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Progress Report

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**General-Purpose Heat Source Project and
Space Nuclear Safety and Fuels Program**

January 1980

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University of California



LOS ALAMOS SCIENTIFIC LABORATORY

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Progress Report

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January 1980

Compiled by

W. J. Maraman



By

ABSTRACT

This formal monthly report covers the studies related to the use of $^{238}\text{PuO}_2$ in radioisotopic power systems carried out for the Advanced Nuclear Systems and Projects Division of the Los Alamos Scientific Laboratory. The two programs involved are the following:

General-Purpose Heat Source Development

Space Nuclear Safety and Fuels.

Most of the studies discussed here are of a continuing nature. Results and conclusions described may change as the work continues. Published reference to the results cited in this report should not be made without the explicit permission of the person in charge of the work.

GENERAL-PURPOSE HEAT SOURCE PROJECT AND
SPACE NUCLEAR SAFETY AND FUELS PROGRAM

JANUARY 1980

Compiled by

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I. GENERAL-PURPOSE HEAT SOURCE

A. Management

The current schedule for the remainder of the General-Purpose Heat Source (GPHS) project is given in Fig. I-1.

B. Design Impact Tests

No impact tests were carried out this month. Plans for the CBCF insulation impact tests and the pyrolytic insulation impact tests were completed.

PHASE/TASK	FY79			FY80						FY81												
	J	A	S	O	N	D	J	F	M	A	M	J	J	A	S	O	N	D	J	F	M	
REDESIGN																						
IMPACT TESTS																						
PG																						
CBCF																						
VIBRATION TESTS (GE)																						
PG																						
CBCF																						
FINAL DESIGN DEFINITION																						
VERIFICATION																						
AGING (2)																						
IMPACT (4)																						
EXPLOSION (4)																						
FIRE																						
DYNAMIC TEST (GE)																						
PERFORMANCE ANALYSIS REPORT																						
FINAL REVIEW																						

Fig. I-1.
GPHS Project Schedule.

C. Fuel Development

The porosities of the edge and center of the Los Alamos Scientific Laboratory (LASL) fuel pellet from sample IRG-19 (GP 25) were determined to be 31.3 and 17.4%, respectively, while the porosities in the comparable regions of the fuel from IRG-23 (GP 30) were found to be 18.7 and 12.2%.

D. Clad Development

The accumulated exposure time of the Pt-3008 vs $^{238}\text{PuO}_2$ (1100°C, vacuum) was 12.3×10^7 h as of February 1, 1980.

E. Graphite Properties

1. Air Permeation Rate. A flow apparatus was constructed for measuring the permeation of air through the fine-weave, pierced fabric (FWPF) 3D graphite used in the GPHS program. A Wallace-Tiernan gauge was used to measure the differential pressure across the samples and the vacuum side was maintained at 1 mm with a forepump. The total void volume of the gas manifold was estimated to be 2035 cm³. The 50-mm-diameter samples were glued into the holder, one with the Z-axis aligned parallel and other with it perpendicular to the direction of gas flow. A third sample had a standard-diameter aeroshell plug, with the Z-axis perpendicular to the air flow. The plain samples were 5 mm thick, while the closure sample was 10 mm thick. Variations in the pressure and flow rate were measured as a function of time. The molecular flow rate (dn/dt) was calculated from the rate of pressure decrease by using the relationship:

$$dP/dt(VA/RT) = dn/dt = dV'/dt (1 \text{ atm}/RT) \quad [1]$$

where dp/dt = rate of pressure decrease (atm/s),
V = volume (cm³)
A = Avogadro's number (6.023×10^{23} molecules/mol),
R = gas constant (82.06 cm³ atm/K/mol),
dn/dt = molecular flow rate (molecules/s),
dV'/dt = standard flow rate (sccs).

The results from this study are listed in Table I-1. The measured time, pressure, and flow rate are listed along with the calculated molecular flow. The case where the flow is parallel to the Z-axis shows the highest flow rate at a given pressure, whereas the sample with the threaded plug displayed a rate between the other samples. This observation is expected since the former was a combination of the other two characteristic directions in the graphite. The flow rate is plotted as a function of the pressure in Fig. I-2 for the three samples. The diagram shows the significantly higher flow rates for the case of flow parallel to the Z-axis. The flow rate is plotted as a function of time in Fig. I-3 starting at the pressure of 300 mm for all three samples. If the thread root diameter is used to calculate the relative volumes of the two directions in the closure sample and the flow rates measured for the two solid samples, the calculated flow rate is in good agreement with the measured flow rate through the closure sample.

This study has shown that the FWPF 3D graphite does not appear to provide a major barrier to the flow of air into the interior of the GPHS module, especially in the case of the Z-axis parallel to the air flow direction and that the presence of the joint in the aeroshell has little or no effect on the total permeation rate.

TABLE I-1

PRESSURE AND FLOW RATES OF AIR PERMEATION THROUGH FWPF FOR FLOW PERPENDICULAR AND PARALLEL TO Z-AXIS AND FOR A THREADED CLOSURE

Time (sec)	P (mm)	Flow (sccm)	log flow (molecules/sec)
<u>A. Flow Perpendicular to Z-axis</u>			
0	760	494	--
20	700	413	20.296
42	650	362	20.176
64	600	320	20.176
92	550	277	20.071
125	500	232	20.000
163	450	195	19.938
203	400	161	19.865
323	300	102	19.758
528	200	58	19.508
696	150	39	19.293
937	100	26	19.136
1090	80	20	18.936
1410	50	13	18.791
<u>B. Flow Parallel to Z-axis</u>			
0	300	845	--
31	200	456	20.328
59	150	285	20.071
100	100	157	19.905
120	80	117	19.819
160	60	79	19.518
184	50	63	19.439
215	40	49	19.328
255	30	36	19.217
312	20	25	19.063
428	10	14	18.755
<u>C. Threaded Cap</u>			
0	670	980	--
13	700	802	20.550
37	500	575	20.439
75	400	393	20.372
130	300	235	20.079
172	250	169	19.895
229	200	117	19.762
315	150	74	19.584
445	100	42	19.404
485	90	36	19.217
530	80	31	19.166
580	70	27	19.120
640	60	23	19.041
717	50	19	18.933
815	40	15	18.828
950	30	11	18.689
1140	20	8	18.540
1495	10	4	18.269

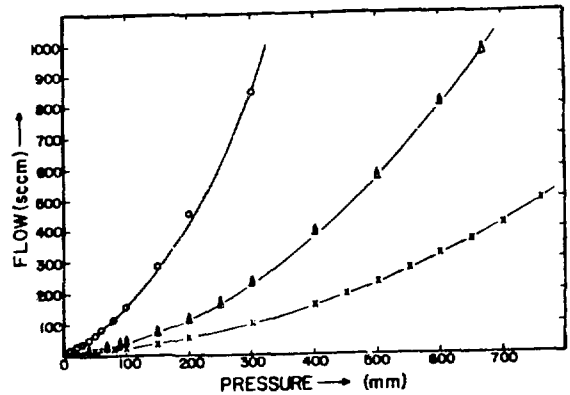


Fig. I-2. Pressure dependence of flow rate of air through FWPF graphite for flow parallel (o) and perpendicular (x) to Z-axis and for threaded cap (Δ).

