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^{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 ^{238}Pu Fuel Form Program. This program is supported primarily by the DOE Advanced Nuclear Systems and Projects Division (ANSPD) and also by the Division of Military Applications (DMA).

Goals of the Savannah River Laboratory (SRL) program are to provide technical support for the transfer of ^{238}Pu fuel form fabrication technology from Los Alamos Scientific Laboratory (LASL) to 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, 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 of General-Purpose Heat Source (GPHS) fuel forms continued at the Savannah River Laboratory (SRL) Plutonium Experimental Facility (PEF). GPHS Pellet 8 underwent final heat treatment without incident and was integral and free of surface cracks after heat treatment. GPHS Pellet 9 was hot pressed without incident but was badly thermally shocked during the initial attempt at final heat treatment due to sintering furnace failure.

GPHS Pellets 7 and 8 remained integral after exposure to flowing helium at ambient pellet temperatures of 150-200°C. Prior to the helium exposure, GPHS Pellet 7 survived 10 to 12 thermal cycles between about 800°C and 400°C without fracturing.

Fabrication Conditions

During this reporting period, GPHS Pellet 8 was heat treated and GPHS Pellet 9 was hot pressed. The initial attempt to heat treat GPHS Pellet 9 was interrupted after only a few minutes at 1525°C by a water leak in the heat exchanger of the cooling water system for the sintering furnace. Additional fabrication tests were postponed because of repairs to the heat exchanger and to the sintering furnaces.

Process conditions for fabrication of GPHS Pellets 7, 8, and 9 are summarized in Tables 1 and 2. Essentially the same conditions were used to fabricate all three pellets. GPHS Pellet 7 is included in this table because the fabrication conditions for this pellet are the current SRL centerline conditions. GPHS Pellet 9 was fabricated to demonstrate that SRL centerline conditions produce acceptable fuel with reproducible characteristics. For GPHS Pellet 8, the charge to the die was increased in the first attempt to produce a pellet with a density of 86% of theoretical density (TD); the maximum expected density for GPHS fuel forms.

TABLE 1

Process Conditions Used for Fabricating GPHS Pellets 7, 8, and 9

^{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

TABLE 2

Hot Pressing Conditions for GPHS Pellets

	GPHS Pellet No.		
	7	8	9
Preload, lb	200	200	200
Heating			
Time to 1100°C, min	3	3	3
Max Temp, °C	1530	1530	1530
Time to Max Temp, min	8	8	8
Load			
Temp of Initiation, °C	1360	1360	1300
Max Load, lb	2600	2600	2600
Ramp, min	5	5	5
Time Between Initiation of Heat and Load, min	4	4	4
Time to Die Closure after Max Load, min	2	10	2
Time at Max Load and Temp after Closure, min	5	4	5

Pellet Characteristics

The good pellet quality previously observed in full-scale tests with the reference shard mixture[1] continued in the most recent tests. GPHS Pellet 9 was integral with no surface cracks as pressed. This pellet remained integral, but several hairline surface cracks were observed on one end after equipment problems interrupted the heat treatment operation. The thermal shock associated with rapid cooling from 1525°C to 200°C in about 1-1/2 hr probably was the major contributor to these cracks. Normal cooling time over this temperature range is 8 to 10 hr (i.e., <200°C/hr).

GPHS Pellet 8 was the first pellet to be free of surface cracks after final heat treatment. All other pellets had one or more surface cracks after final heat treatment. The improved surface quality may be related to the positioning of the pellet during final heat treatment. GPHS Pellet 8 was placed in a bed of ThO_2 shards with approximately one-half of the pellet buried. All other pellets were heat treated on alumina boats. A slight color change on the portion of the pellet surface which was in contact with the ThO_2 and slight sticking of ThO_2 shards to the pellet indicated a ThO_2-PuO_2 interaction. Additional tests are necessary to determine whether heat treatment in ThO_2 shards does improve the surface quality.

GPHS Pellet 8 showed excellent dimensional stability during final heat treatment (Table 3) as did previous pellets made from the reference shard mixture. Linear shrinkages on diameter and length were 0.3% and 0.4% (0.003 and 0.004 in.), respectively. GPHS Pellet 9 showed essentially no shrinkage as a result of the interrupted heat treatment. This pellet will be regaged after undergoing the full heat treatment cycle.

The final density of GPHS Pellet 8 was lower than expected, i.e., 84.9% instead of 86% TD. As discussed in Reference 1, the lower density occurred because of additional corrosion of the die material at the PuO_2 - graphite interface. The additional corrosion occurred because of the additional time (10 min versus 2 min) required to obtain die closure with the larger PuO_2 charge to the die. The as-pressed diameter and length of GPHS Pellet 8 were 0.004 to 0.005 in. greater than the original dimensions of the die cavity. On the other hand, the as-pressed diameter and length of GPHS Pellets 7 and 9 were the same as or slightly smaller than the original dimensions of the die cavity (Table 4).

Microstructural characterization of GPHS Pellets 6 and 7 is discussed in a later section of this report.

TABLE 3

GPHS Pellet Data

GPHS Pellet No.	Condition	Diameter, in.	Length, in.	Weight, g	Density, % TD	O/Pu
2*	As-pressed	1.065	1.066	146.683	82.3	1.89
	Heat Treated	1.064**	1.055	147.630	>83.8**	
	Difference	-0.1%	-1.0%	0.947	1.5	
3*	As-pressed	1.072	1.074	145.714	80.4	1.85
	Heat Treated	1.065**	1.066	146.999	>82.4**	
	Difference	-0.7%	-0.7%	1.285	2.0	
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	84.3	1.93
	Heat Treated	1.092	1.093	152.351	84.3	
	Difference	-0.3%	-0.4%	0.644	1.0	
6††	As-pressed	1.107	1.107	152.069	81.1	1.090
	Heat Treated	¶	1.099	152.934¶¶¶	¶	
	Difference		-0.7%			
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%	0.718	1.2	
9†,¶¶	As-pressed	1.093	1.098	151.790	83.5	1.93
	Heat Treated¶¶	1.093	1.097	152.440	83.9	
	Difference	-0.0%	-0.1%	0.650		

* Shard mixture: 60% sintered at 1150°C and 40% sintered at 1450°C; pellet geometry: right circular cylinder.

