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Progress Report

DR. 961

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

November 1979

MASTER

University of California



LOS ALAMOS SCIENTIFIC LABORATORY

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Progress Report

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November 1979

Compiled by

W. J. Maraman

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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 three programs involved are the following:

- General-Purpose Heat Source Development
- Space Nuclear Safety
- Fuels Program.

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

NOVEMBER 1979

Compiled by
W. J. Maraman

I. GENERAL-PURPOSE HEAT SOURCE

A. Management

The current-schedule for the General-Purpose Heat Source (GPHS) project is given in Fig. I-1 for the two insulation options. The pyrolytic-graphite (PG) design was designated as the prime design by the Department of Energy (DOE) this month, but work will continue on the potentially lighter CBCF design. Fairchild-Industries (FI) is creating the CBCF design. FI is creating the CBCF design and will test it for vibration resistance. A satisfactory design will then be subjected to impact tests at LASL.

B. Design Impact Tests

Three impacts were conducted this month to evaluate the effects of a high temperature reentry pulse and the use of CBCF on impact performance.

1. DGCI-27-R1-23. This was double impact at 27°. The graphite impact shell (GIS) was of design DGIS-12, shown in Fig. I-2, with a side wall thickness of 4.24 mm, and end-wall thickness of 5.84 mm with a 16% area fraction of holes in the end, and standoff rings, 0.25 mm high, in the end and on the side wall (Fig. I-3). It contained a single threaded closure on the impact end and a standard web. The GIS was machined from fine-weave, pierced fabric (FWPF) 3D carbon-carbon composite and outgassed 1 h at 1800°C. The GIS was enclosed in a bulk graphite liner (ATJ-S) 1.55 mm thick. The leading capsule was IRG-44, Ir-0.3% W. It was annealed for 20 h at 1500°C before welding and subjected to a thermal reentry pulse with the $^{238}\text{PuO}_2$ pellet after welding. The maximum temperature of the reentry pulse was 1740°C, with 6 minutes above 1300°C. The reentry temperature profile is shown in Fig. I-4. The capsule contained a $^{238}\text{PuO}_2$ fuel pellet of 84.9% theoretical density. The trailing capsule was GCI-26R, a reused Pt-3008 capsule containing a UO_2 pellet. The assembly was impacted at 27°, 58 m/s, and 965°C.

The leading capsule survived impact without failure. The GIS crushed at the impact corner, tore off circumferentially and split partially up the side wall, as shown in Fig. I-5. The impacted capsule IRG-44 is shown in Fig. I-6. The overall strains were substantial, as shown in Table I-1. A severe local bend along a longitudinal crack in the fuel is shown in Fig. I-6. The magnitude of the local strains at the bend will be measured after the capsule is sectioned. The maximum tensile strain of the weld was 9.2%, which did not cause weld failure.

| 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 | | | | ▲ | | | | | | | | | | | | | | | | | | |
| VIBRATION TESTS | | | | △ | △ | | | | | | | | | | | | | | | | | |
| PHYSICAL PROPERTIES | | | | △ | | | | | | | | | | | | | | | | | | |
| REENTRY SENSITIVITY CALCULATIONS | | | | ▲ | | | | | | | | | | | | | | | | | | |
| 3D REENTRY CALCULATIONS | | | | △ | △ | | | | | | | | | | | | | | | | | |
| ATTITUDE CONTROL TESTS | | | | △ | | | | | | | | | | | | | | | | | | |
| FINAL DESIGN | | | | | | △ | | | | | | | | | | | | | | | | |
| VERIFICATION | | | | | | | | | | | | | | | | | | | | | | |
| AGING | | | | | | | → | | | △ | | | | | | | | | | | | |
| IMPACT | | | | | | | → | | | △ | | | | | | | | | | | | |
| EXPLOSION | | | | | | | → | | | △ | | | | | | | | | | | | |
| FRAGMENT | | | | | | | → | | | △ | | | | | | | | | | | | |
| FIRE | | | | | | | | | | △ | | | | | | | | | | | | |
| REENTRY CALCULATIONS | | | | | | | | | | | | | | | | | | | | | △ | |
| DYNAMIC TEST (GE) | | | | | | | | | | △ | | | | | | | | | | | | |
| THERMAL | | | | | | | | △ | | | | | | | | | | | | | | |
| PERFORMANCE ANALYSIS REPORT | | | | | | | | | | | | | | | | | | | | | △ | |
| FINAL REVIEW | | | | | | | | | | | | | | | | | | | | | | △ |