2. Lock-Member Shear Tests. Some concern was raised regarding the shear strength of the GPHS locking members when the X and Y fiber directions were oriented at 45° to the shear loading direction. To determine this property, simulated shear tests were performed with the assembly shown in Fig. I-4. The locking plate was FWPF graphite with the X-Y axes at 45° to the loading direction. The locking members were bulk graphite.

Two tests with double lock members were performed. In the first test, failure occurred at a load level of 4.54 KN (1020 lbs) resulting in a failure stress of 31.6 MPa (4580 psi) per lock member. For the second test, the failure load was 4.71 KN (1060 lbs) and the failure stress was 32.8 MPa (4760 psi). The scatter in these two values

is probably a result of the statistical strength nature of brittle materials. In both tests, load-displacement curves were nonlinear, but lock member failure occurred in a brittle fashion.

Characteristics of failed lock members are shown in Fig. I-5. The lock members tended to fail by splitting down the mid-plane. The FWPF plate showed very little damage after test. In a previous test with the FWPF graphite X and Y axes at 0 and 90° to the shear loading direction, the failure load was 4.35 KN (979 lbs) with a stress per lock member of 30.3 MPa (4400 psi). This comparison shows that FWPF X-Y fiber orientation has virtually no effect on the shear of the locking members.

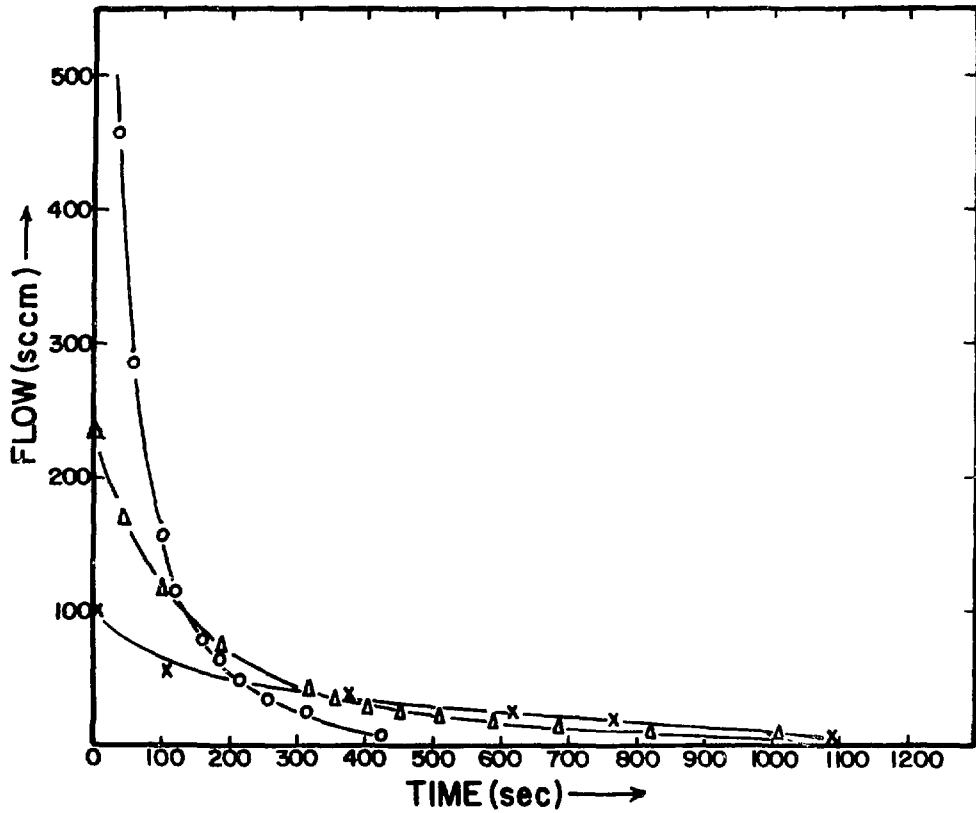
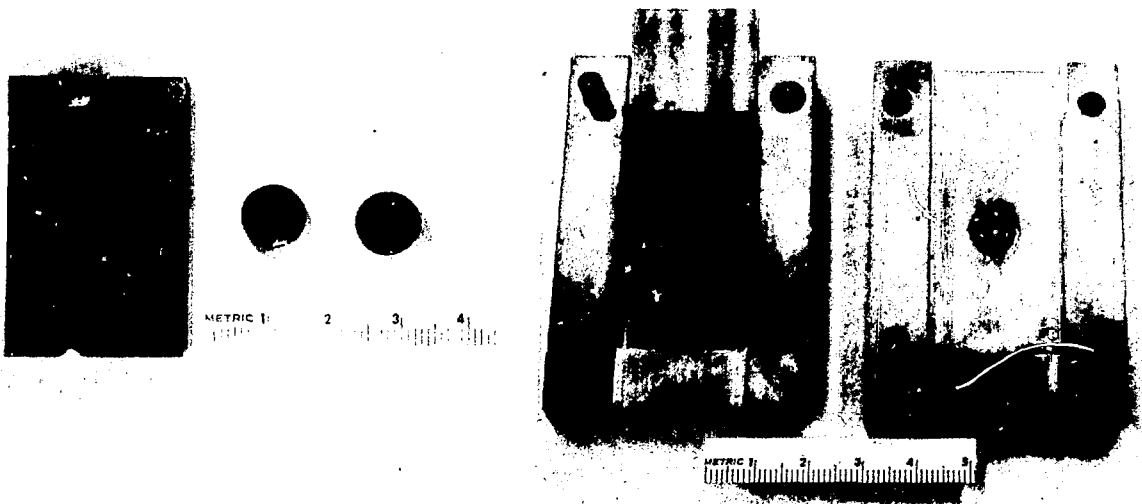


Fig. I-3.

Time dependence of air flow rate through FWPF graphite for flow parallel (o) and



(a)

Fig. I-4.

(b)

Shear strength test assembly for GPHS locking members. (a) FWPF locking plate and bulk graphite locking members and (b) graphite components in test fixture.