** Pellet fractured; diameter measured and density calculated from reassembled pieces.

† Reference shard mixture and reference geometry.

†† Remnant shards and reference geometry.

¶ Pellet was sectioned longitudinally prior to final heat treatment.

¶¶ Heat treatment interrupted when furnace reached 1525°C because of equipment problems.

¶¶¶ Final weight calculated from weight gain during heat treatment of 1/2 pellet.

TABLE 4

Pellet Versus Die Cavity Dimensions* For GPHS Pellets

GPHS Pellet No.	Condition	Diameter, in.	Length, in.
7	Die Cavity	1.095	1.102
	As-pressed	1.093	1.099
	Heat Treated	1.089	1.096
8	Die Cavity	1.094	1.107
	As-pressed	1.098	1.112
	Heat Treated	1.095	1.108
9	Die Cavity	1.094	1.097
	As-pressed	1.093	1.098
	Heat Treated		

* Measurement variation is ±0.001 in.

Thermal Cycling

Although GPHS Pellet 7 had several hairline surface cracks after final heat treatment, this pellet remained integral when cycled 10-12 times between about 800°C and 400°C. This thermal cycling of the pellet occurred during successful efforts to photograph the pellet using only the light from the decay heat. However, this thermal cycling may have caused some damage to the pellet since there were slight increases in the pellet diameter (Table 5) and the amount of surface cracking (visually observed).

TABLE 5

Pellet Characteristics After Thermal Cycling And Helium Exposure Tests

GPHS Pellet No.	Date	Condition*	Diameter, In.	Length, in.	Wt, g	Density, % TD
8	7/5	Heat Treated	1.095	1.108	156.30	84.9
	7/5 - 7/19	Storage	1.097	1.109	156.26	84.5
	7/19 - 7/20	Exposure to Flowing Helium	1.096	1.109	156.25	84.6
7	6/26	Heat Treated	1.089	1.096	153.47	85.2
	7/16	Thermal Shock	1.092	1.096	153.5	84.7
	7/16 - 7/17	Exposure to Flowing Helium	1.095	1.099	153.495	84.0

* Pellet was gaged after it was subjected to condition described in this column.

Exposure to Flowing Helium

GPHS Pellet 8 remained free of surface cracks and its physical characteristics were unchanged (Table 5) after 2-1/2 weeks of storage in a graphite container followed by overnight exposure to flowing helium at 150 to 200°C. GPHS Pellet 7 also survived a similar helium exposure. However, slight increases in both the diameter and length of GPHS Pellet 7 (Table 5) indicated that the pellet may have sustained additional damage (cracking) that was caused either by exposure to helium or by the additional handling associated with pellet gaging. (GPHS Pellet 7 had undergone thermal cycling prior to being exposed to helium.)

These experiments in flowing helium demonstrated that GPHS fuel, after storage, should be strong enough to survive the thermal shock in PuFF associated with the transfer of fuel from the argon atmosphere of fuel fabrication cells to the helium atmosphere of the welding cell. Multi-Hundred Watt (MHW) fuel occasionally cleaves during this transfer.

The resistance to fracture demonstrated by GPHS Pellets 7 and 8 during thermal cycling and exposure to helium indicates the ruggedness of GPHS fuel fabricated in PEF.

MICROSTRUCTURAL ANALYSIS OF GPHS PELLETS 6 AND 7

Microstructural analysis of GPHS Pellets 6 and 7 indicated that (1) as-pressed pellets reoxidize at self-heat temperatures during storage, (2) this low-temperature reoxidation causes microcracking, (3) density gradients are more severe in pellets pressed from MHW feed that in pellets pressed from GPHS grog feed, and (4) a GPHS pellet subjected to thermal cycling and exposure to flowing helium can survive these thermal stresses without microcracking. Additional experiments are planned to evaluate the effects of low-temperature reoxidation and to measure the thermal gradients within GPHS pellets.

This study is part of a continuing microstructural examination of GPHS fuel forms being fabricated in PEF. Microstructural analysis of GPHS pellets has previously confirmed that centerline GPHS conditions, developed by Los Alamos Scientific Laboratory [J. W. Congdon, "Trip Report, GPHS Technology Transfer Meeting, LASL, April 17-20, 1979." Memorandum to M. L. Hyder (May 29, 1979)] and modified for use in the PEF, produce a homogeneous microstructure and a relatively uniform density distribution. The primary goal of