(a)

| 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 | | | | | | △ | | | | | | | | | | | | | | | | |
| VIBRATION TESTS | | | | | | △ | | | | | | | | | | | | | | | | |
| PHYSICAL PROPERTIES | | | | | | △ | | | | | | | | | | | | | | | | |
| REENTRY SENSITIVITY CALCULATIONS | | | | ▲ | | | | | | | | | | | | | | | | | | |
| 3D REENTRY CALCULATIONS | | | | | | △ | | | | | | | | | | | | | | | | |
| ATTITUDE CONTROL TESTS | | | | | | △ | | | | | | | | | | | | | | | | |
| FINAL DESIGN | | | | | | | △ | | | | | | | | | | | | | | | |
| VERIFICATION | | | | | | | | | | | | | | | | | | | | | | |
| AGING | | | | | | | → | | | △ | | | | | | | | | | | | |
| IMPACT | | | | | | | → | | | △ | | | | | | | | | | | | |
| EXPLOSION | | | | | | | → | | | △ | | | | | | | | | | | | |
| FRAGMENT | | | | | | | → | | | △ | | | | | | | | | | | | |
| FIRE | | | | | | | | | | △ | | | | | | | | | | | | |
| REENTRY CALCULATIONS | | | | | | | | | | | | | | | | | | | | | △ | |
| DYNAMIC TEST (GE) | | | | | | | | | | △ | | | | | | | | | | | | |
| THERMAL | | | | | | | | | | | | | | | | | | | | | | |
| PERFORMANCE ANALYSIS REPORT | | | | | | | | | | | | | | | | | | | | | △ | |
| FINAL REVIEW | | | | | | | | | | | | | | | | | | | | | | △ |

(b)

Fig. I-1.

General-Purpose Heat Source project schedules. (a) Schedule for the pyrolytic graphite insulator design; (b) estimated schedule for the CBCF-3 insulator design.

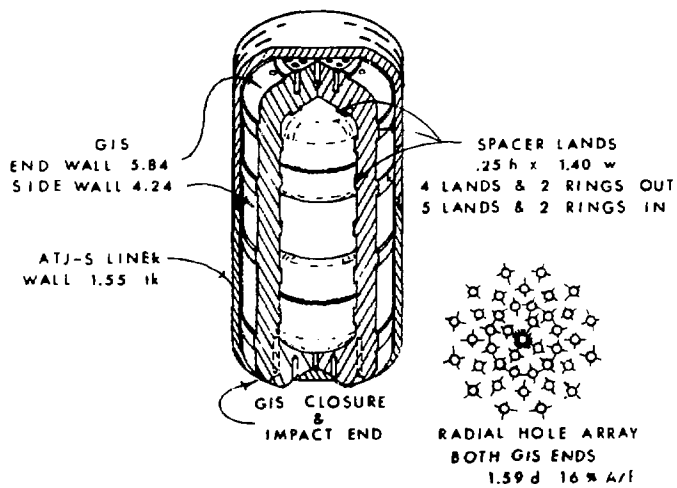


Fig. I-2.
Schematic drawing of DGIS-12 (DGC1-27-R1-23) showing details of graphite impact assembly.

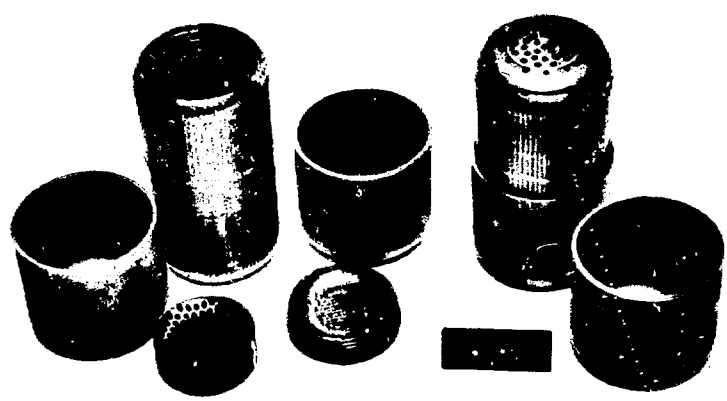


Fig. I-3.
DGIS-12 used in DGC1-27-R1-23. Photograph shows two sets of hardware, unassembled at left and partially assembled at right. The FWPF GIS contained standoff rings on both the interior and exterior cylindrical walls and on the interior and exterior of the impact end. The bulk liners were machined to fit over the exterior of the GIS.

