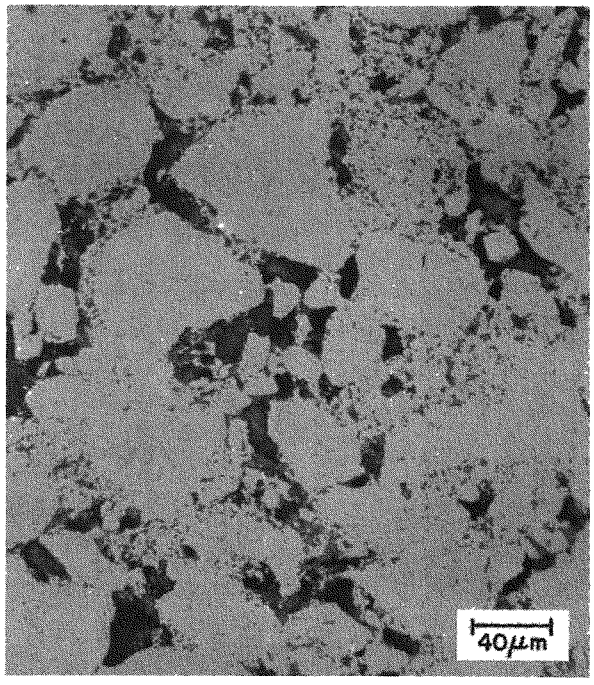
As expected, heat treatment of the as-pressed pellet sections in air at either low or high temperature caused cracking that increased in severity with increasing fuel density. The 80%-TD pellet sections remained nearly crack-free after heat treatment at 180°C, 275°C, or 1350°C (Figure 12). The 84%-TD pellet sections, after heat treatment at the same conditions, developed a few relatively long cracks (Figure 13) as a result of internal stresses caused by reoxidation. Previous cold-press sinter tests had indicated that this type of cracking was definitely the result of hot pressing in graphite.[3] Thermal stresses were too small to affect the observed cracking because of the small size of the pellets. The extent and severity of reoxidation-induced cracking increased considerably in the 91%-TD pellet (Figure 14). The crack pattern of Figure 14 indicates that the entire cross section of the pellet had been subjected to localized tensile stresses induced by reoxidation.

The extent of cracking did not appear to be affected significantly by reoxidation temperature in either the 84%-TD (Figure 13) or 91%-TD pellets. Cracking might be related to the rate of reoxidation and thus might depend on the concentrations of oxygen present during heat treatment. In that case, reoxidation in the argon cell atmosphere at low temperature, as recommended by LASL, would mitigate reoxidation-induced cracking.

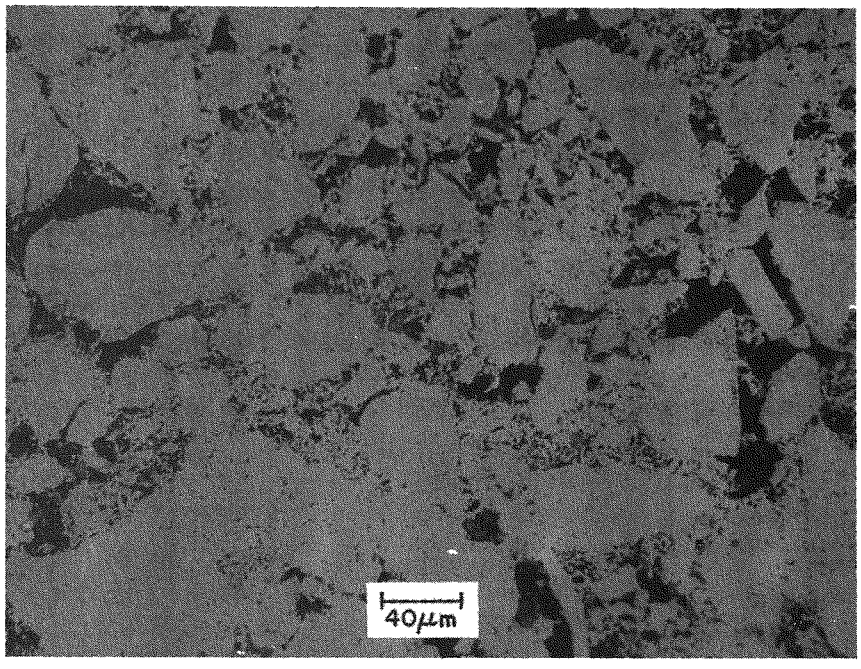
Although no apparent difference in cracking behavior of PuO_2 fuel sections reoxidized at low and high temperatures was observed, the microstructures of these fuel sections differed significantly (Figure 15). After reoxidation at low temperature, an apparent remnant or image structure (small pores outlining location of suboxide phase in as-pressed microstructure) remained in the fuel. Heat treatment at 1350°C eliminated most of the remnant pore structure, although stringers of porosity were still present within some shards.



5 hr at 180°C

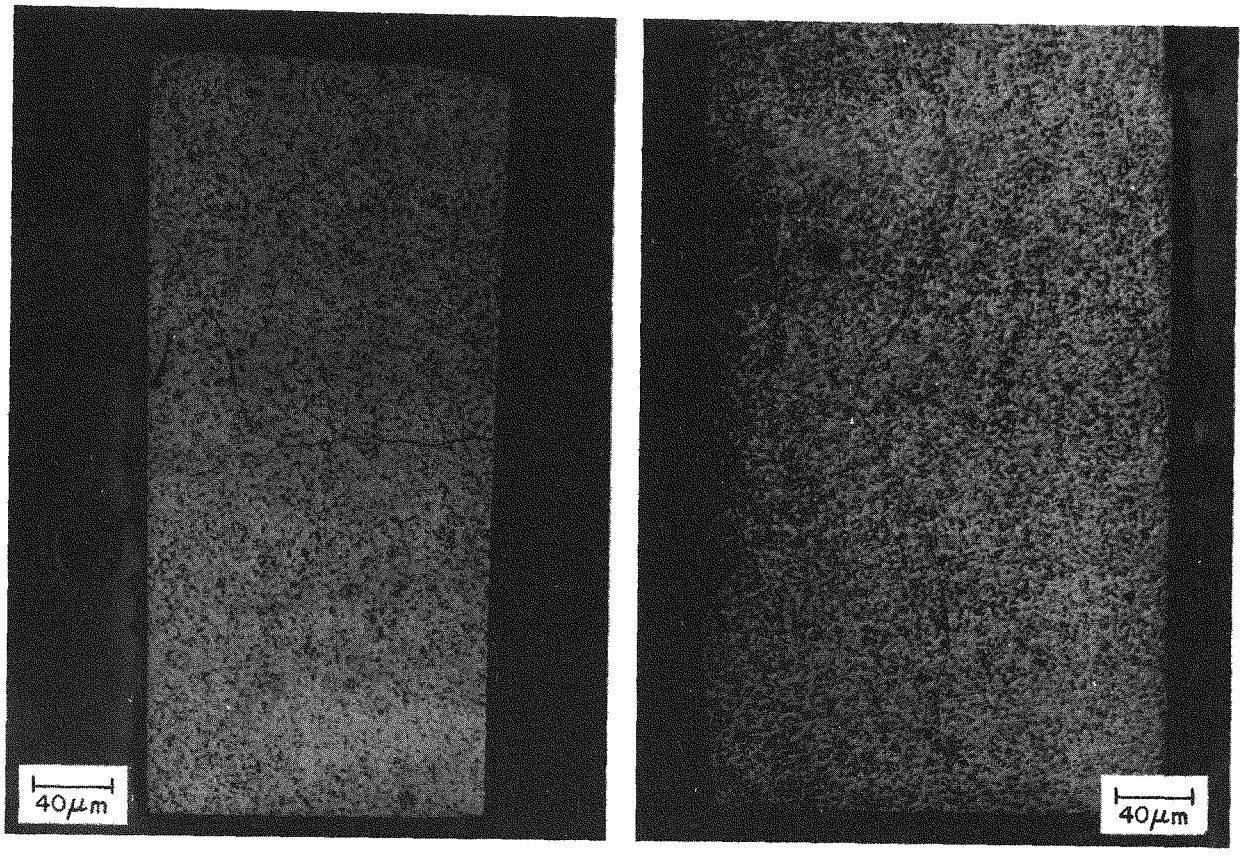


5 hr at 275°C



6 hr at 1000°C and
6 hr at 1350°C

FIGURE 12. Effect of Heat Treatment Temperature on the Microstructure of an 80%-TD Pellet MHP 102 (Polished Surface)