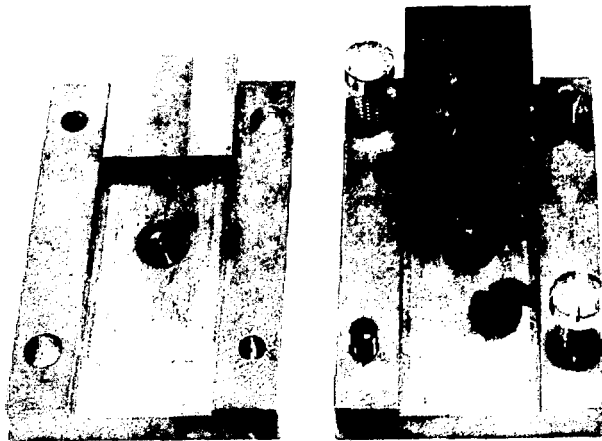


Fig. I-5.
Appearance of failed lock member
assembly after test.

II. ENVIRONMENTAL STUDIES

A. Terrestrial Environments

1. Environmental Chamber Experiments. Six terrestrial environmental chamber experiments are under way as described in Table II-1.

The fragments of Pressed Plutonium Oxide (PPO) greater than 6 mm in diameter (224 g) that resulted from the impact test of MHFT-12 have been exposed to simulated humid weather conditions on loam soil for 2409 days. The environmental system was programmed for summer conditions (20 to 40°C, 87 to 95% RH) for 174 days, followed by 182 days under winter conditions (0 to 17°C, 71 to 100% RH). The system was then operated for 173 days of its second summer, 207 days of the second winter, 193 days of the third summer, 169 days of the third winter, 211 days of the fourth summer, 174 days of the fourth winter, 195 days in its fifth summer, 154 days in its fifth winter, 168 days in its sixth summer, 185 days in its sixth winter, and 178 days in its seventh summer, before being programmed for its seventh winter. The averages of the plutonium contents from liquid scintillation analysis and the volume measurements of the water that percolated through the soil for the first five 32-mm winter rains show the increase in volume expected for the change to winter conditions, but the plutonium content was almost unchanged (Table II-2). The average water volume was 8.9 l, compared with 5.0 l last month, while the average plutonium content decreased slightly from 1.7 to 1.6 μCi . The rate of plutonium collection by the dehumidifier condensate stayed the same at 0.5 $\mu\text{Ci/wk}$.

The finer fraction of MHFT-12, which consists of 28 g of PPO particles with diameters between 0.01 and 6 mm, was placed on loam soil and subjected to 110 days of humid summer conditions, followed by 147 days of humid winter weather. Subsequently, the seasons changed on the same days as in the chamber containing the large pieces. It has now been in its seventh winter for 43 days. The average of the measured volumes for four 32-mm rains is higher, as is the aver-

TABLE II-1
TERRESTRIAL ENVIRONMENTAL CHAMBER EXPERIMENTS

<u>Experiment Number</u>	<u>Description</u>	<u>Date Started</u>
1	MHFT-12 chunks on loam; humid climate	6-21-73
3	MHFT-12 fines on loam, humid climate	9-27-73
6	MHFT-50 fines on loam; circular soil partitioner; humid climate	4-9-75
7	MHFT-27 fines on sand; arid climate	2-25-75
8	MHFT-50 chunks on loam; circular soil partitioner; humid climate	4-9-75
10	MHFT-27 chunk on sand; arid climate	2-26-75

age plutonium content (Table III-3). A fifth rain of 153 mm was not averaged with rest, as it would be expected to have a much higher volume and plutonium content. The average volume increased from 3.3 to 3.8 l, the average plutonium content from 0.36 to 0.53 μCi . The collection rate for plutonium by the dehumidifier condensate increased from 0.24 to 1.06 $\mu\text{Ci}/\text{wk}$.

The fragments of PPO greater than 6 mm in diameter (221 g) from the impact test of MHFT-27 have been on sand in an environmental chamber programmed for arid winter conditions (3 to 28°C, 21 to 87% RH) for 122 days, under arid summer conditions (26 to 53°C, 7 to 33% RH) for 196 days, under arid winter conditions again for 137 days, under arid summer conditions again for 211 days, in the third arid winter for 188 days, in the third summer for 181 days, 155 days in the fourth arid winter, 169 days in its fourth arid summer, 186 days in its fifth arid winter, 178 days in its fifth arid summer, and now in its sixth arid winter for 43 days. Liquid scintillation analysis and volume measurement of one 81-mm rain gave results of 0.36 μCi of plutonium in 11.2 l, compared with 0.04 μCi in 2.8 l last month. The collection rate for plutonium in the dehumidifier condensates was 0.10 $\mu\text{Ci}/\text{wk}$, compared with 0.11 $\mu\text{Ci}/\text{wk}$ last month.

The finer fraction of MHFT-27, 34 g of particles of PPO with diameters between 0.01 and 6 mm, has experienced the same weather and soil conditions as the larger pieces. Volume measurement and liquid scintillation analysis of

TABLE II-2

PLUTONIUM IN PERCOLATED RAINWATER
(LARGE PIECES FROM MHFT-12)

<u>No.</u>	<u>Date</u>	<u>Vol. Collected (ℓ)</u>	<u>Pu Found (μCi)</u>
332	12/17	6.6	1.2
333	12/26	8.4	1.4
334	1/2	9.0	1.8
335	1/7	11.2	2.2
336	1/14	<u>9.3</u>	<u>1.5</u>
Average		8.9	1.6

water that percolated through the soil for one 81-mm rain gave results of 0.185 μ Ci of plutonium in 6.4 ℓ of water, higher in plutonium than the 0.008 μ Ci of plutonium in 2.6 ℓ of water for the last rain. The dehumidifier condensates collected 0.08 μ Ci/wk of plutonium, higher than the 0.07 μ Ci/wk last month.