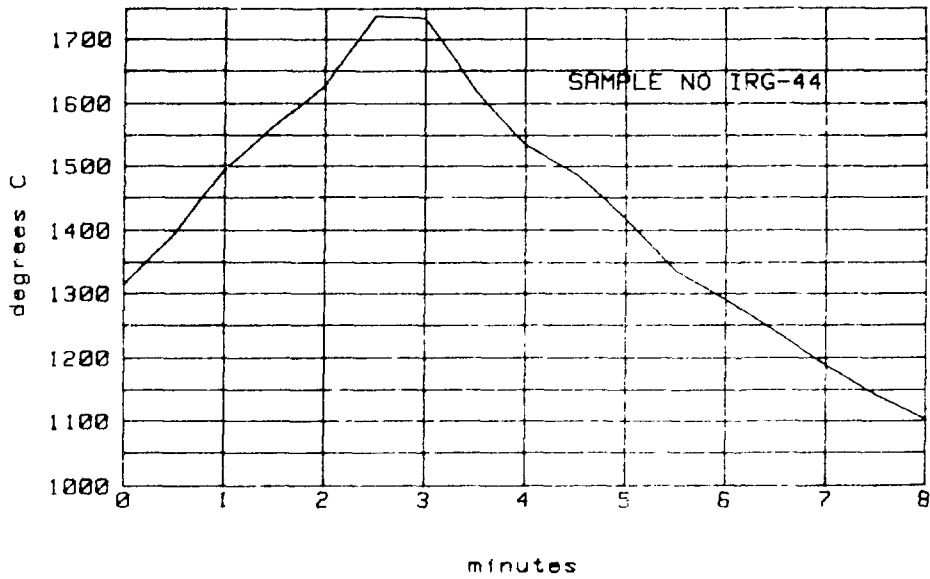


Fig. I-4.

Reentry profile used for GPHS impact DGCI-27-R1-23 with capsule IRG-44. The impact temperature was 965°C.

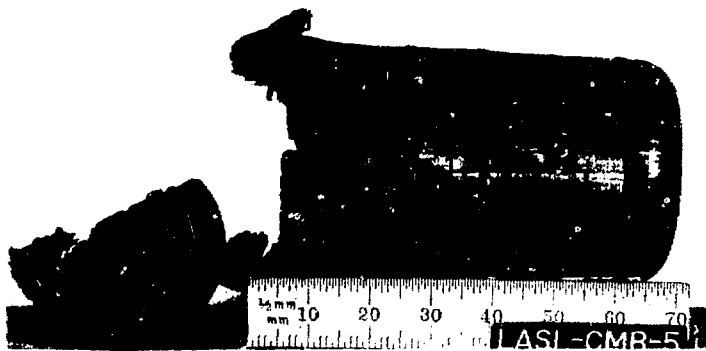


Fig. I-5.

DGCI-27-R1-23: Double 27° impact with leading capsule IRG-44 in DGIS-12. Impacted at 27°, 58 m/s, and 965°C.



a)



b)



c)



d)



e)

Fig. I-6.
DGGI-27-R1-23: Capsule IRG-44 (Ir-0.3 W) was the leading capsule in this double 27 impact at 58 m/s and 965 C. a) Front view with impact face on top, b) side profile, c) side profile at 90°, d) impact face, and e) back end.