5 hr at 180°C Heat Treatment

6 hr at 1000°C and 6 hr at 1350°C Heat Treatment

FIGURE 13. Reoxidation-Induced Cracking in 84%-TD Pellet MHP 103 (Polished Surface)

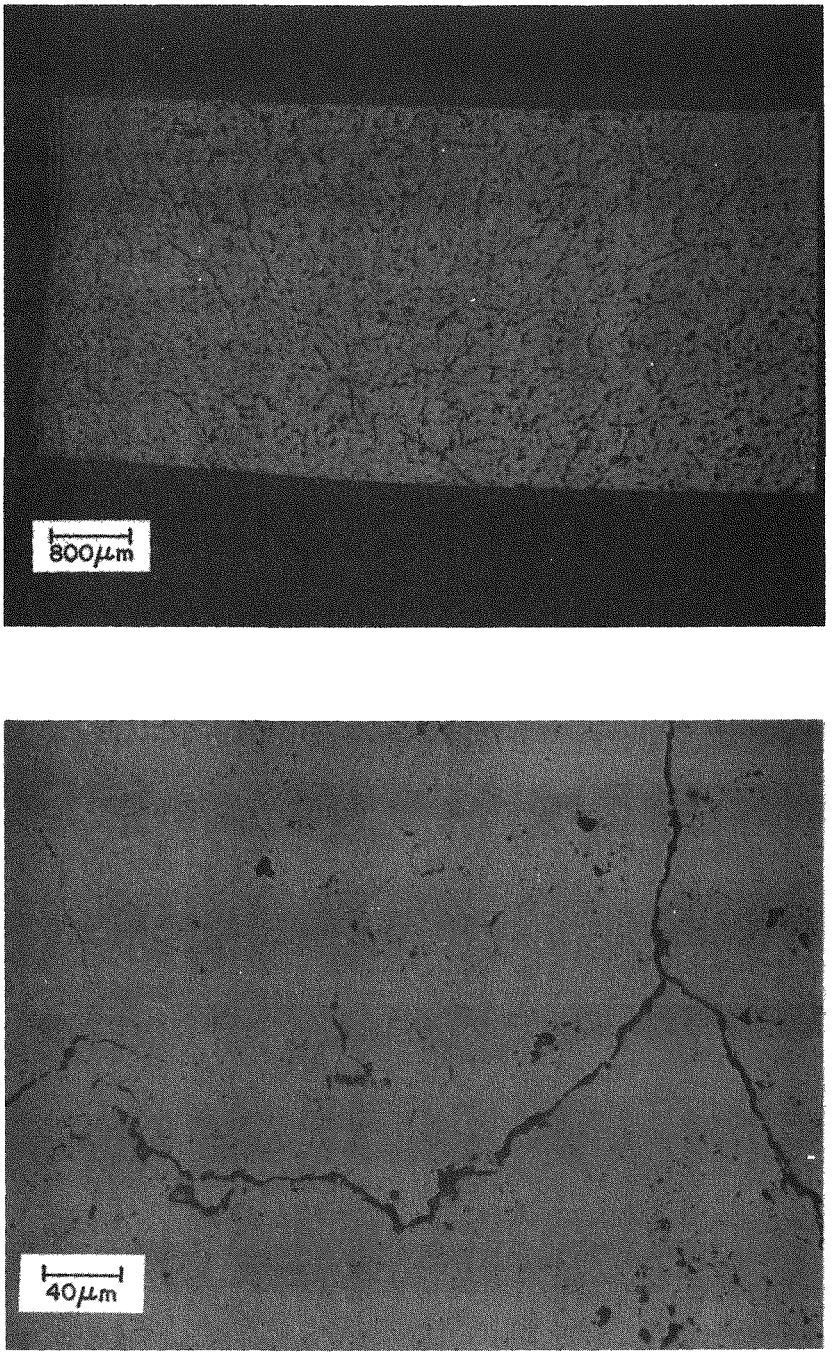
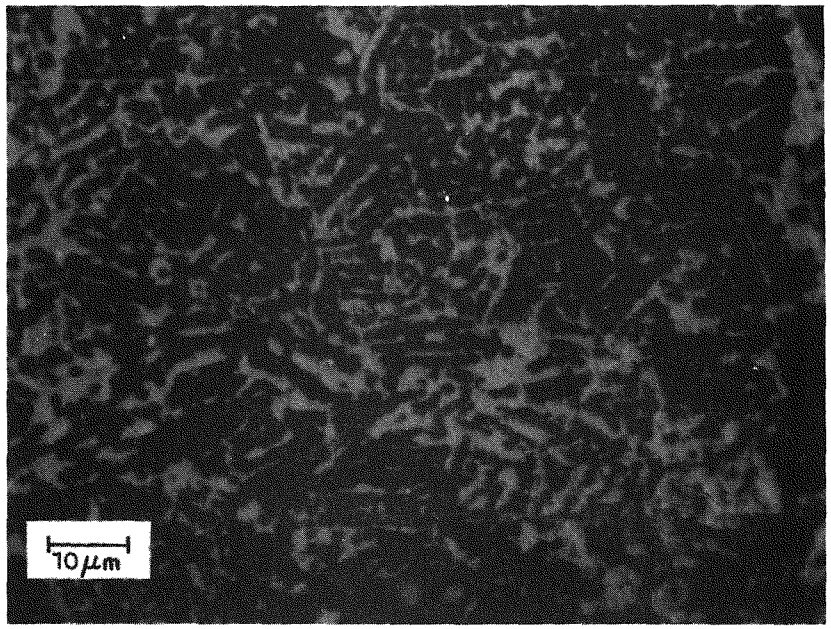
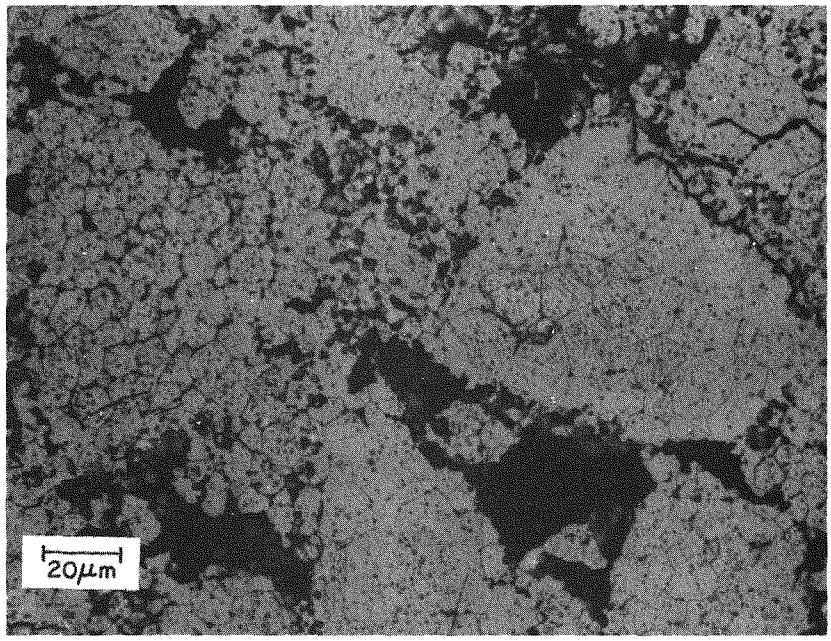


FIGURE 14. Reoxidation-Induced Cracking in 91%-TD Pellet MHP 101. Heat Treated 6 hr at 1000°C and 6 hr at 1350°C (Polished Surface)



a. 275°C, 84% TD



b. 1350°C, 80% TD

FIGURE 15. Microstructure of Heat-Treated Pellet MHP 103 (Acid Etched)

NEW FURNACE RACK FOR SINTERING GPHS SHARDS

A new furnace rack-tray assembly made of ZGS Pt-10% Rh* was designed for the PuFF sintering furnace to provide capability for sintering high-fired (1600°C) shards for the production of GPHS fuel pellets. To estimate the effective operating life of the new racks, creep tests of the alloy will be conducted at the maximum operating temperature (1600°C) for stress levels comparable to and exceeding the maximum design stress.