The pieces (186 g) of PPO greater than 2 mm in diameter from MHFT-50 after an impact test have been on loam soil in an environmental test chamber programmed for humid winter conditions for 79 days, under humid summer weather for 193 days, under winter humid conditions for 169 days, under summer humid conditions again for 211 days, under winter humid conditions for the third time for 188 days, in the third summer for 181 days, in the fourth winter for 155 days, in the fourth summer for 167 days, in the fifth winter for 185 days, 178 days in the fifth summer, and now 43 days into the sixth winter. The soil in this

TABLE II-3

PLUTONIUM IN PERCOLATED RAINWATER
(FINE MATERIAL FROM MHFT-12)

<u>No.</u>	<u>Date</u>	<u>Vol. Collected (ℓ)</u>	<u>Pu Found (μCi)</u>
324	12/17	4.0	0.57
325	12/26	6.8	1.40
326	1/2	0.8	0.02
327	1/7	<u>3.5</u>	<u>0.13</u>
Average		3.8	0.53
328	1/14	104.	10

TABLE II-4

PLUTONIUM IN PERCOLATED RAINWATER
(LARGER PIECES FROM MHFT-50 ON COMPARTMENTED SOIL)

No.	Date	Rainfall (mm)	Inner		Outer	
			Vol (ℓ)	Pu (μ Ci)	Vol (ℓ)	Pu (μ Ci)
241	12/17	34	1.4	0.070	11.8	0.14
242	12/26	34	1.4	0.019	13.2	0.10
243	1/2	34	1.4	0.089	14.5	0.11
244	1/7	23	1.8	0.167	8.2	0.36
245	1/14	34	3.2	0.157	14.5	0.17
Average			1.8	0.100	12.4	0.18

chamber is divided into two compartments. A thin cylindrical 45-cm-diameter steel shell separates the central portion of the soil, which contains the PPO, from the outer portion. The rainwater that percolates through the soil in the two compartments is collected separately for analysis. In the liquid scintillation analysis and volume measurements of rainwater from five rains, the average values for both compartments are higher this month except for the average plutonium content of the inner compartment, which changed very little (Table II-4). In the inner compartment the average volume was 1.8 ℓ and the average plutonium content was 0.100 μ Ci, compared with 1.5 ℓ and 0.109 μ Ci last month. In the outer compartment the average volume increased from 11.2 to 12.4 ℓ, the plutonium content from 0.13 to 0.18 μ Ci. The rate of collection of plutonium by the dehumidifier condensate increased from 0.7 to 2.0 μ Ci/wk.

The fine particles (68 g) of MHFT-50, with particle diameters between 0.01 and 2 mm, have experienced the same weather conditions as the larger pieces and are also on loam in a similarly compartmented tray. Liquid scintillation analyses and measured volumes of percolated rainwater from five rains show decreases in the average volumes and plutonium contents for both compartments (Table II-5). The averages for the outer compartment volumes and plutonium contents are 3.2 ℓ and 0.104 μ Ci, respectively, compared with 5.1 ℓ and 0.142 μ Ci last month. The inner compartment averages are 1.3 ℓ and 0.015 μ Ci, compared with 3.0 ℓ and 0.036 μ Ci last month. The collection rate for plutonium in the dehumidifier condensate changed from 0.4 to 0.2 μ Ci/wk.

The experiment in which a 254-g sphere of plutonium dioxide clad with iridium and encased in a Graphite Impact Shell (GIS), MHFT-23, was on sand under alternate winter and summer humid conditions for 1103 days was terminated April 17, 1978. A draft of the topical report for this experiment has been prepared and is being reviewed.

2. Terrarium Experiments. Chambers 5 and 9 have been fitted with fluorescent lights so that they can be used to measure plutonium uptake in growing plants and simple animals. A 24-h timer, wired to control the lights, was installed in each chamber and set in a summer cycle (sunrise at 7 AM and sunset at 8 PM). Blue grass was planted in each chamber, but two temperature excursions killed the grass in chamber 9. Baffles had been installed in the airstream of

TABLE II-5
 PLUTONIUM IN PERCOLATED RAINWATER
 (FINE MATERIAL FROM MHFT-50 ON COMPARTMENTED SOIL)

No.	Date	Rainfall (mm)	Inner		Outer	
			Vol (ℓ)	Pu (μ Ci)	Vol (ℓ)	Pu (μ Ci)
242	12/17	24	2.0	0.018	3.0	0.098
243	12/26	24	1.1	0.015	3.9	0.127
244	1/2	24	0.9	0.015	2.5	0.081
245	1/7	23	1.8	0.019	2.6	0.075
246	1/14	23	0.7	0.009	4.0	0.137
Average			1.3	0.015	3.2	0.104

these chambers to reduce the effect of strong winds, which seem to be inimical to the growth of the grass, probably because of rapid drying of the soil. The grass has been cut regularly and the cuttings analyzed for plutonium to establish a blank, which is 0.5 pCi/g, a value that is probably not significantly different from our lower detection limit. An improved method of cutting the grass is being sought, preparatory to the introduction of plutonium. The grass in chamber 9 has been replanted. Equipment to correct the control problem in this chamber has been installed.

3. Soil Column Experiments. Three soil column experiments are under way to test the transport of plutonium derived from plutonium oxide particles on the soil surface. The soil columns are made of silt loam soil, ~ 14 cm high and 2.5 cm in diameter. These columns are identified as 7, 8, and 9. Column 7 has 3.5 mg of PuO₂ particles on its soil surface, with a particle size distribution mainly in the 40- and 60-diameter range, column 8 contains 2.2 mg PuO₂ particles with an average diameter of 12 μ m, and 9 contains 2.6 mg with an average diameter of 7 μ m. These three size ranges are from the same three batches of plutonia particles that we are using in the experiments for measuring dissolution rate as a function of particle size. The columns are being eluted with distilled water (a rainwater simulant). The columns have been in operation for 1164 days. Measurements made after 1066 days of operation (Table II-6) show that the flow rates have not changed significantly. The variation in flow rate between columns is unexpected, inasmuch as they all contain soil from the same source. Radiation measurement show that at least 99% of the PuO₂ particles still remain at the top of each column; radiation cannot be detected from any particle that may move downward into the soil columns. Almost 90% of the plutonium found in the eluates was in the collection representing the first 5 days of operation. This was probably from the water suspension of particles used to place the particles on the columns.

B. Aquatic Environments

1. Aqueous Release Rates As A Function of PuO₂ Particle Size. Three sets of sized ²³⁸PuO₂ particles, each having a different size distribution, are

TABLE II-6
Pu TRANSPORT FROM PuO₂ PARTICLES

Column	Elution Duration			
	1066 days		1164 days	
	Total Vol (ℓ)	Total Pu (ng)	Total Vol (ℓ)	Total Pu (ng)
7	77.4	30.3	83.8	31.5
8	67.4	27.0	73.2	20.4
9	35.8	39.9	39.3	40.9

suspended in 1M HClO₄ in 50-ml volumetric flasks in a 37°C water bath. Periodically samples are removed, centrifuged to remove suspended particles, and then analyzed for plutonium. These data are needed to obtain information about dissolution under carefully controlled solution conditions and to determine the effects of particle size on dissolution rates.

The data (Table II-7) have been collected for 1534 days for the three particle size distributions. Each of these particle size distributions was used to prepare 4 experiments, 2 containing 2 mg of particles and 2 containing 10 mg. Distribution A consists of the largest particles, having a size range of ~ 1 to 120 μm. Distribution B is intermediate in particle size and distribution C contains a preponderance of the smallest particles. The dissolution rates, grams dissolved/second (g/s), have changed only slightly during the past 3 months, and, as expected, the smaller particles release plutonium at a faster rate than the larger ones.