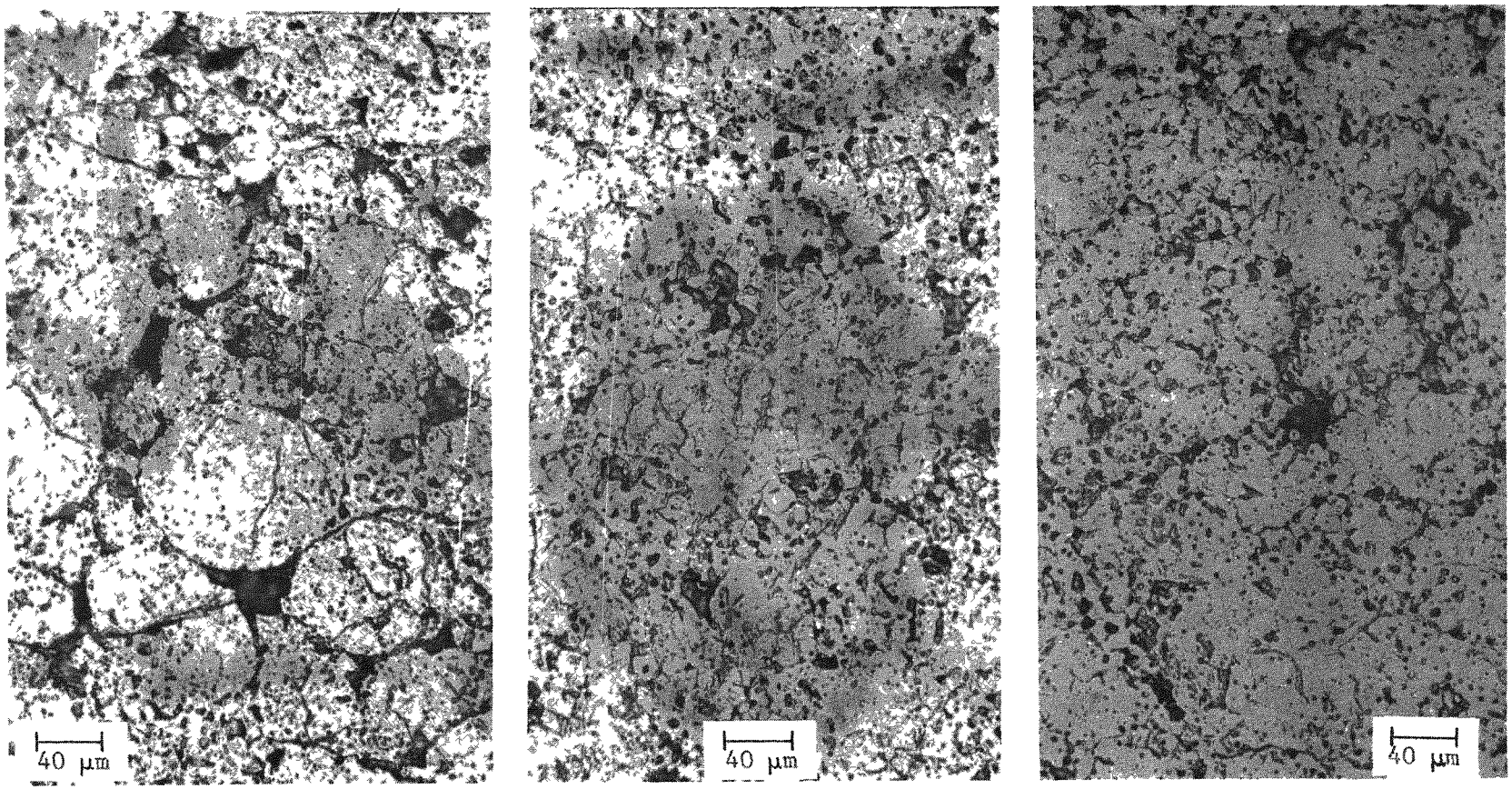
the microstructural analysis of GPHS pellets is to determine the effects of process variations on their microstructure and fracture tendency in order to help establish centerline process conditions and provide a basis for GPHS fuel form technical standards.

GPHS Pellet 6

GPHS Pellet 6 was hot pressed from a shard mixture of 86% 1300°C shards and 14% 1450°C shards (similar to MHW feed). The fabrication conditions and physical characteristics of GPHS Pellet 6 are reported elsewhere.[1] GPHS Pellet 6 was stored at self-heat temperature for five days and was then cut in half longitudinally. One half was stored as pressed in a graphite container at self-heating temperatures (about 200°C) for an additional 5 weeks. The other half was given a final heat treatment at 1525°C for 6 hr. The bulk density of this pellet as-pressed was 81.1% TD.