The existing support racks for the shard sintering furnace and fuel heat treatment furnace in the PuFF facility were designed for operation up to 1440°C. New furnace racks are required that will operate at 1600°C in an oxygen atmosphere for more than 1000 hours. Design of the sintering furnace rack and plans to develop needed creep rate and stress rupture data for the rack alloy, ZGS Pt-10% Rh, are described below. Design of a new rack for the heat treatment furnace was recently initiated and will be described in a subsequent report.

Rack-Tray Assembly Design

The alloy ZGS Pt-10% Rh was selected as the most promising material for the new rack because of its excellent high-temperature properties in oxygen and its availability. Ceramic materials were considered unreliable because ceramics of complex shapes are difficult to fabricate and are susceptible to failure from thermal shock or mechanical impact (the PuFF furnace racks are handled with master-slave manipulators).

The design of the new sintering furnace rack is based on stress rupture data for ZGS Pt-10% Rh at 1400°C extrapolated to 1600°C (Figure 16).[4] Neither creep rate nor stress rupture data are available above 1400°C for ZGS Pt-10% Rh or other potential alloy candidates.

The new furnace rack design (Figure 17) is a modification of the existing sintering furnace rack (Figure 18) with a factor of ~4 reduction in critical stresses. An analysis of stresses for the existing design, summarized in Figure 16, indicated that the shelf and ring (Figure 19) limit the expected life of the rack assembly. Assuming that the shelves and rings of the existing rack are made of ZGS Pt-10% Rh (they are currently made of ZGS platinum, which has lower strength and creep resistance),

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* ZGS (Zirconia Grain Stabilized) platinum-10% rhodium is a high-temperature, oxygen-resistant alloy made by Matthey Bishop, Inc., of Malvern, Pennsylvania.

conservative calculations indicate these components would fail after about 40 and 250 hours of operation, respectively. However, by doubling the total height of the shelves and rings and by using the stronger ZGS Pt-10% Rh alloy, the time to rupture increases to over 1000 hours (Figure 16). All other stresses in the rack-tray assembly would be less than the shelf and ring stresses.

PuFF personnel have noticed that the tray and shelves tend to weld together at 1300°C. This should be an even greater problem at 1600°C. To prevent this self-welding, a flat ceramic disk of Al_2O_3 will be placed between the trays and the shelves. Because of the increase in the shelf and ring height and the addition of the ceramic disks, the total number of trays has been decreased from 6 to 5, and the total height of the tray has been decreased from 3/4 to 5/8 inch.

Creep Tests

Creep tests are planned on the furnace rack to 1) determine if the extrapolated stress rupture data used to set the maximum design stresses are acceptable and 2) to estimate the deformation in high-stress components as a function of time. The results will be used to predict the effective operating life of the furnace racks.

The initial creep test, scheduled to begin in May, will expose a ZGS Pt-10% Rh sample to maximum load (0.25 kg/mm^2) and temperature (1600°C) conditions for the sintering furnace rack. If the results of this initial test indicate a rack life of less than 1000 hours, the rack design will have to be modified to reduce stresses further. Final design drawings of the furnace racks for both the shard-sintering and fuel-heat-treatment furnaces will be submitted for a project to procure the racks after completion of the initial creep test. Additional tests will be made at 1500°C and 1600°C to establish relationships between creep rate and time-to-rupture versus stress.

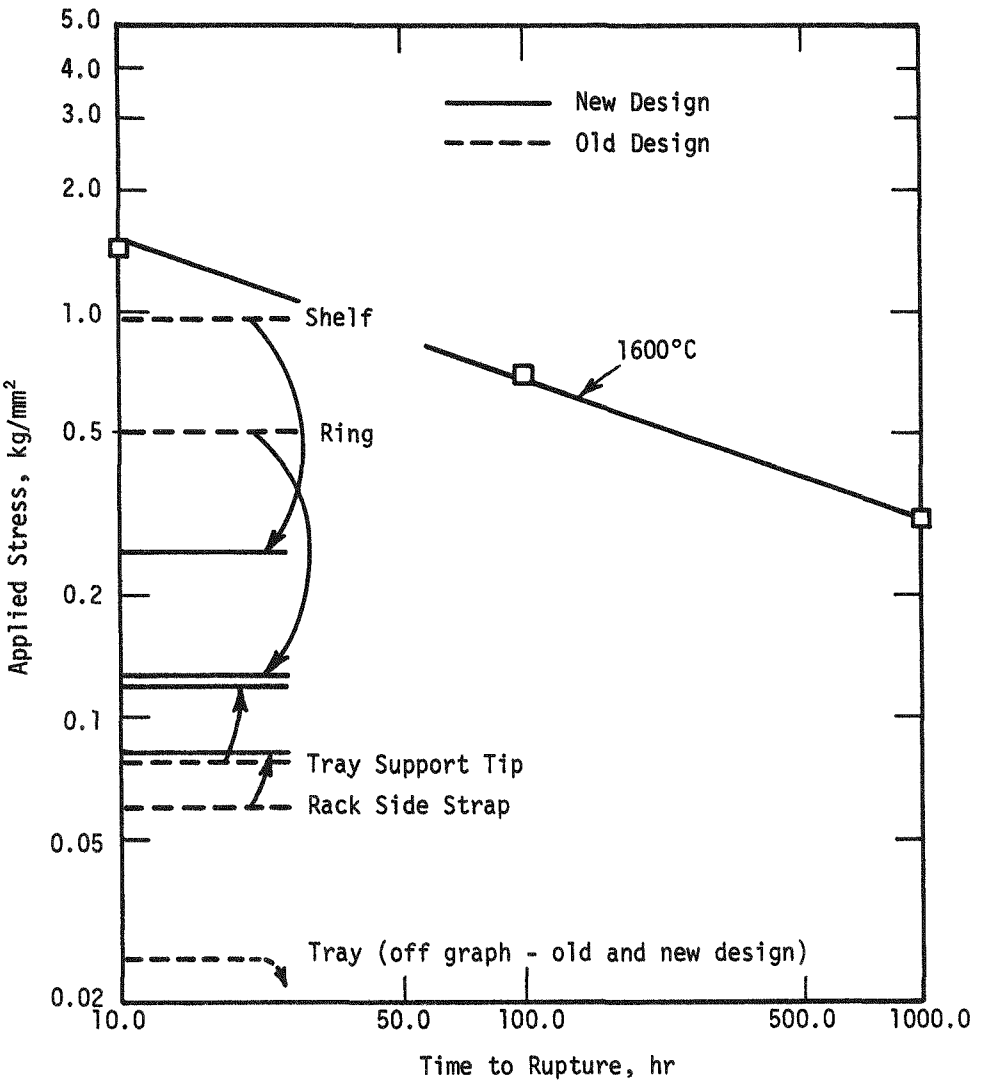


FIGURE 16. Old versus New Design Stresses and Stress Rupture Information for ZGS Pt-10% Rh