A comprehensive review of this experiment, including a more detailed characterization of the three particle distributions, has been completed. The review revealed that the large particles in distribution A were actually aggregates of smaller particles. The suspension technique used in the particle sizing produced a combination of "large particles" (did not suspend) and small particles thought to be individual members of aggregates that had broken apart. It is not possible to determine a quantitative estimate for the amount represented by the large fraction, therefore, the calculation of an available surface area is not possible.

Distribution B contained both large and small particles. In this case the large particles consisted of a preponderance of single large particles with some fines adhering to them, whereas the large particles in distribution A consisted of aggregates of similar size particles. Again, it is not possible to determine a total surface area because the sizing technique does not produce a quantitative estimate for the amount represented by each fraction.

Distribution C behaved normally in that all particles could be suspended and there were no anomalous large particles. This distribution lends itself to a calculation of total surface area but there is no assurance that the initial area has remained constant. In fact, it can be argued that the surface area has changed during the course of the experiment.

Grams dissolved versus time plots of representative experiments (A-1, B-1, C-1) exhibits no discernible discrepancies. Each plot shows increasing dissolution of ²³⁹PuO₂ with C-1 > B-1 > A-1. Integral dissolution rate plots show

TABLE II-7
SUMMARY OF DISSOLUTION EXPERIMENT IN 1M HClO₄

Experiment Number	PuO ₂ (mg)	Percent Dissolved	Dissolution Rate	
			(g/s)	(g/s-g)
A1	2.02	4.8	7.3x10 ⁻¹³	3.6x10 ⁻¹⁰
A2	2.05	4.5	6.9x10 ⁻¹³	3.4x10 ⁻¹⁰
A3	10.19	4.5	3.5x10 ⁻¹²	3.4x10 ⁻¹⁰
A4	9.90	4.5	3.4x10 ⁻¹²	3.4x10 ⁻¹⁰
B1	2.02	8.2	1.3x10 ⁻¹²	6.2x10 ⁻¹⁰
B2	2.03	9.7	1.5x10 ⁻¹²	7.3x10 ⁻¹⁰
B3	10.25	8.4	6.6x10 ⁻¹²	6.5x10 ⁻¹⁰
B4	10.45	8.5	6.8x10 ⁻¹²	6.5x10 ⁻¹⁰
C1	2.23	18.6	3.2x10 ⁻¹²	1.4x10 ⁻⁹
C2	2.31	17.7	3.1x10 ⁻¹²	1.3x10 ⁻⁹
C3	9.89	17.8	1.3x10 ⁻¹¹	1.4x10 ⁻⁹
C4	10.67	17.4	1.4x10 ⁻¹¹	1.3x10 ⁻⁹

significant differences among the three experiments. A-1, coarse particles, has a shallow minimum at $\sim 3 \times 10^7$ s, whereas B-1, medium size particles, has a low maximum at $\sim 2 \times 10^7$ s. C-1, fine particles, exhibits a strong maximum at $\sim 2 \times 10^7$ s and has since decreased by 30%. The maximum is 3 times that of B-1. It can be postulated that the initial rapid increase in the dissolution rate of C-1 represents the solubilization of the smallest particles. The rate is now decreasing because of the reduction in surface area since only "larger" particles remain.

The third column in Table II-7 lists the percent of original material that has dissolved. Since a substantial amount of the original material has dissolved, $\sim 17\%$ in the case of experiment C, the distribution of particles in each experiment may have changed considerably. An ultrafiltration technique is being investigated whereby the size distribution in each sample may be determined. If the technique is successful part or all of the experiment will be terminated.

An extraction procedure, designed to extract only Pu⁴⁺, is being applied to these experiments. Preliminary results indicate that the plutonium in solution is 90%, or greater, in the ionic form. Work is continuing in this area.

2. Fuel Form Exposures.

a. Seawater. The large 25-W pellet, HPZ-59-4, has been in cold seawater for 1923 days (Table II-8). Its release rate is 14 nCi/m²-s, down 12% from last month.

The tidal simulation aquarium, using the 19-W source HPZ-186-4, has been under way for 814 days. The source is on sand in a stainless steel tray that is lowered into the water and subsequently raised above the surface twice

TABLE II-8
SUMMARY OF
Pu RELEASE RATE FROM PPO IN AQUATIC ENVIRONMENTS

Sample	Power (W)	Immersion (days)	Water	Temperature (C)	Release Rate (nCi/m ² -s)
HPZ-60-2	2.5	2130	Fresh	10	130
HPZ-111-1	25	1829	Fresh	10	370
HPZ-59-4	25	1923	Sea	10	14
HPZ-174	18	818	Sea	10	6
HPZ-186-4	19	814	Sea (Tidal)	10	56

every 24 hours. Figure II-1 is a plot of plutonium concentration in $\mu\text{g}/\ell$ versus time for the tidal simulation experiment. On day 103 (point A on Fig. II-1) a crack was observed on the face of the source and on day 393 (B) the source had split into two pieces. By day 404 (C) the source had separated into seven pieces. On day 451 (D) the elevator motor failed and the source was left submerged in the seawater. On day 481 (E) a sample of the source was removed for metallurgical examination and five sand samples were obtained from the tray on the bottom of the aquarium. At the present time the source is fragmented into 30 or more pieces. Therefore, a calculation of release rate per unit area is not possible because there is no reasonable estimate of the source surface area. On the basis of its original area, the release rate is $56 \text{ nCi}/\text{m}^2\text{-s}$, down 5% from last month's value.

The five sand samples, taken from the four corners and the center of the tray, were analyzed for plutonium content. The results ranged from $55.1 \mu\text{g}$ to $26.2 \mu\text{g}$ with an average of $42 \pm 12 \mu\text{g}$. Based on this average and on the surface areas of the sand tray and the sand samples it can be estimated that there are 28 mg of plutonium on or in the sand.

The 18-W pellet HPZ-174 has been immersed in seawater at 10°C for 818 days. The release rate is $6 \text{ nCi}/\text{m}^2\text{-s}$, down 14% from the value for the last 4 months. This is the same system in which we maintained a live sea cucumber for 230 days.

b. Fresh Water. The 2.5-W pellet, HPZ-60-2, is part of a set of experiments to compare release rates from PPO in seawater and in fresh water. Its average release rate after 2130 days of immersion is $130 \text{ nCi}/\text{m}^2\text{-s}$, unchanged from last month's value. This is the second highest release rate observed in this set of PPO pellets.