TABLE I-1

GPHS IMPACTS OF 62.5 W Ir-0.3% W/PuO₂ CAPSULES

| Capsule Number | Type of Impact | Capsule Location | Heat Treat and grains/thick. in at 1500°C | Impact Shell | Impact Temp. (°C) | Impact Velocity (m/s) | Max. Diam. Strain (%) | Diagonal or Diagonal Shrinkage (%) | Max. Positive Weld Strain | Local Strains | | |
|----------------|-----------------------|----------------------------|---|-------------------------------|-------------------|-----------------------|-----------------------|------------------------------------|---------------------------|--------------------|---------|------------------------|
| | | | | | | | | | | Shear or Thickness | Bending | Fracture |
| IRG-10 | Double 90° | Leading Caps. | - | DGIS-2 | 965 | 58 | 2.5 | -1.3 | 4% | a | b | No |
| IRG-30 | Double 0° | Leading Caps. | - | DGIS-6 | 965 | 58.1 | 5.5 | -6.6 (Height) | 4.3 | 46% | 16 | No |
| IRG-17 | Mockup Module 45° | Leading GIS. Leading Caps. | - | DGIS-2 | 965 | 58 | 9.0 | -12.4 | 8.9 | 40 | 15 | No |
| IRG-14 | Double 27° | Leading Caps. | - | DGIS-2 | 965 | 57.2 | 3.5 | -8.9 | 3.5 | a | 9 | No |
| IRG-15 | - | - | - | DGIS-2 | 965 | 56 | 9.1 | -7.4 | 8.1 | 75 | 20 | No |
| IRG-23 | - | - | - | DGIS-4 | 965 | 57.9 | 5.8 | -7.9 | 5.8 | a | 15 | No |
| IRG-24 | - | - | - | DGIS-5 | 965 | 58 | 5.1 | -8.3 | 5.1 | a | 8 | No |
| IRG-27 | - | - | - | DGIS-6 | 900 | 58 | 4.3 | -7.7 | 4.3 | 14 | 9 | No |
| IRG-43 | - | - | - | DGIS-6 | 800 | 58 | 8.8 | -9.2 | 8.8 | a | 16 | No |
| IRG-42 | - | - | 1h at 1650°C | DGIS-6 | 800 | 58 | 4.1 | -6.8 | 4.1 | a | 10 | No |
| IRG-19 | Module 45° Short Edge | Leading GIS. Leading Caps. | 1h at 1500°C 25-28 | DGIS-3 | 965 | 57.6 | 4.6 | -9.3 | 4.1 | a | 13 | 2 possible microcracks |
| IRG-20 | - | Trailing GIS Leading Caps. | - | DGIS-3 | 965 | 57.6 | 3.0 | -2.4 | 2.8 | a | <5 | No |
| IRG-29 | Module 45° Corner | Leading GIS. Leading Caps. | - | DGIS-6 | 965 | 58 | 6.7 | -7.1 | 5.0 | a | 16 | No |
| IRG-28 | - | Trailing GIS Leading Caps. | - | DGIS-6 | 965 | 58 | 0.8 | -1.7 | 0.8 | a | <5 | No |
| IRG-34 | Double 27° | Leading Caps. | 1h at 1800°C 5-6 | DGIS-6 | 800 | 58.0 | >10 | Could not be measured | <10 | b | b | Yes catastrophic |
| IRG-40 | Double 27° | Leading Caps. | 20h at 1500°C 13-14 | DGIS-7 | 1075 | 78.9 | 17 | -11.7 | 17 | >90 | 20 | Push-through failures |
| IRG-37 | Double 27° | Leading Caps. | 20h at 1500°C 13-14 | DGIS-8 Std. thick. | 1075 | 58 | 10.4 | -14.6 | 10.4 | a | 6 | No |
| IRG-50 | Double 90° | - | - | DGIS-9 Std. thick. | 1075 | 58 | +12.6 (length) | -13.0 | 9.7 | a | 11 | No |
| IRG-51 | Double 0° | - | - | DGIS-10 Std. thick. 10% holes | 965 | 56 | 6.0 | -7.3 | b | a | b | No |
| IRG-25 | Double 0° w/12M | - | 1h at 1500°C 25-28 | Half GIS No holes | 965 | 58 | 5.7 | -2.5 | b | b | b | No |
| IRG-26 | - | - | - | Half GIS 40% holes | 965 | 58 | 4.4 | -2.7 | b | b | b | No |
| IRG-44 | Double 27° | Leading Caps. | 20h at 1500°C + 1700°C | DGIS-12 Bulk liner on OD | 965 | 58 | 10.6 | -6.4 | 9.2 | b | b | No |
| IRG-48 | Double 0° | - | 20h at 1500°C + 1700°C Reentry | DGIS-13 CBCF + PG Ring | 965 | 57.7 | 7.3 | -8.0 (Height) | 6.9 | severe | severe | Yes |
| IRG-49 | Double 27° | - | - | - | 965 | 58.1 | 10.9 | -13.9 | 11.4 | severe | severe | Yes |

a - not significant
b - not measured

The grain size of the Ir-0.3% W capsule has not been measured and we cannot yet compare the observed deformation to predicted failure levels. It does appear at this time that the Ir-0.3% W clad can survive impact at 965°C even after a reentry pulse of 1740°C.

2. DGIS-0-R2-23. This was a double impact at 0° (end-on). The GIS was of design DGIS-13, shown in Fig. I-7. The side wall, end walls, and web are of the same thickness as DGIS-12. The impact end was enclosed in a cup of CBCF, 2.03 mm thick with a PG standoff ring, 2.15 mm thick. The assembly prior to impact is shown in Fig. I-8. The GIS was machined from FWP 3D carbon-carbon composite. All graphitic components were outgassed for 1 h at 1800°C. The leading capsule was IRG-48, Ir-0.3% W. It was annealed for 20 h at 1500°C before

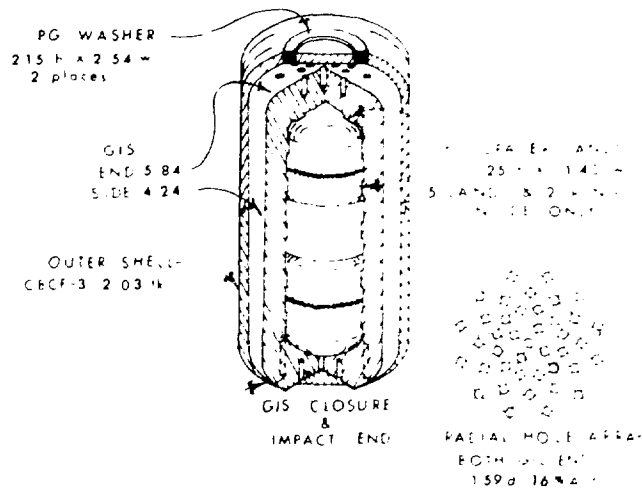


Fig. I-7.
Schematic drawing of DGIS-13 (DGCI-0-R2-23 and DGCI-27-R3-23) showing details of graphite impact assembly.