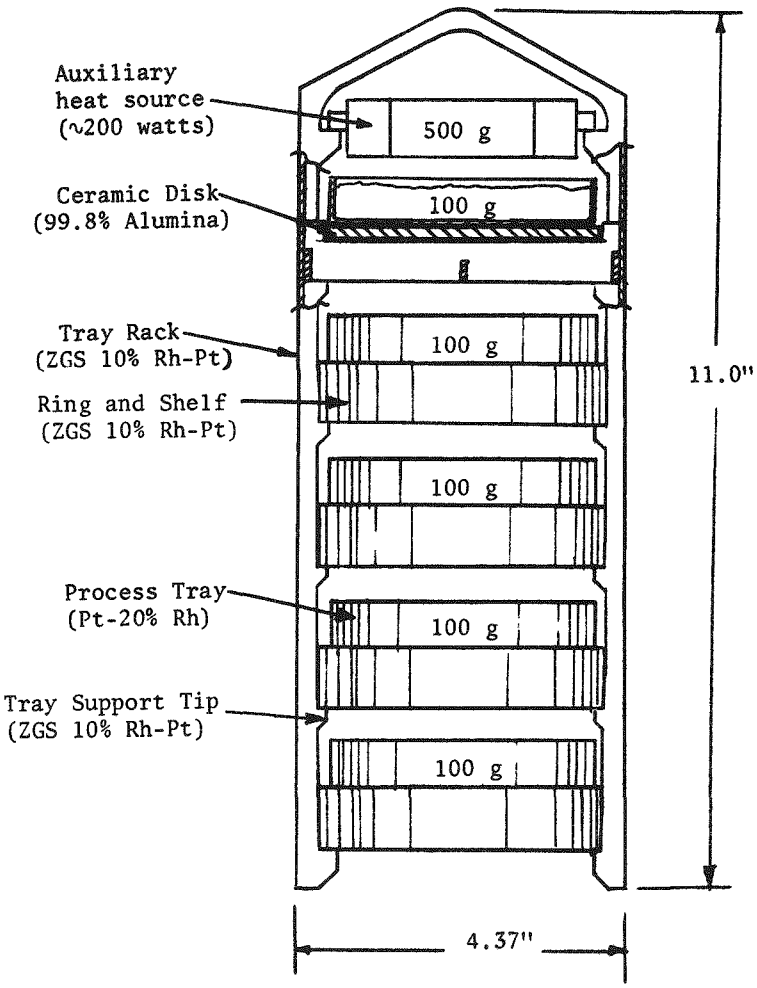


FIGURE 17. Redesigned Furnace Rack and Tray Assembly for PuFF Sintering Furnace

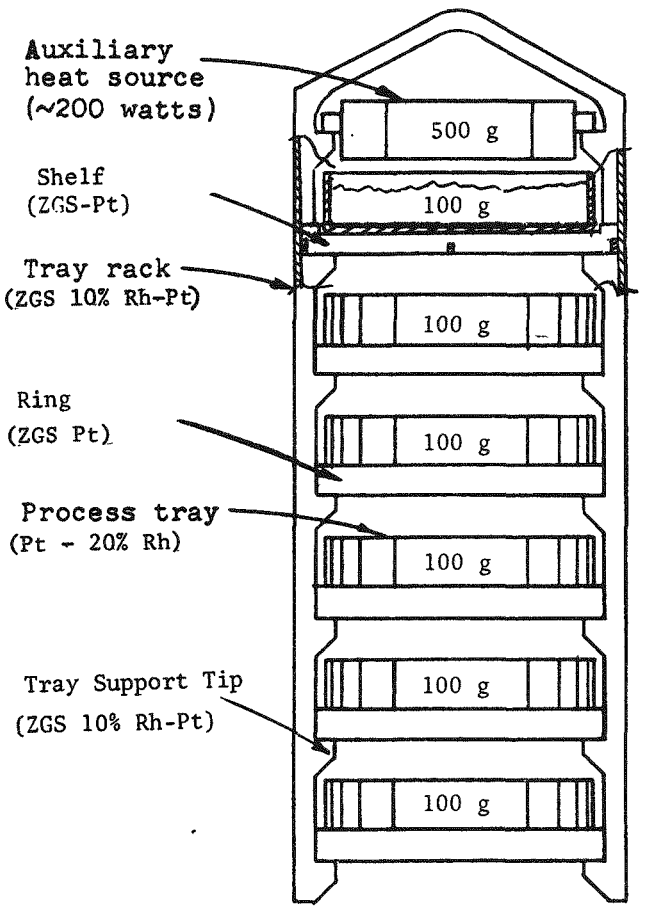


FIGURE 18. Existing Furnace Rack and Tray Assembly for PuFF Sintering Furnace

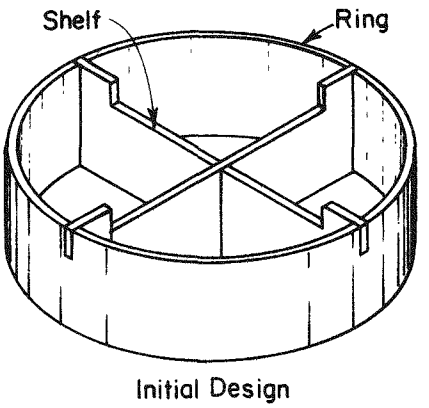


FIGURE 19. Tray Support Rings for PuFF Sintering Furnaces Rack

PROCESS DEVELOPMENT

HOT PRESSING PuO_2 IN ALUMINA DIE LINERS

The first unreduced, hot-pressed pellets of PuO_2 have been successfully fabricated using alumina die liner and punches in a graphite die-susceptor. The pellets were uncracked after a final heat treatment in oxygen. The alumina provides a noninteractive barrier between the PuO_2 and graphite, thereby preventing reduction of the PuO_2 and subsequent reoxidation and reoxidation-induced fracture.

This experiment is part of a continuing effort by SRL to develop new and improved processing techniques for $^{238}PuO_2$ fuel forms. Fracture problems with MHW fuel spheres currently made in the PuFF facility are, in part, associated with the carbothermic reduction of PuO_2 during hot pressing.[5] Phase (and volume) changes during the subsequent reoxidation heat treatment create tensile stresses of sufficient magnitude to cause cracks in the PuO_2 structure. These cracks weaken the microstructure and make the fuel susceptible to fracture from thermal stresses. The use of alumina die liners and punches is being investigated to eliminate the contaminating and reducing atmosphere of graphite hot press dies. If successful, noninteractive dies are expected to reduce fracture, increase production yield, and possibly improve the mechanical performance of $^{238}PuO_2$ fuel forms.

Background

Exploratory hot pressing experiments by Spriggs et al.[6] have demonstrated that dense (94 to 100% TD) polycrystalline ceramic oxides (including many rare earth oxides) can be fabricated using alumina dies at relatively low temperatures (800°C to 1180°C) and high pressures (10,000 to 20,000 psi). Lower temperatures are required for strength considerations (strength of alumina falls off markedly with temperature above about 1200°C) and to prevent reaction between PuO_2 and Al_2O_3. The reaction between PuO_2 and Al_2O_3

$$2\ PuO_2 + Al_2O_3 \xrightarrow{1400°C} 2\ PuAlO_3 + \tfrac{1}{2}O_2$$

occurs only under strongly reducing conditions.[7] According to the phase diagram for the binary system PuO_2 - Al_2O_3, the eutectic

point at an oxygen partial pressure of 1 atm is located at 42 mol % PuO_2 and 1900°C. The mutual solubility of the oxides is less than 1%. Therefore, little, if any, reaction is expected between PuO_2 and Al_2O_3 during hot pressing at temperatures below 1400°C.

Experimental Procedure

Two small PuO_2 pellets (~2.3 g each) were successfully fabricated using Al_2O_3 die inserts. The die assembly consisted of an alumina tube (3/8 in. OD x 1/4 in. ID x 2 in. long) inserted through a 3/8 in. ID graphite die body and alumina rods (1/4 in. OD) for punches (Figure 20).