The fastest release rate is shown by the 25-W pellet, HPZ-111-1, immersed for 1829 days at 10°C . Its average release rate is $370 \text{ nCi}/\text{m}^2\text{-s}$, down 5% from the value observed last month. The highest rates from PPO are found in 10°C fresh water, and the lowest in warm seawater. There is little, if any, rate difference caused by a 10-fold difference in power level.

c. Source Term. The plutonium release rates shown in Table II-8 are essentially minimum release rates in that the removal of plutonium by deposition on underwater surfaces cannot be measured. A total release rate, or source term, is not obtainable in an aquarium experiment. Therefore, a glass chamber was designed and constructed to permit such measurements.

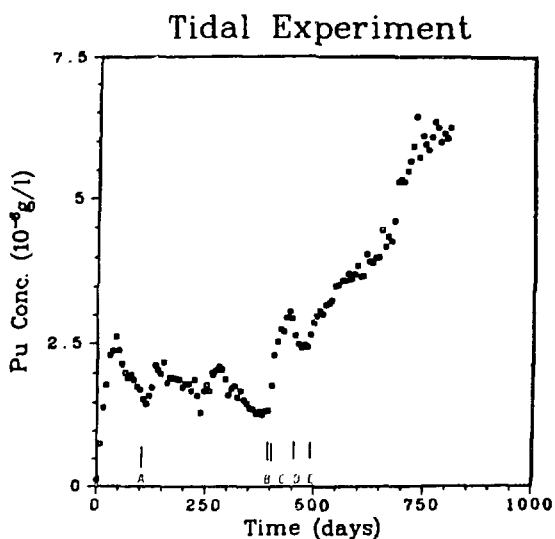


Fig. II-1.
Plutonium concentration in the Tidal
Simulation Experiment.

This chamber is a 15.25-cm i.d. cylinder, 25.4-cm tall. It can be separated into two parts by means of an O-ring joint midway up the cylinder. The top half has a side arm for attaching a membrane filter and an O-ring sealed port through which water samples can be withdrawn. The bottom half contains a glass pedestal for positioning the plutonium source.

A 48.6-g, bare $^{238}\text{PuO}_2$, source, HPZ-186-2, was positioned in an empty glass chamber on December 20, 1977, and the chamber was placed in an aquarium at 10°C for temperature control. This experiment will provide baseline data for an unclad source exposed to air only. Such conditions occur in some of the environmental chambers. The $0.05\text{-}\mu\text{m}$ pore-size filter is changed weekly and analyzed for ^{238}Pu . This experiment was terminated on day 454. Only 2 filters out of 63 had shown any plutonium, 6 pg on one and 2 pg on the other. Neither result was confirmed by subsequent filters and are, therefore, attributed to contamination from an unknown source external to the experiment. The source was transferred to a second chamber and the first chamber was resealed and transferred for analysis. The source in the second chamber was submerged in deionized water for 30 min and then removed. At this point the source fractured into three pieces and further experiments with this source were cancelled. The second chamber was resealed in preparation for analysis.

Analysis of the first chamber consisted of the quantitative recovery of the $^{238}\text{PuO}_2$ on the interior surfaces. This was accomplished using a $\text{HNO}_3\text{-HF}$ leach followed by a wash with a decontamination solution. No residual activity could be detected on the surfaces available for monitoring.

The total ^{238}Pu recovered was $1.41\ \mu\text{g}$, which represents a release rate per unit surface area of $22.1\ \text{pg}/\text{m}^2\text{-s}$. This number can be compared to the release rates seen in the aquatic environment experiments which vary from 0.59 to $36\ \text{ng}/\text{m}^2\text{-s}$. The analysis of the second part of the experiment in which the source was submerged indicated the release of $49.3\ \mu\text{g}$. Calculations based on the same surface area and a 30-minute submergency yields a release rate of $16.8\ \mu\text{g}/\text{m}^2\text{-s}$. This calculation may be invalid since the initial release may be that of loose material on the "old" surface of the source. The next experiment, that of the release from a "new" surface of the same source, could not be performed because the source broke into three parts.

Four glass chambers are being prepared for the next sequence of experiments. These experiments will use GROG type source material and will consist of two fresh water systems and two simulated seawater systems, one each at 10°C and one each at 37°C. This sequence should provide information on the effects of the type of water used and the temperature of the water.

When the four chambers were placed in two aquariums, three chambers leaked at the center seal. Neoprene rubber gaskets were found to provide a suitable seal. These experiments will be initiated during the next report period.

d. Water Samplers. Two water samples were shipped to the Naval Ocean Systems Center at San Clemente Island. On May 15, 1979, the two samplers were attached to two cages and placed on the ocean floor. One sampler was placed in the Cermet area and activated by navy divers. The second sampler was placed and activated on the ocean floor approximately 60 feet NNE of the first unit. These two units were designed to collect 1.5 l of water in 90 days. They have been received back at Los Alamos.

Sample #1 has been opened and the contents removed. Severe corrosion was observed on all four valves and a deposit was found on the interior walls and on the top of the piston. This deposit was easily removed with a water wash and a light rubbing. Samples of the interior deposit and samples of the deposit on the exterior of the sampler will be submitted for elemental analyses.

The total seawater sample obtained during the 3-month sampling period amounted to 440 ml. It was noted that the lower receiver had lost its vacuum. Corrosion of the vacuum valve may have resulted in this vacuum loss and subsequent equalization of the pressure on both sides of the piston which terminated sampling.

The 440 ml sample, the sludge deposit, and the tissue used to wipe the interior walls were treated with $\text{HNO}_3\text{-HClO}_4$. The resulting solution was highly colored (yellow) which produced considerable quenching during liquid scintillation counting. Therefore, the sample was split into four aliquots. Two aliquots were processed using a microporous anion exchange resin. This procedure had been previously tested with simulated seawater spiked with ^{238}Pu in the concentration range expected in the water samplers. The recovery of plutonium for the test with the spiked simulated seawater was $96 \pm 10\%$. However, when the aliquots of the solution from the water sampler were processed by this anion exchange resin procedure, values of 230 and 13 d/m were obtained. Because the first aliquot was handled in an area where cross contamination was possible, the high value was discarded. The aliquots processed by lanthanum fluoride precipitation produced values of 49 d/m and 16 d/m. The three aliquots give an average of $(9.6 \pm 7.3) \times 10^{-12}\text{g/l}$ for the concentration of ^{238}Pu in the seawater at the Cermet site. The second sampler, containing a seawater sample from outside the Cermet site, is being processed at this time.

C. Analytical Studies of PuO_2 -Soil Agglomeration (Purchase Order LP9-3589K; LFE Environmental)

A progress report was not received from LFE Environmental this month.