Fig. I-8.
DGIS-13 used in DGCI-0-R2-23 and DGCI-27-R3-23. The FWPF GIS contained standoff rings on the interior cylindrical and end walls. The CBCF is shown at the top right as a cup with a center hole. The disc at the bottom right consisted of a ring of PG (2.15 mm thick) and a center disk of CBCF (2.03 mm thick).

welding and subjected to a thermal reentry pulse with the $^{238}\text{PuO}_2$ pellet after welding. The maximum temperature of the reentry profile was 1700°C with 6 minutes above 1300°C . The temperature profile is shown in Fig. I-9. The capsule contained a $^{238}\text{PuO}_2$ fuel pellet of 82.8% theoretical density. The trailing capsule was GCI-27R, a reused Pt-3008 capsule containing a UO_2 pellet. The assembly was impacted at 0° , 57.7 m/s, and 965°C .

The remains of the GIS after impact are shown in Fig. I-10. The impact end crushed and fractured circumferentially. The center web crushed substantially. The leading capsule deformed substantially and failed at a fuel push-through on the impact face. The impacted capsule is shown in Fig. I-11. The diametral strain was 7.3% which is greater than the 5.5% experienced by IRG-30 which was impacted with two 0.76-mm-thick PG liners inside a GIS with the same GIS end-wall thickness. The push-through in capsule IRG-48 was severe, as shown in Fig. I-11. The impact face was also slightly tilted, as shown, suggesting that the PG standoff ring was not crushed evenly. We are currently evaluating the local strain levels and will determine the grain size of the Ir-0.3% W clad.

3. DGCI-27-R3-23. This was a double impact at 27° . The GIS and CBCF outer liner with PG standoff ring were identical to those used in DGCI-27-R2-23. The leading capsule was IRG-49 with a heat treatment and reentry profile identical to that used for IRG-48. It contained a $^{238}\text{PuO}_2$ fuel pellet of 86.3% theoretical density. The trailing capsule was GCI-25R, a reused Pt-3008 capsule containing a UO_2 pellet. The assembly was impacted at 27° , 58.1 m/s and 965°C .

The remains of the GIS after impact are shown in Fig. I-12. The impact corner crushed, the impact end tore off and the GIS split longitudinally. The

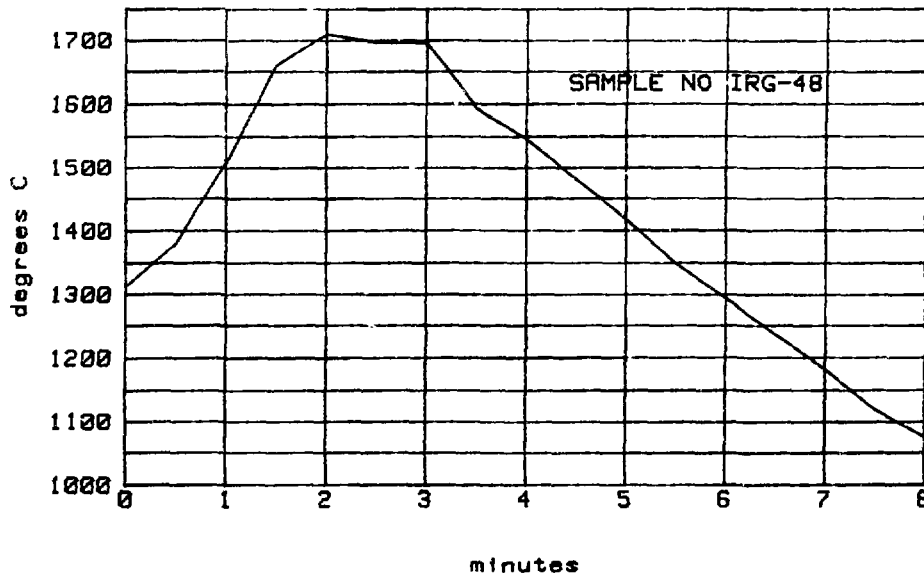


Fig. I-9.

Reentry profile used for GPHS impact DGIS-0-R2-23 with capsule IRG-48. The impact was at 965°C .