For the first pellet (MHP 106), a grog mixture of $^{238}PuO_2$ shards (60% presintered at 1100°C and 40% presintered at 1600°C) prepared according to the GPHS flowsheet (Figure 1) was used. These shards were hot pressed at 1200°C and ~10,000 psi for 1 hour using the die assembly described above and the pressing schedule illustrated in Figure 21. The alumina-graphite die assembly was reused to directly hot press a second pellet (MHP 107) from $^{238}PuO_2$ powder obtained from Pu(IV) reverse-strike oxalate (direct fabrication process). A hot pressing schedule similar to the one of Figure 21 was used; however, the maximum temperature and pressure were increased to 1275°C and 17,000 psi, respectively, and were maintained for 15 min.

After their bulk densities were determined, the pellets were sectioned diametrically. One section was retained for as-pressed metallography, while the other section was heat treated and then examined. Heat treatment conditions were 6 hours at 1000°C followed by 6 hours at 1350°C (original GPHS flowsheet)[1] in static air.

Characterization of Fuel

The measured bulk densities of the GPHS-type pellet and the "direct fabrication" pellet were ~75% TD and ~90% TD respectively, which brackets the expected density of GPHS fuel (84-85% TD).

Microstructural analysis of an as-pressed section of the pellet prepared from GPHS shards (MHP 106) showed that much of the shard structure and intershard porosity were retained (Figure 22) despite the relatively high hot-pressing pressure (10,000 psi compared to ~2800 psi for GPHS fuel). An acid etch, which preferentially etches the suboxide phase, indicated that no reduced phases were present in the as-pressed microstructures (Figure 23). Aligned pores imaging the suboxide phase (remnant structure) were not observed in the heat-treated specimen

(Figure 24), again indicating that the $^{238}PuO_2$ was not reduced during hot pressing. No cracks were observed in either the as-pressed or heat-treated sections of the GPHS pellet. The microstructure of the heat-treated and acid-etched section contained grains of two size distributions (Figure 25). The grain size in shards presintered at 1600°C ranged from 12 to 30 μm. The grain size within shards presintered at 1100°C was less than 10 μm. The relatively small grain size within the low-fired shards suggests that a higher shard presintering temperature or higher heat treatment temperature may be necessary to increase grain size and minimize fines generated on impact. (In the current GPHS flowsheet, Figure 1, the final heat treatment temperature has been increased from 1350°C to 1525°C.)

Microstructural analysis of the "direct fabrication" pellet confirmed that a relatively high density (~90% TD) was achieved and revealed that the pellet was uncracked in the as-pressed condition. Suboxide etching of the as-pressed section showed that the PuO_2 was not reduced (Figure 26). In the heat-treated section, no microcracking or macrocracking was observed (Figure 27a) even though the density was ~90% TD. Cracking has commonly been observed at densities of 85-87% TD) and greater in heat-treated PuO_2 hot pressed (and reduced) in graphite.[2,4,7] Severe cracking normally occurs in 90%-TD PuO_2 during heat treatment (Figure 27b).

Conclusions

The above results indicate that:

- Alumina die liners and punches eliminate the carbothermic reduction of PuO_2 during hot pressing.
- Small PuO_2 pellets up to 90% TD can be hot pressed at relatively low temperatures and high pressures in alumina.
- The use of alumina die liners and punches eliminates reoxidation-induced microcracking in PuO_2 even at high density (90% TD).

Program

SRL plans to explore further the use of noninteractive dies, die liners, or coatings to eliminate the fracture problems associated with hot pressing $^{238}PuO_2$ fuel forms in graphite dies. Future small-scale pellet experiments using alumina die liners will be designed to adjust hot pressing parameters to obtain fuel 85% TD, to increase the minimum grain size, and to evaluate aluminum impurities in PuO_2 hot pressed in alumina.

Full-scale testing of alumina die liners to produce 62.5-W fuel pellets for the GPHS is also planned. Hot-press die assemblies with alumina liners and punches have been designed and are being fabricated to produce full-scale GPHS fuel in the PEF. If these initial full-scale tests are successful, a development program will be undertaken to refine hot pressing variables and die design, to analyze fuel microstructures, to verify the absence of excess aluminum in the fuel, and then to demonstrate the process in the PuFF facility for future production of GPHS fuel.