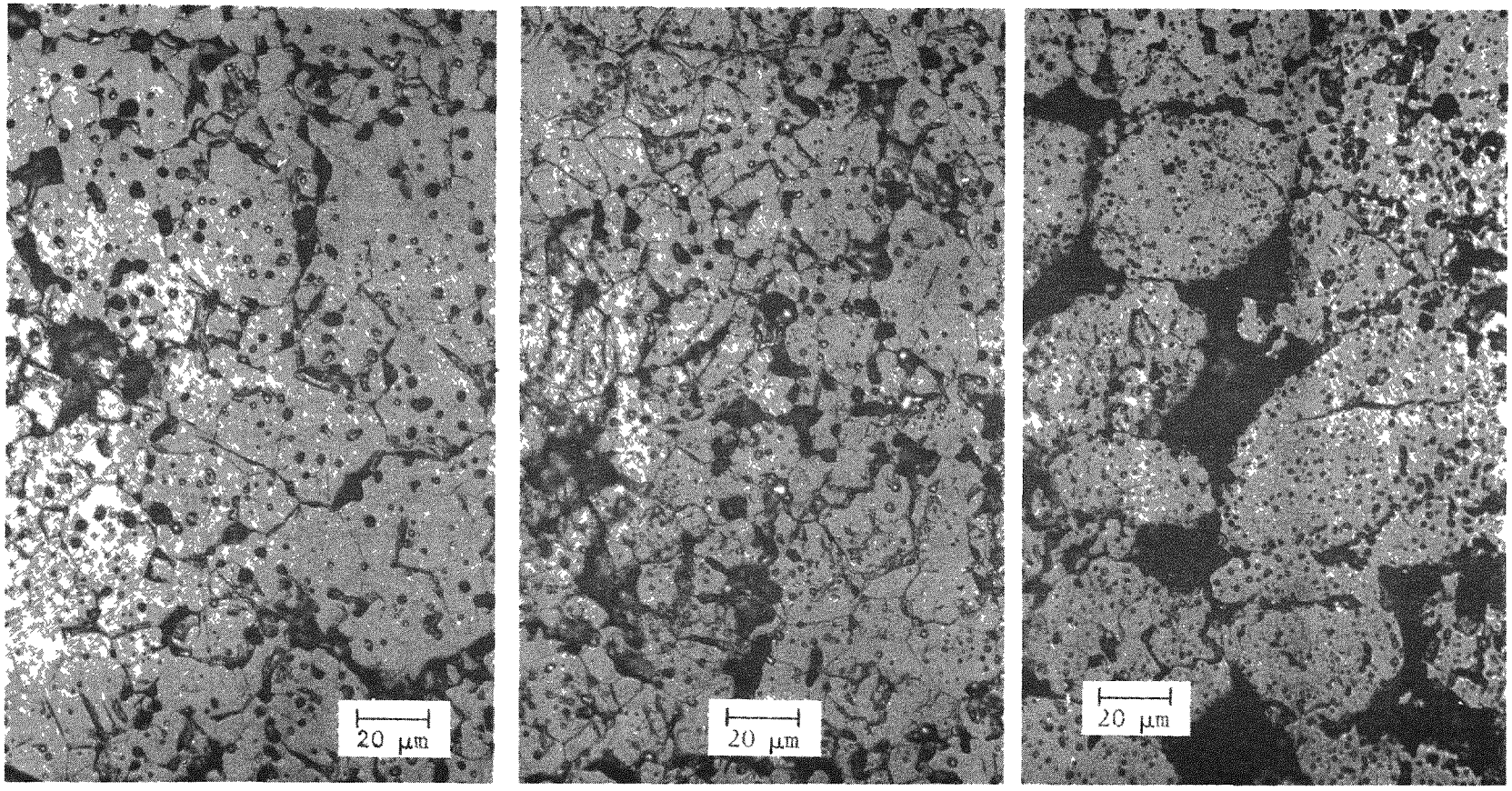
Metallographic characterization of a section of the aged, as-pressed half of GPHS Pellet 6 showed that it was microcracked after storage at the ambient self-heat temperature for about 6 weeks (Figure 1). Microcracking was less severe near the center of the pellet despite the higher density. Suboxide etching indicated that no suboxide phase was present and, therefore, that the pellet had reoxidized during storage in argon with an O_2 content of 100 to 500 (Figure 2). This low-temperature reoxidation is apparently detrimental to the microstructure of the pellet and suggests that the storage time of as-pressed pellets prior to heat treatment should be limited.

Metallographic characterization of the half of GPHS Pellet 6 which was heat treated showed that it had a bulk density of approximately 83% TD. The density variations ($\pm$3% TD) observed in this pellet (Figures 3 and 4) are slightly higher than observed in earlier GPHS pellets. This observation suggests that the pressing characteristics of the GPHS grog feed (40% 1600°C shards and 60% 1100°C shards) may be superior to the presing characteristics of the shard mixture used for GPHS Pellet 6 (86% 1300°C shards and 14% 1450°C shards). The shards used for GPHS Pellet 6 were similar to the feed (100% 1300°C shards) presently used for MHW fuel forms. The surface tensile cracks in GPHS Pellet 6 were also larger and more frequent than typically observed in other GPHS pellets. Cracks were also observed in the interior of the pellet. The larger density gradients in GPHS Pellet 6 apparently resulted in higher stresses throughout the pellet. These results indicate that the MHW fuel form could be improved by using a grog feed material as used for the GPHS fuel form.



End Interior Center

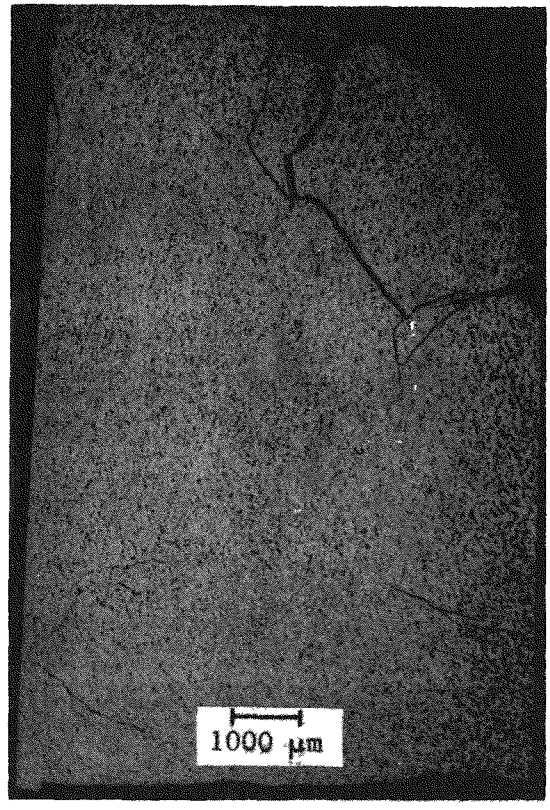
**FIGURE 1. Longitudinal Axis of GPHS Pellet 6.
As pressed, stored six weeks, polished.**