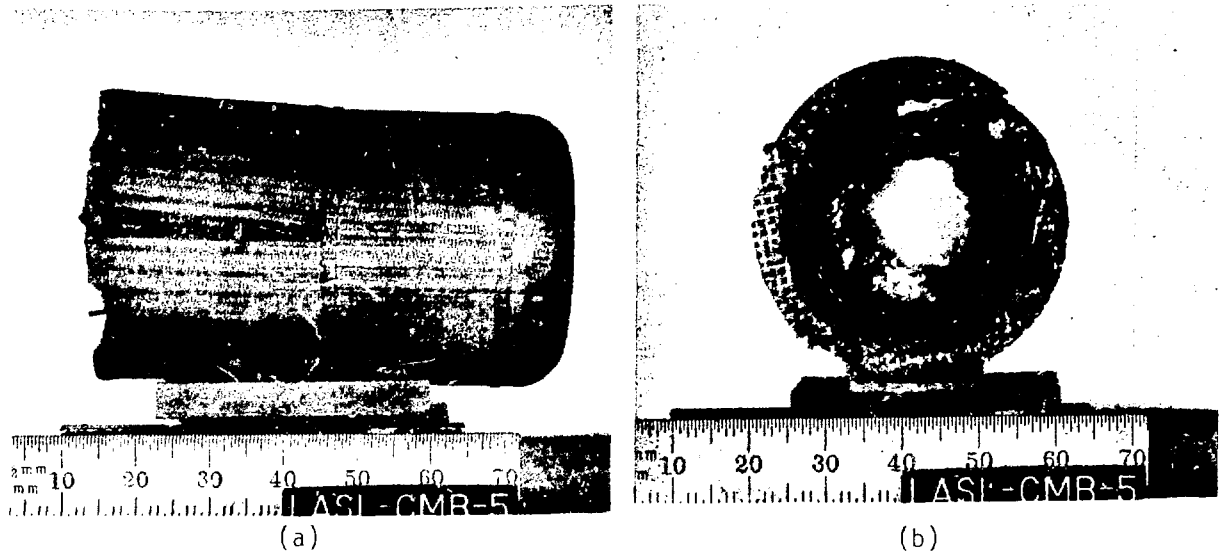


Fig. I-10.

DGCI-0-R2-23. Double 0° impact with leading capsule IRG-48 in DGIC-13 with CBCF cup and PG standoff. Impacted at 0° , 57.7 m/s, and 965°C . (a) Side profile of double GIS and (b) view of center web.

leading capsule deformed substantially and failed at a fuel push-through on the impact face. The impacted capsule is shown in Fig. I-13. The maximum diagonal shrinkage was considerably greater than that observed in IRG-44, which was impacted with a bulk exterior liner instead of the CBCF-PG combination. The maximum diametral strains were similar. The push-throughs in the impact face were severe and caused failure of the Ir-0.3% W clad. Local strain measurements and grain size determinations are yet to be made.

A complete explanation of the two failures involving the CBCF-PG outer liner must await further analysis of local strains, grain size, and grain boundary chemistry measurements. It does appear certain at this time that the CBCF-PG liner with the inherent mismatch in density and strength between the CBCF and PG aggravates the push-through problem.

C. Fuel Development

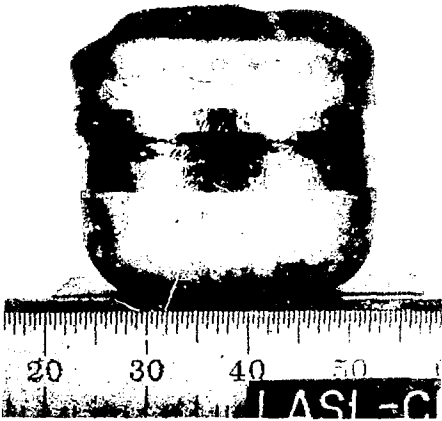
Two $^{238}\text{PuO}_2$ pellets (62.5-W size) were encapsulated in Ir-0.3% W for impact testing. Each capsule was cleaned by successive immersions in 3 acid solutions (5/2/2 of $\text{H}_2\text{O}/\text{HNO}_2/\text{HF}$) followed by immersions in distilled water and ethanol. The capsules were cleaned to fewer than 50 counts per second per capsule (swipe) and then transferred to the test area.

D. Clad Development

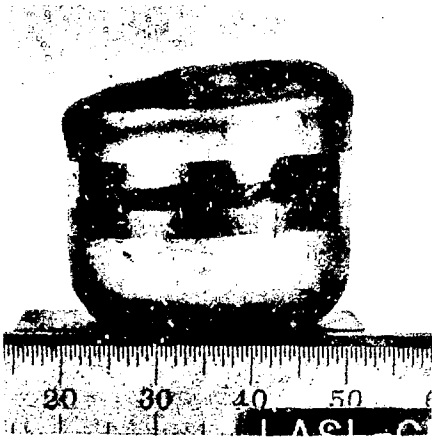
The accumulated exposure time of the Pt-3008 vs $^{238}\text{PuO}_2$ (1100°C , vacuum) was 19 909 h as of December 1, 1979.