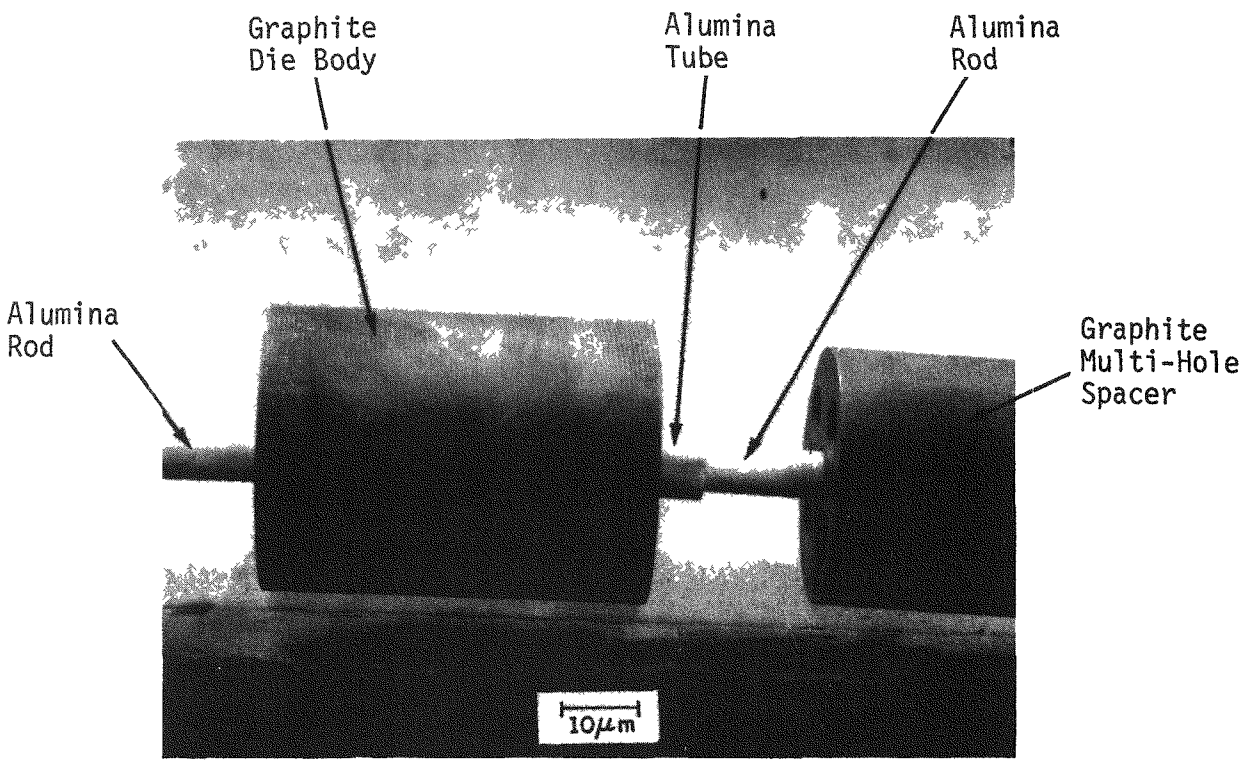


FIGURE 20. Hot Press Die Assembly

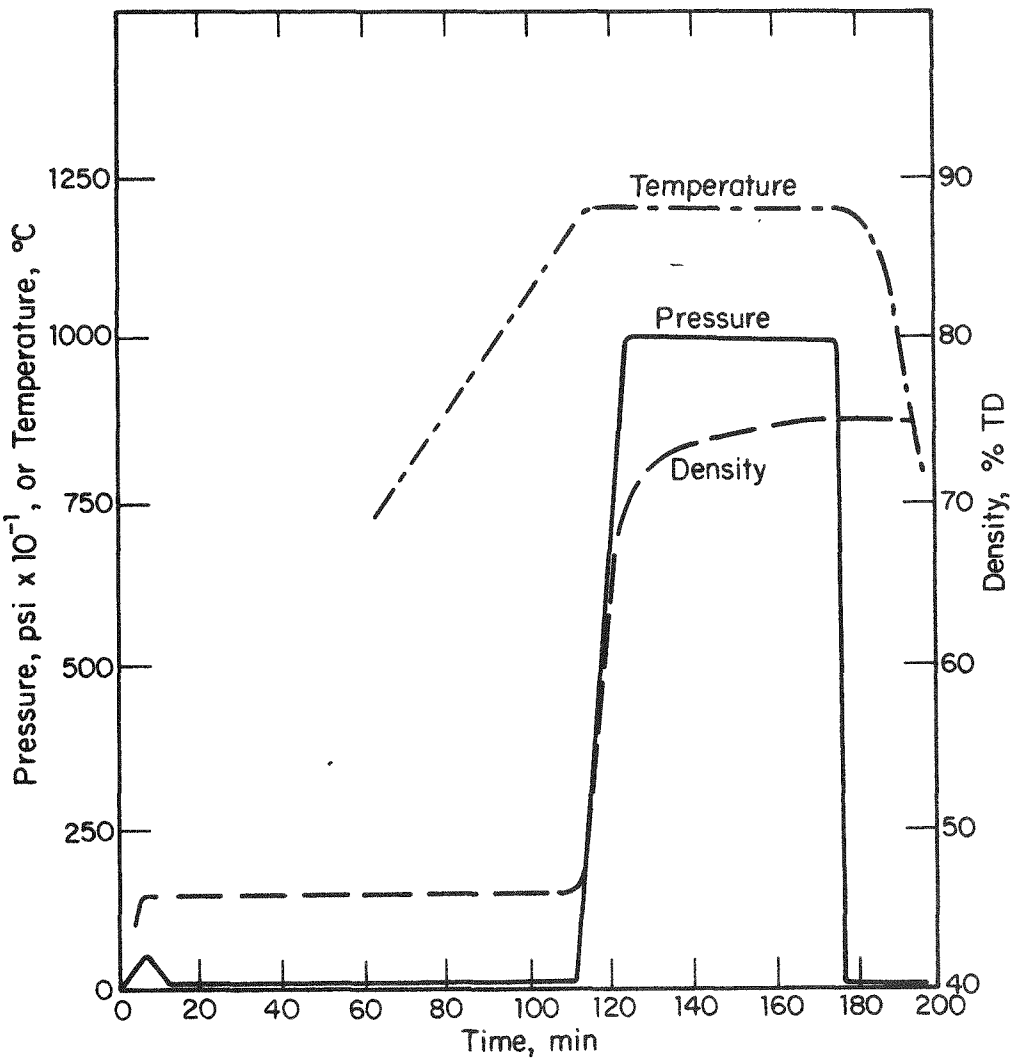


FIGURE 21. Pressing Schedule for GPHS Grog Shards Hot Pressed in Alumina (MHP 106)

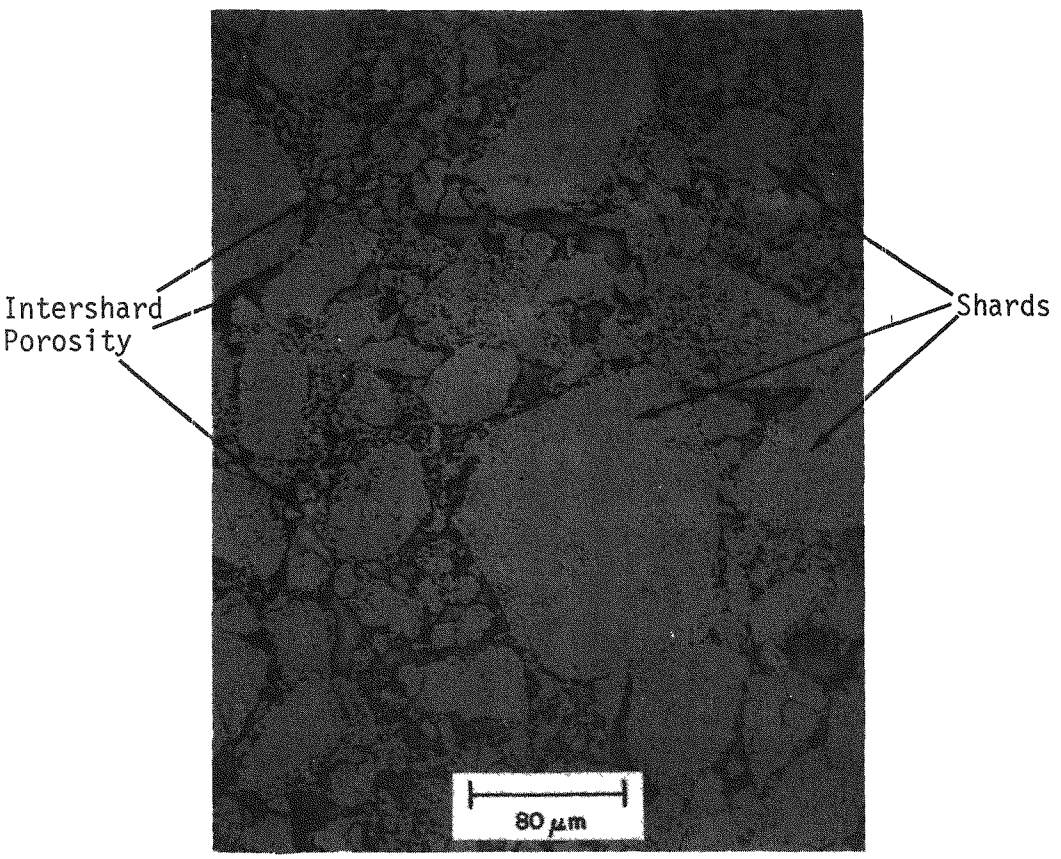
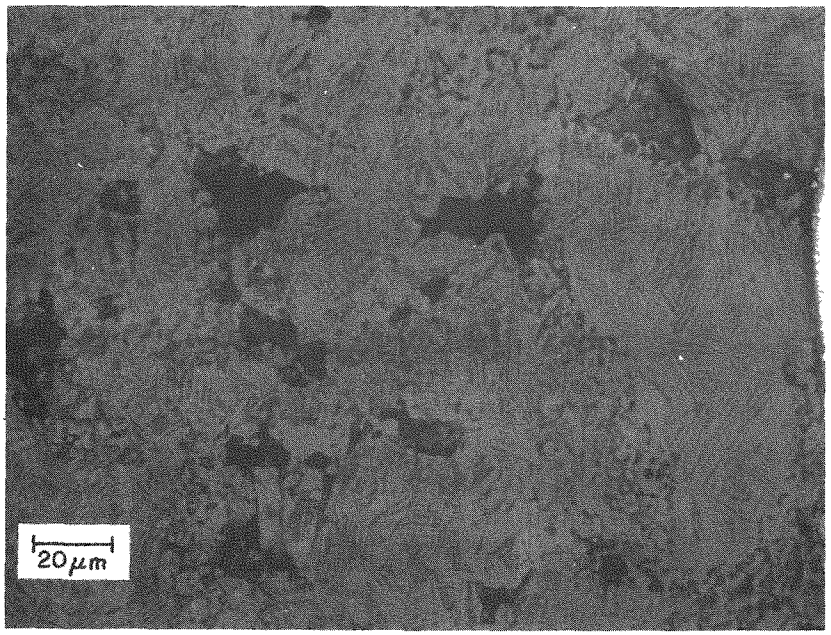
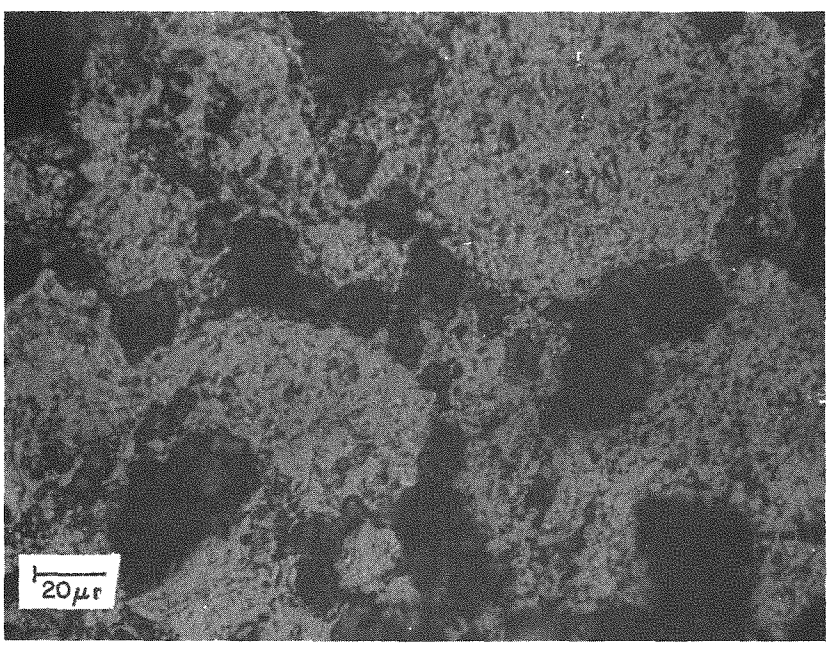