End Interior Center

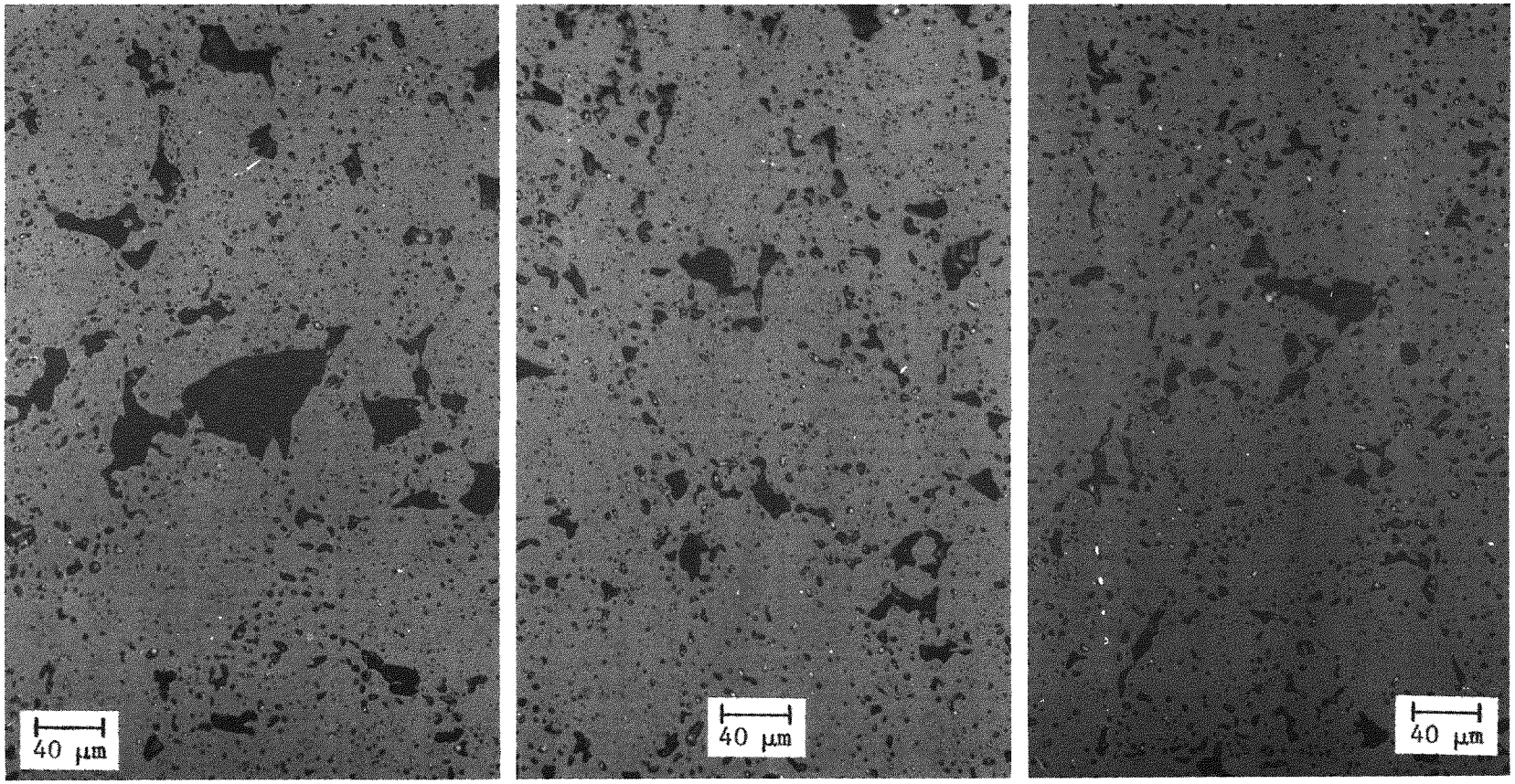
**Figure 2. Longitudinal Axis of GPHS Pellet 6.
As pressed, stored six weeks, 15-min suboxide etch.**

End



Center

FIGURE 3. Longitudinal Section of GPHS Pellet 6. Heat treated, polished.



End Interior Center

FIGURE 4. Longitudinal Axis of GPHS Pellet 6. Heat treated, polished.

GPHS Pellet 7

GPHS Pellet 7 was hot pressed using centerline process conditions.[1] After heat treatment the pellet was subjected to thermal stresses from repeated (10 to 12 cycles) thermal cycling between about 800°C and 400°C over twenty-minute intervals. GPHS Pellet 7 was also exposed to thermal stress when it was exposed to flowing helium for 12 hr at ambient pellet temperatures of 150-200°C. A longitudinal section of the heat-treated pellet was prepared for metallography after helium exposure.

The bulk density of GPHS Pellet 7 was 82.5% TD. As shown in Figure 5, the density distribution of this pellet was relatively uniform (±2% TD). Only one crack was observed in the cross-section of the pellet, although this crack did extend about 0.3 inches into the interior (Figure 6). The thermal stresses to which the pellet was exposed did not initiate additional surface crack or microcracking in the cross-section which was examined.