Cumulative costs on this purchase order through November, 1979, were \$5,752, leaving a balance of \$7 828. This purchase order has been extended to a new termination date of September 30, 1980. Final payment of \$3 775.68 was made on the previous purchase order, LP-8-8136D, for a total payment of \$18 878.33 at close out.

III. SYSTEMS SUPPORT

A. Stirling Isotope Power System

The accumulated exposure time for the 800°C test assembly was 18 202 h as of February 1, 1980. We expect to remove this sample from test and examine it within the next two or three months in support of our efforts to evaluate the effects of the delayed Galileo launch on the heat source components.

B. Multi-hundred Watt

1. Impact Test. The Multi-hundred Watt (MHW) fueled sphere assembly (FSA) MHFT-61 was examined after aging, reentry, and impact.

The components for this FSA were identified as follows:

Plutonia sphere	: HS-2
Iridium Hemi-shells	: DOP-26 5-1 and 5-4
GIS	: 936837-2

The aging treatment consisted of 8 834 h exposure at 1210°C (GIS) in vacuum. Continuous monitoring of the helium signal derived from the FSA during the aging treatment indicated that at least one vent remained open throughout the test exposure.

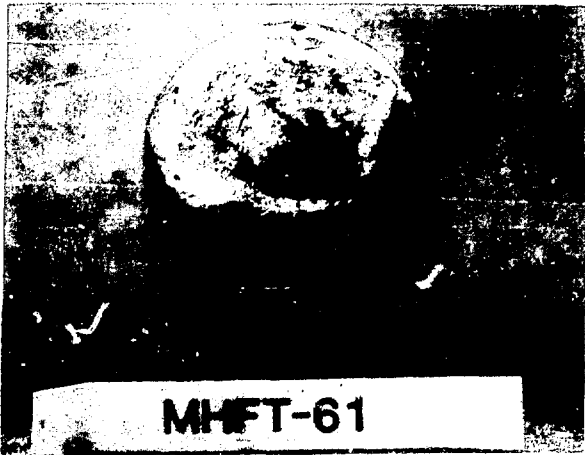
Following a safety verification test reentry pulse to 1550°C, post-impact containment shell (PICS), the FSA was impacted at 74 m/s and 1440°C (PICS). The cap of the GIS was oriented at 180° to the initial impact point.

The height strain of the impacted post-impact sphere assembly (PISA) was -23.5% and the diametral strain was +6.9%. Two linear punch-through strains were observed on the impact face of the PISA, as shown in Fig. III-1(a) and in the profile views (b and c) in the figure. The iridium PICS was not breached at these locations.

Small "fingerprint" type cracks were present on the impact face, however, visual examination of the interior of the PICS did not indicate that there was penetration. The back side of the PISA showed several locations at which the iridium was stretched over the edge of fuel sphere fragments. No failures of the iridium occurred at these locations. Figure III-2 illustrates the mode of the fuel sphere crush-up. The shear plane between fuel fragments which resulted in one of the punch through strains of the PICS is evident in the section on the left-hand side of the photograph.

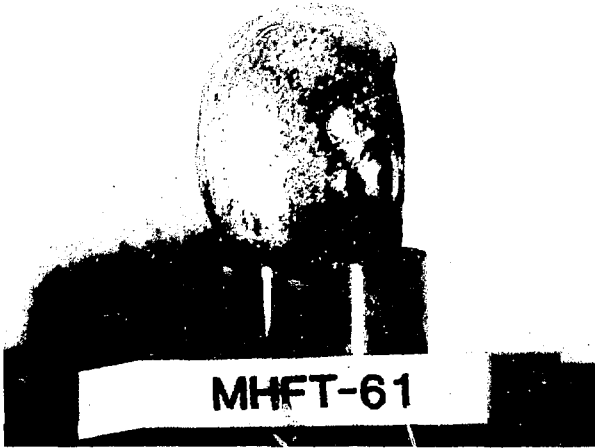
It is apparent that the lower velocity of impact (74 m/s vs the 85 m/s used for the LES 8/9 and Voyager MHW impact tests) resulted in decreased overall deformation of the PISA, however, a complete evaluation of the test will require metallographic determination of the iridium grain size and Auger analysis of grain boundary chemistry.

2. Iridium Impurity Effects. We have continued to examine the effect of impurities on iridium performance in MHW impacts. This month we concentrated on a further examination of the effect of phosphorus on grain growth. We reported previously that DOP-26 Ir-0.3% W suffers enhanced grain growth when annealed in the presence of P₂O₅. However, a clear trend that phosphorus affects grain growth in Ir-0.3% W PICS has never been established. Previous experiments with phosphorus effects on grain growth and grain boundary chemistry are summarized in the top portion of Table III-1. The effect of phosphorus available from P₂O₅ is clearly established. There is some different grain size response depending on the specific lot of DOP-26 alloy examined.



(a)

Fig. III-1.
Photographs of the impacted PISA from
MHFT-61. (a) Impact face, and (b & c)
profile views.



(b)



(c)

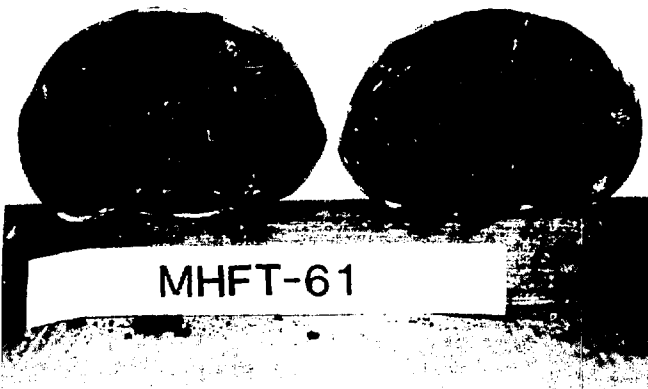


Fig. III-2.
Photograph of the impacted fuel sphere
of MHFT-61.