FIGURE 22. Shard Structure and Intershard Porosity of Pellet MHP 106, Hot Pressed in Alumina (Heat Treated, as Polished)



Reduced $^{238}PuO_{2-x}$



Unreduced $^{238}PuO_2$

FIGURE 23. Reduced Microstructure of Pellet Hot Pressed in Graphite and Unreduced Microstructure of Pellet Pressed in Alumina (MHP 106). As Pressed, 1 hr Suboxide Etch.

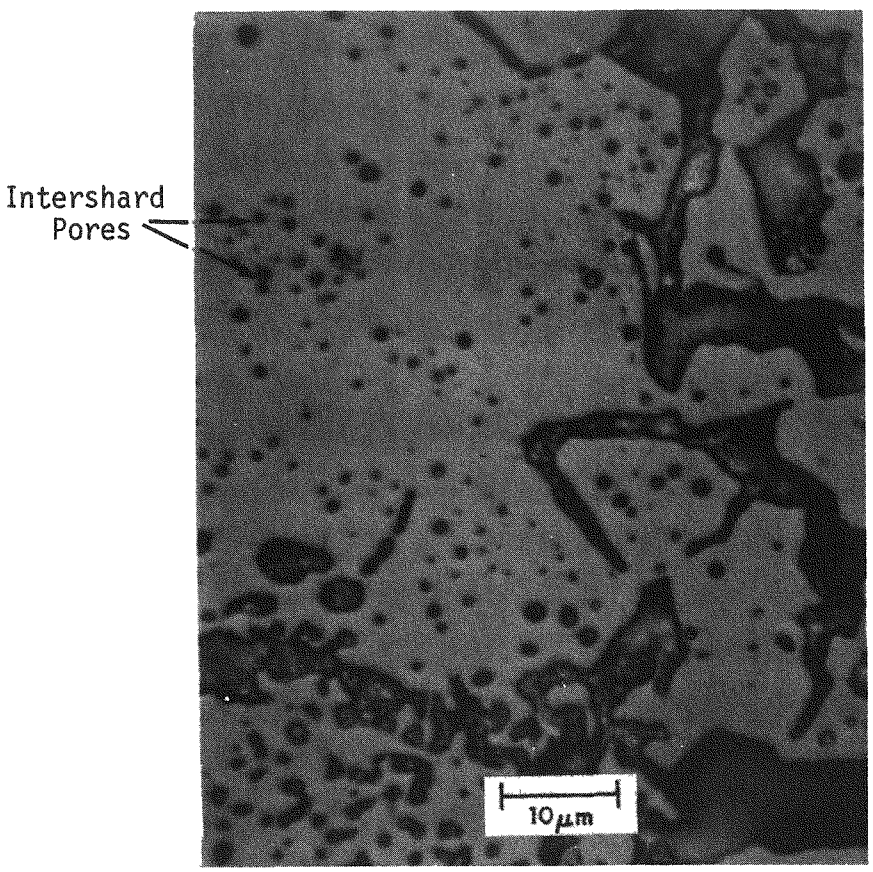


FIGURE 24. Intershard Porosity in $^{238}PuO_2$ Pellet Hot Pressed in Alumina and Heat Treated in Air (MHP 106). Note absence of aligned porosity stringers "ghosting" suboxide phase.