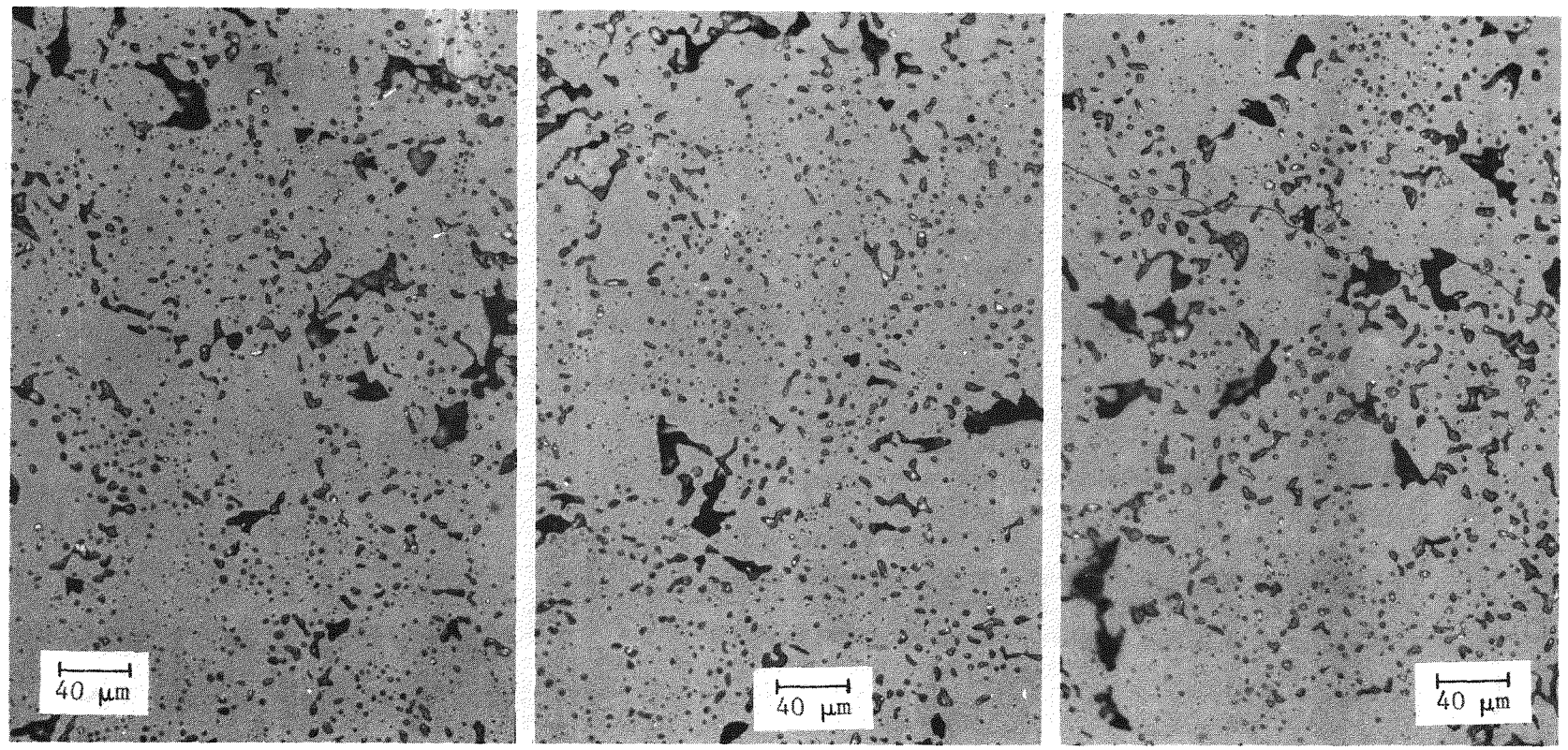
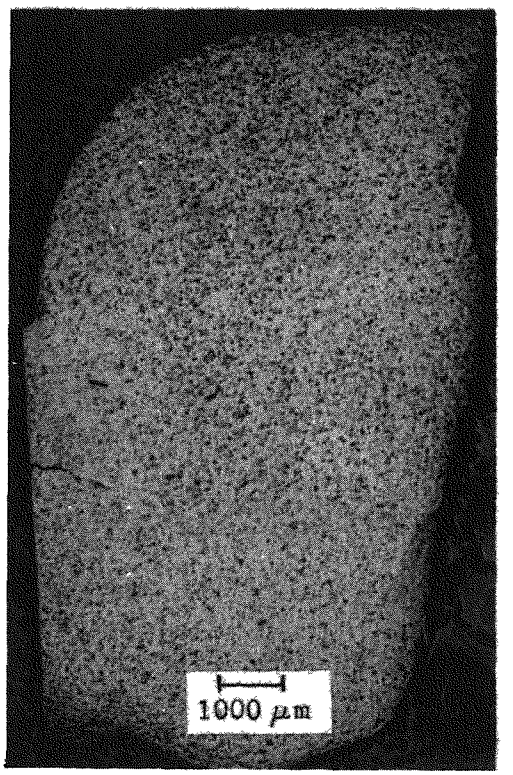


FIGURE 5. Longitudinal Axis of GPHS Pellet 7. Heat treated, polished.

End



Center

FIGURE 6. Longitudinal Section of GPHS Pellet 7. Heat treated, polished.

MULT-HUNDRED WATT PROCESS SUPPORT

CERAMIC FURNACE HEARTH FABRICATION

Ceramic furnace hearths for use in the vertical process furnaces of the Plutonium Fuel Form (PuFF) Facility were successfully fabricated at SRL.

Production of Multi-Hundred Watt (MHW) $^{238}PuO_2$ fuel forms is currently in progress in the Plutonium Fuel Fabrication (PuFF) facility. Recently, only two "ceramic furnace hearths" were available for the three vertical process furnaces and replacement hearths could not expeditiously be obtained from an outside vendor. SRL was therefore requested to cast, cure, dry, and sinter ceramic furnace hearths.

Two hearths of the standard design, and two multi-piece hearths, fired to 1300°C, were fabricated for SRP. Several castings were also made in the development stage of this program but were discarded due to mold release problems and differential expansion cracks in the castings and in the high-fired Al_2O_3 tubes.

Fabrication Procedure

Mold

1. The aluminum mold shown in Figure 7 was designed by Separations Technology and fabricated by the Maintenance Department. Both sections of the mold must be disassembled (Figure 8) to remove the casting.