TABLE III-1
GRAIN SIZE AND AES RESULTS FOR P-DOPING
EXPERIMENTS ON Ir-0.3% W

Specimen	Lot	Treatment	Grain Size # grains/thickness	Auger Intensities	
				Th ₆₅ /Ir ₂₂₉	P ₁₂₀ /Ir ₂₂₉
DOP-26	GPHS Lots	1 h at 1500°C in vacuum ^a	25-28	--	--
DOP-26	GPHS Lots	19 h at 1500°C in vacuum	13	--	--
DOP-26	Early DOP-26 Lot, (1977)	18 h at 1575°C in vacuum	8.5	0.60	0
OLMF-8-65	OLMF-8	19 h at 1575°C in vacuum	7.4	--	--
L-30-1	L-30	19 h at 1575°C in vacuum	6.2	0.34	0
DOP-26	Early DOP-26 Lot, (1977)	18 h at 1575°C with P ₂ O ₅	5.6	0.81	0.40
L-30-5	L-30	19 h at 1500°C with P ₂ O ₅	8.7	0.45	0.36
OLMF-8-61	OLMF-8	19 h at 1500°C with P ₂ O ₅	7.8	0.81	0.38
OLMF-8-44	OLMF-8	1 h at 1500°C with P ₂ O ₅	19	0.82	0.47
OLMF-8-44B	OLMF-8	1 h at 1500°C with P ₂ O ₅ + 18 h at 1500°C in vacuum	4.7	--	--
OLMF-8-5	OLMF-8	18 h at 1500°C in vacuum + 1 h at 1500°C with P ₂ O ₅	12.6	--	--

In order to determine if phosphorus present in the iridium, rather than in the annealing atmosphere, will also enhance grain growth, we ran the following experiment. Sample OLMF-8-60 was annealed with P₂O₅ for 1 h at 1500°C. The grain size was measured as 19 grains per thickness, compared to 25 to 28 per thickness typically observed for DOP-26. As shown in Table III-1, the grain boundaries contained high phosphorus concentrations. A piece of this specimen (OLMF-8-44B) was then annealed for an additional 18 h at 1500°C in vacuum in a clean graphite crucible. The grains grew enormously, to average 4.7 grains/thickness. The grain growth was very nonuniform with some grains as large as two-thirds of the thickness of the specimen (see Fig. III-3). These results indicate that phosphorus in the grain boundaries of iridium will enhance grain growth significantly even in the absence of phosphorus in the atmosphere.

Since the 1 h at 1500°C treatment was successful in putting phosphorus in the grain boundaries, it makes it possible to control the grain size of phosphorus-doped specimens. To achieve a grain size of ~12 grains/thickness, specimens were

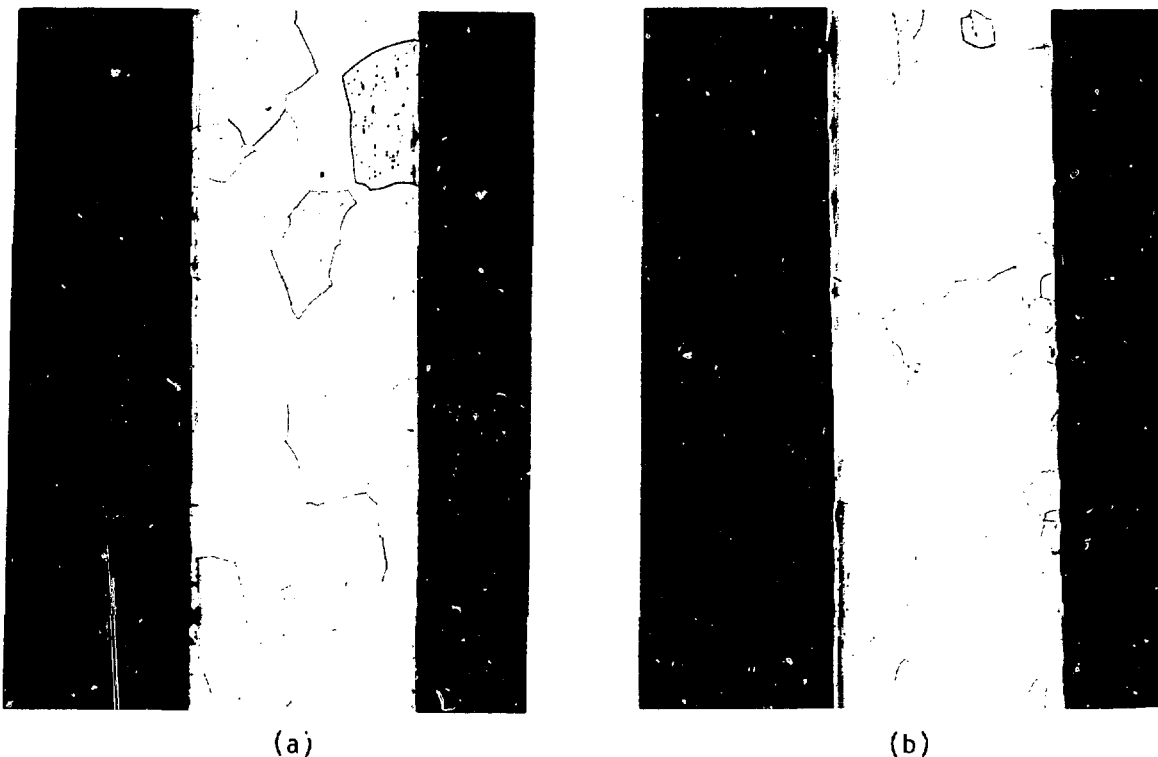


Fig. III-3.

Metallographic cross sections of OLMF-8-44B annealed 1 h at 1500°C with P_2O_5 plus 18 h at 1500°C in vacuum. a) and b) represent cross sections from the same specimen.

annealed at 1500°C for 18 h in vacuum followed by 1 h at 1500°C with P_2O_5 . The resulting grain size was 12.6 grains/thickness (see Table III-1). We are currently examining this material by Auger Electron Spectroscopy to check the phosphorus levels at the grain boundaries. Four tensile specimens that received this treatment were sent to Oak Ridge National Laboratory (ORNL) for tensile impact testing. One small section was sent to Don David at Mound Facility (MF) for Auger studies. One disk for biaxial testing was also given the phosphorus-doping treatment.

3. Fuel Impurities. An attempt to detect phosphorus impurity in the plutonia spheres of assemblies on hand at LASL by gamma-ray spectroscopy has been carried out. The technique yields analytical data for aluminum and sodium in addition to phosphorus. The results obtained with FSAs MHFT-67 through -72 are summarized in Table III-2 along with available corresponding analyses from Savannah River (SR). The agreement between the analyses for sodium are quite good, but less so for aluminum, with the gamma-ray results showing lower concentrations than the emission spectrographic technique. The gamma-ray results indicate the phosphorus content to be very low.

TABLE III-2
 SUMMARY OF γ -RAY SPECTROSCOPY
 ANALYSIS OF PHOSPHORUS, ALUMINUM, AND SODIUM
 IN MHW-FSAs

	Phosphorus		Aluminum		Sodium	
	γ -ray	SR*	γ -ray	SR	γ -ray	SR
MHFT-67	<25	--	--	>200	--	10
MHFT-68	<20	--	20	75	20	11
MHFT-69	<20	--	29	>200	--	25
MHFT-70	<20	--	10	75	19	20
MHFT-71	<20	--	7	75	10	10
MHFT-72	<20	--	24	--	--	--

*SR analysis on feed powder from HB line.