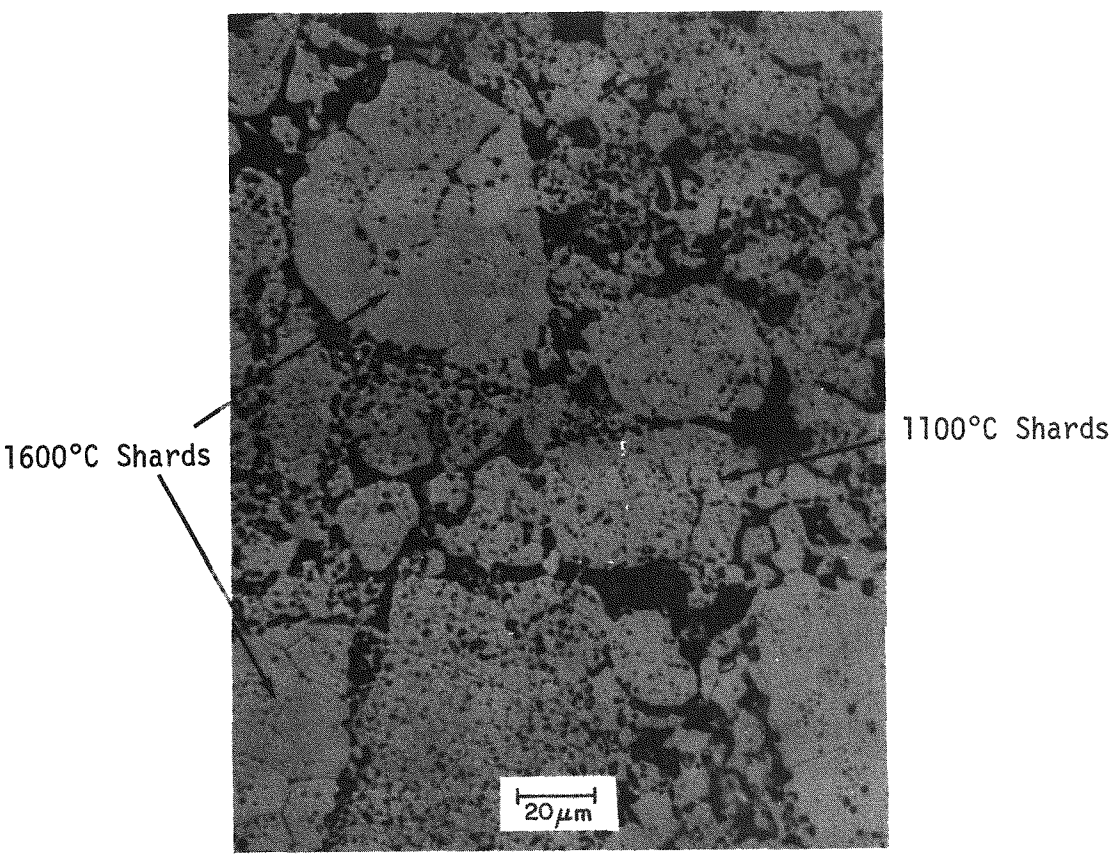
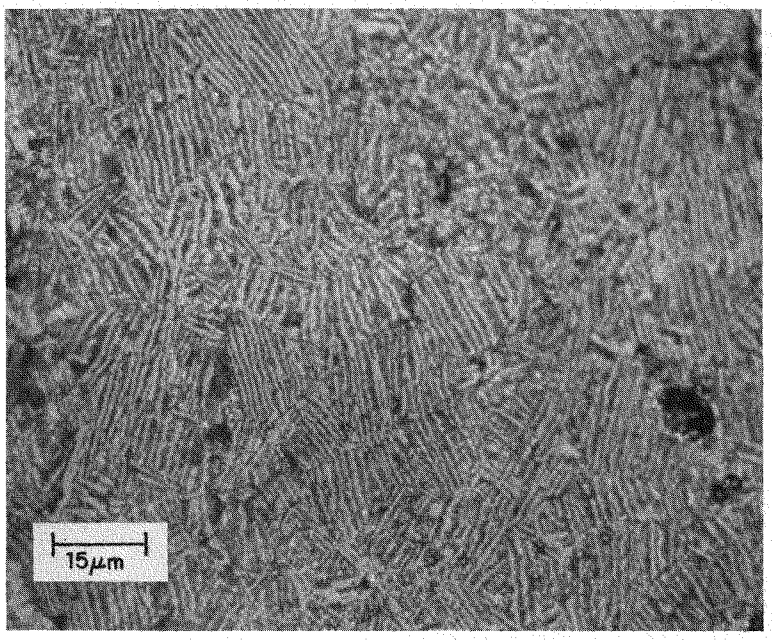
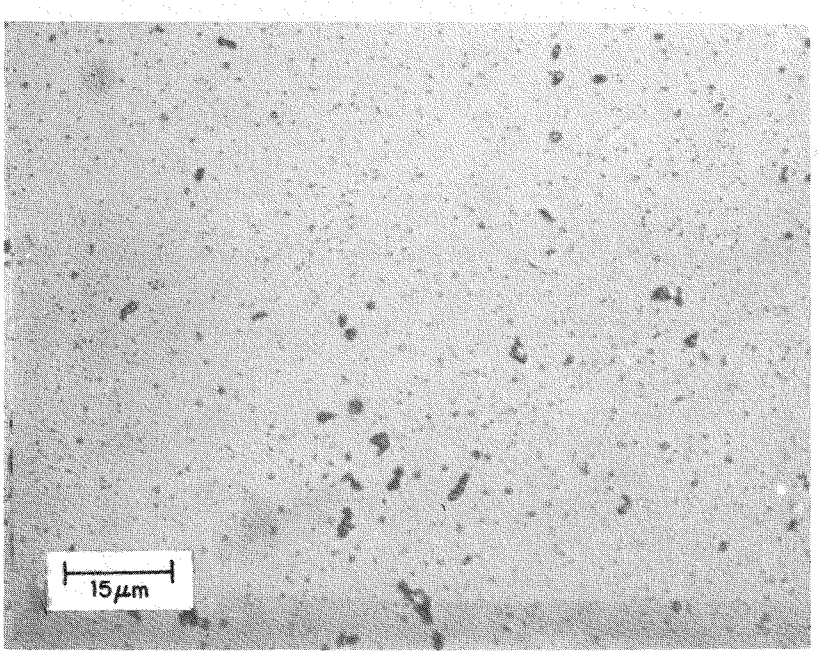


FIGURE 25. Bimodal Grain-Size Distribution in Grog Shards Hot Pressed in Alumina. Heat treated, 1-hr grain-boundary etch.

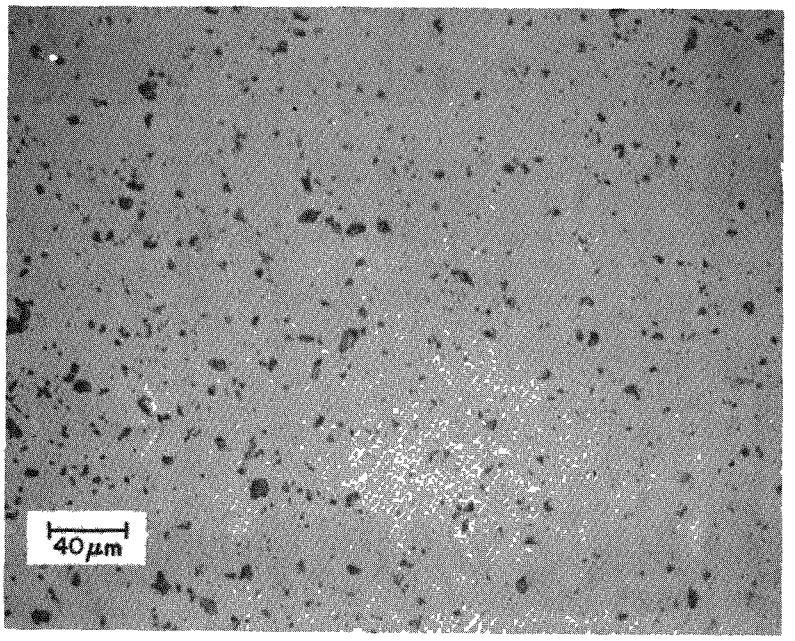


Hot Pressed in Graphite (MHP 101)

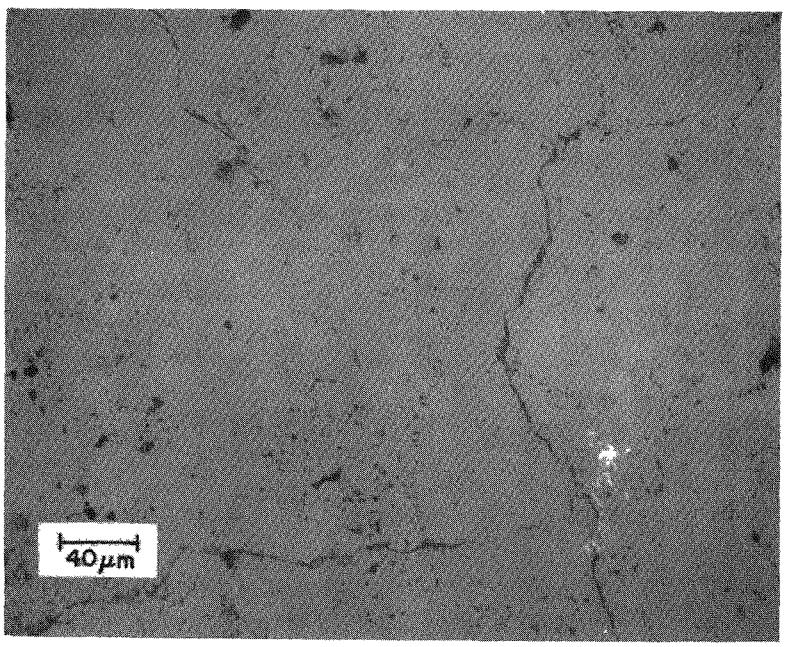


Hot Pressed in Alumina (MHP 107)

FIGURE 26. Alumina Die Inserts Eliminate Suboxide Structure in Direct Hot Pressing of Pu(IV)-Base $^{238}PuO_2$. Suboxide Etch, 90% TD.



a. Hot Pressed in Alumina (MHP 107)



b. Hot Pressed in Graphite (MHP 107)

FIGURE 27. Alumina Die Inserts Eliminate Reoxidation-Induced Fracture. Polished Surface, 90% TD.

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