2. Wood dowels were used as inserts where holes were required for thermocouples (Figure 9). These dowels burned out during sintering of the casting. Burnout was assisted by drilling holes longitudinally through the center of each dowel.

Mold Preparation

1. To prevent the casting from adhering to the mold, the interior surfaces of aluminum were coated with **Vaseline®** (Chesebrough-Ponds, Inc., Greenwich, Connecticut) and then the surfaces were lined with polyethylene. The **Vaseline®** helped to hold the polythylene in place and also lubricated corners which were not adequately covered by the plastic.

2. The wood dowels were coated with paraffin to prevent water adsorption from the casting which could cause drying cracks.

3. Differential thermal expansions between the stainless steel, castable, and high-fired Al_2O_3 were shown to cause cracking of the castable and the Al_2O_3 during the heat treatment. Therefore, all of the interfaces were coated with paraffin to provide a gap during sintering and to eliminate stresses from differential expansion. Paraffin was also used to prevent infiltration of the castable into gaps at interfaces, especially between the stainless and high-fire Al_2O_3 pieces. These gaps were required to prevent stresses in the Al_2O_3 caused by expansion of the steel.

Casting

1. Alundum[3] Insulating Castable CA 333 (3000 g) was blended by hand with 500 ml of water.

2. The castable was then poured into the mold as the mold was vibrated by hand. The vibrations increased the fluidity of the castable and helped to remove entrapped air.

Curing

1. The entire mold assembly and casting were wrapped in polyethylene and the casting was allowed to cure for 24 hours at room temperature.

Drying

1. After curing, the plastic covering was completely disassembled to remove the casting.

2. The casting was dried in air for 24 hours at room temperature (about 24°C).

3. Stainless steel foil was used to cover the threads in the steel insert and prevent oxidation of the steel threads during sintering.

4. The casting was heated in static air according to the following schedule:

   - Heat at 15°C/hr to 80°C
   - Hold 6 hr at 80°C
   - Heat at 40°C/hr to 675°C
   - Hold 4 hr at 675°C
   - Cool at <200°C/hr to 25°C

A photograph of a complete hearth is shown in Figure 10.

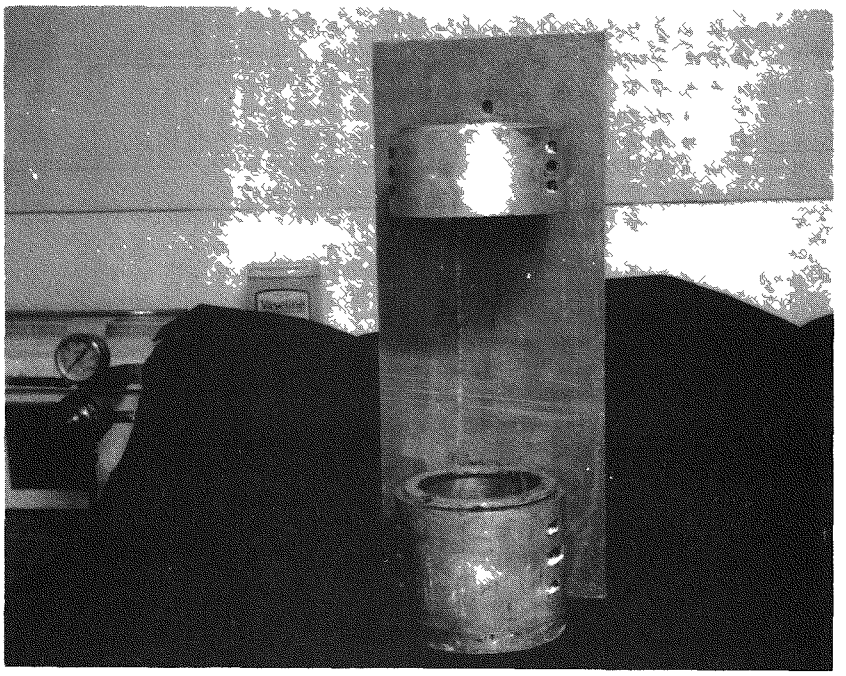


FIGURE 7. Aluminum Mold for Ceramic Hearth Castings (Assembled)

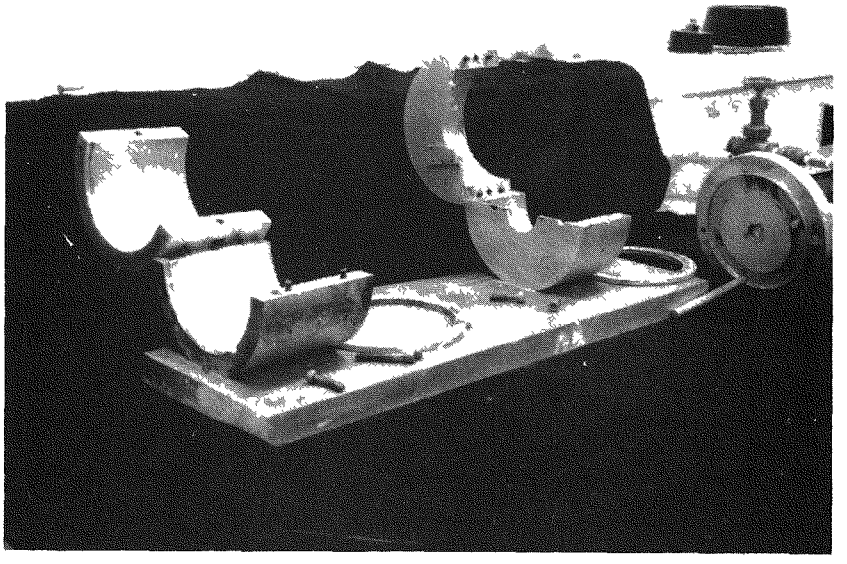


FIGURE 8. Aluminum Mold for Ceramic Hearth Casting (Disassembled)