E. Vent Testing

Monitoring of the exposure of a Mound Facility (MF) GPHS vent to vapor deposited plutonia was continued this month.



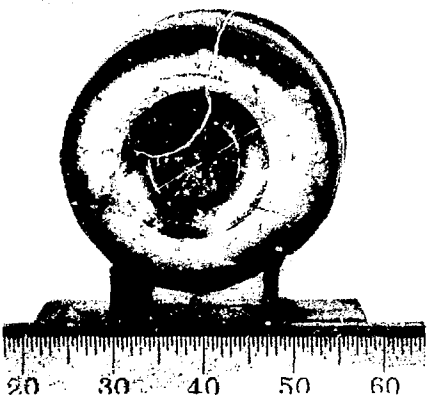
(a)



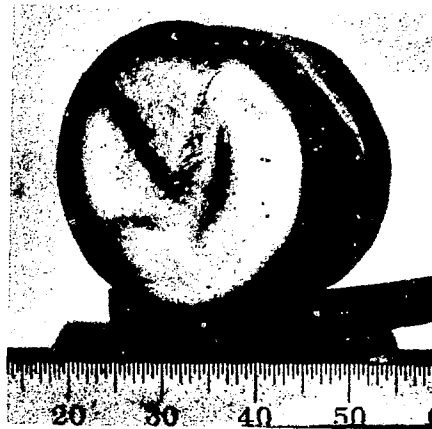
(b)



(d)



(c)



(e)

Fig. I-11.
DGC1-0-R2-23: Capsule IRG-48 (Ir-0.3% W) was the leading capsule in this double impact at 57.7 m/s and 965°C. (a) and (b) side profiles, (c) back end, (d) impact face, and (e) oblique view of impact face showing failure at fuel push-through.

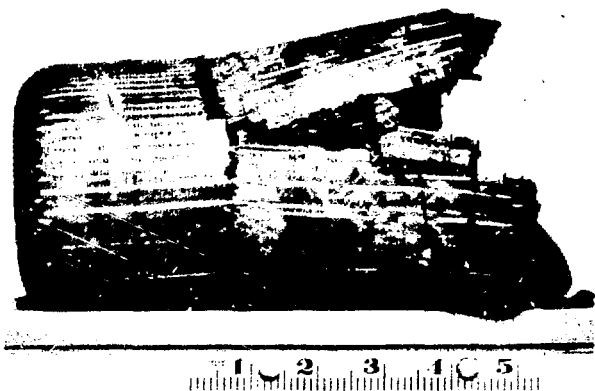


Fig. I-12.
DGCI-27-R3-23. Double 27° impact with
leading capsule IRG-49 in DGIS-13 with
CBCF and PG standoff. Impacted at 27°,
58.1 m/s, and 965°C.

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 2349 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, and 185 days in its sixth winter before being programmed for its seventh summer. The averages of the plutonium contents from liquid scintillation analysis and the volume measurements of the water that percolated through the soil for four 32-mm summer rains decreased this month (Table II-2). The average water volume was 3.4 l, compared with 3.9 l last month, while the average plutonium content changed to 0.99 from 1.05 μCi . The plutonium collected in the dehumidifier condensate changed from 0.3 to 0.4 $\mu\text{Ci}/\text{wk}$.

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 164 days. The averages of the measured volumes and plutonium contents for four 32-mm rains are higher than those for last month (Table II-3). The average volume increased from 2.6 to 3.0 l, and the average plutonium content from 0.52 to 0.59 μCi . The collection rate of plutonium by the dehumidifier condensate decreased from 0.13 to 0.05.

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 chunks on sand; arid climate | 2-25-75 |

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

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> |
|------------|-------------|-------------------------------|---------------------------|
| 324 | 10/22 | 4.2 | 1.39 |
| 325 | 10/29 | 4.2 | 1.61 |
| 326 | 11/5 | 3.6 | 0.66 |
| 327 | 11/13 | 1.5 | 0.31 |
| Average | | 3.4 | 0.99 |

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> |
|------------|-------------|-------------------------------|--|
| 316 | 10/22 | 3.0 | 0.66 |
| 317 | 10/29 | 2.9 | 0.61 |
| 318 | 11/5 | 2.8 | 0.78 |
| 319 | 11/13 | 3.4 | 0.72 |
| Average | | 3.0 | 0.69 |