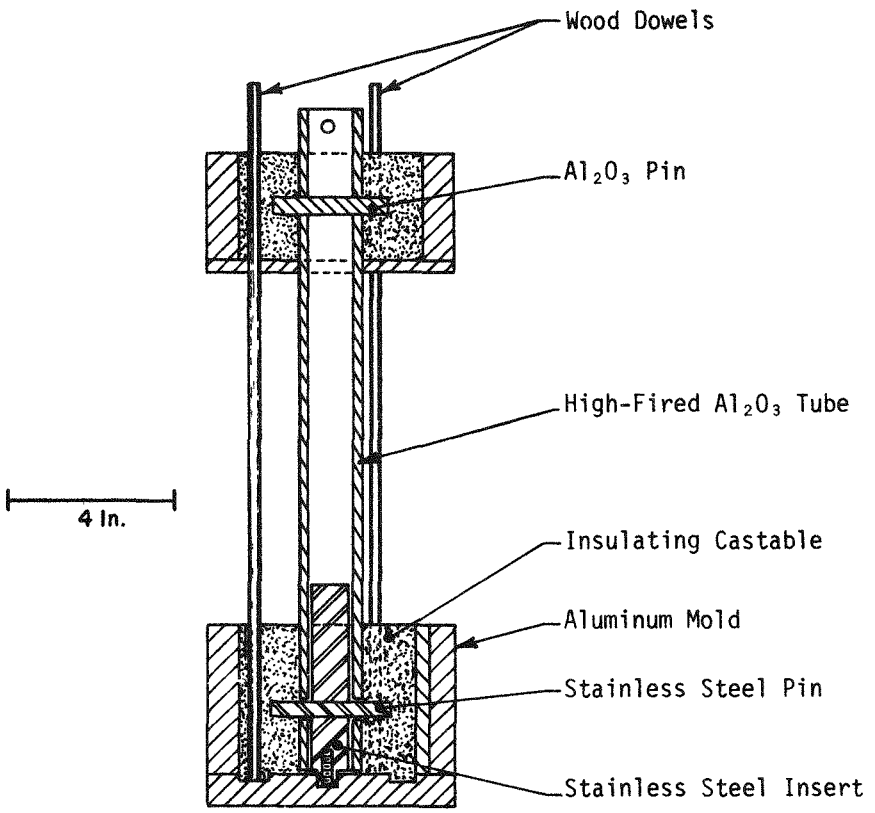


FIGURE 9. Cross-Sectional View of Ceramic Hearth and Mold

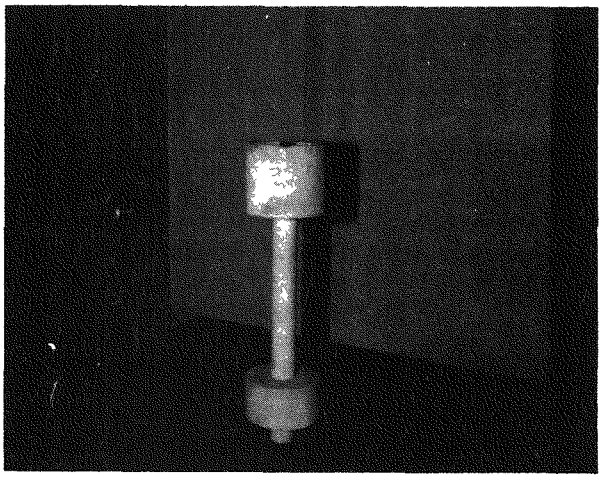


FIGURE 10. Ceramic Process Furnace Hearth

Alternate Hearth Design

Multi-piece hearth assemblies were also cast by modifying the mold as suggested by the Separations Technology Department of SRP. The multi-piece hearth has two advantages: (1) if any piece of the hearth is fractured during service, the piece itself may be replaced rather than replacing the complete hearth assembly and, (2) the cast sections of the hearth can be fired at a temperature high enough to promote sintering of the castable (not possible with a one-piece hearth because of temperature limitation of stainless steel) which is expected to help reduce flaking of the castable and reduce Al contamination in the PuO_2.

The following modifications of the previously described procedure were used to fabricate multi-piece hearths:

1. Wood dowels extending to the casting-mold interface were used as pins to temporarily hold the castings in place. The dowels were pre-drilled and coated with paraffin before casting.

2. The firing cycle described previously (Drying Step 4) was used for these hearths to remove the water and burn out the wood inserts. The hearths were disassembled and the donut-shaped castings were sintered at 1300°C for 2 hours. A heating and cooling rate of about 300°C/hr was used to minimize thermal stresses in the castings.

3. The hearths were reassembled by inserting an Al_2O_3 tube and a stainless steel pin was inserted through the upper castings, the Al_2O_3 tube, and the stainless steel insert.

Improved Hearth Design

Separations Technology suggested that the surface integrity of the alumina hearths should be improved to reduce Al contamination. Moreover, SRL suggested an improved hearth design which included encasing the insulating castable in high fired, closed-end, Al_2O_3 tubes to help reduce Al contamination in the $^{238}PuO_2$ feed material. The donut-shaped castables could be formed directly in the Al_2O_3 tubes with holes located for the gas inlet tube, thermocouples, and the center support tube located by using smaller diameter Al_2O_3 tubing.

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3. **Product Information, ALUNDUM Insulating Castables CA 330 and CA 333.** Norton Industrial Ceramics Division, Worcester, MA (October 1973).