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, and now 164 days into its fifth arid summer. Liquid scintillation analysis and volume measurement of one 81-mm rain gave results of 0.06 μ Ci of plutonium in 4.2 ℓ, compared with 0.03 μ Ci in 2.7 ℓ last month. The collection rate for plutonium in the dehumidifier condensates was 0.27 μ Ci/wk, compared with 0.13 μ Ci/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 water that percolated through the soil for one 81-mm rain gave results of 0.012 μ Ci of plutonium in 3.5 ℓ of water, higher in plutonium than the 0.008 μ Ci of plutonium in 2.0 ℓ of water for the last rain. The dehumidifier condensates collected 0.01 μ Ci/wk of plutonium lower than the 0.33 μ Ci 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 humid winter 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, and now 164 days into the fifth summer. The soil in this chamber is divided into two compartments. A thin cylindrical 45-cm-diam 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 analyses and volume measurements of rainwater from four 34-mm rains, the average values for the outer compartment are almost unchanged (Table II-4). In the inner compartment, the average volume was 1.13 ℓ and the average plutonium content was 0.095 μ Ci, compared with 0.8 ℓ and 0.114 μ Ci last month. In the outer compartment, the average volume decreased slightly, from 10.5 to 9.4 ℓ, while the plutonium content increased from 0.09 to 0.10 μ Ci. The rate of plutonium collection by the dehumidifier condensate increased from 0.5 to 0.9 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

TABLE II-4
 PLUTONIUM IN PERCOLATED RAINWATER
 (LARGER PIECES FROM MHFT-5Q ON COMPARTMENTED SOIL)

| No. | Date | Inner | | Outer | |
|---------|-------|------------|-------------------|------------|-------------------|
| | | Vol (ℓ) | Pu (μ Ci) | Vol (ℓ) | Pu (μ Ci) |
| 233 | 10/22 | 1.65 | 0.019 | 9.0 | 0.09 |
| 234 | 10/29 | 1.60 | 0.175 | 9.8 | 0.12 |
| 235 | 11/5 | 0.60 | 0.037 | 9.9 | 0.10 |
| 236 | 11/13 | 0.65 | 0.150 | 8.8 | 0.10 |
| Average | | 1.13 | 0.095 | 9.4 | 0.10 |

and are also on loam in a similarly compartmented tray. Liquid scintillation analyses and measured volumes of percolated rainwater from four 27-mm rains show increases in the average results for the inner compartment, while the outer compartment averages changed very little (Table II-5). The averages for the outer compartment volumes and plutonium contents are 5.7 ℓ and 0.138 μ Ci, respectively, compared with 5.4 ℓ and 0.15 μ Ci of plutonium last month. The inner compartment averages are 2.2 ℓ and 0.056 μ Ci compared with 1.4 ℓ and 0.029 μ Ci last month. The collection rate for plutonium in the dehumidifier condensate remained at 0.2 μ Ci/wk.

The experiment in which a 254-g sphere of plutonium dioxide clad with iridium and encased in a 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.

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

| No. | Date | Inner | | Outer | |
|---------|-------|------------|-------------------|------------|-------------------|
| | | Vol (ℓ) | Pu (μ Ci) | Vol (ℓ) | Pu (μ Ci) |
| 234 | 10/22 | 2.3 | 0.048 | 5.7 | 0.136 |
| 235 | 10/29 | 2.6 | 0.067 | 5.2 | 0.109 |
| 236 | 11/5 | 1.5 | 0.054 | 6.6 | 0.172 |
| 237 | 11/13 | 2.4 | 0.054 | 5.2 | 0.133 |
| Average | | 2.2 | 0.056 | 5.7 | 0.138 |

TABLE II-6

Pu TRANSPORT FROM PuO₂ PARTICLES

| Column | Elution Duration | | | |
|--------|------------------|------------------|------------------|------------------|
| | 983 days | | 1108 days | |
| | Total Vol (ℓ) | Total Pu (ng) | Total Vol (ℓ) | Total Pu (ng) |
| 7 | 92.0 | 29.3 | 80.1 | 30.8 |
| 8 | 62.4 | 25.7 | 69.9 | 27.6 |
| 9 | 32.9 | 38.2 | 37.3 | 40.4 |

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 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- μ m 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 1108 days. Measurements made after 983 and 1108 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.

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

| Experiment Number | PuO ₂ (mg) | Per Cent Dissolved | Dissolution Rate | |
|-------------------|-----------------------|--------------------|-----------------------|-----------------------|
| | | | (g/s) | (g/s-g) |
| A1 | 2.02 | 4.5 | 7.3x10 ⁻¹³ | 3.6x10 ⁻¹⁰ |
| A2 | 2.05 | 4.2 | 6.9x10 ⁻¹³ | 3.3x10 ⁻¹⁰ |
| A3 | 10.19 | 4.2 | 3.4x10 ⁻¹² | 3.4x10 ⁻¹⁰ |
| A4 | 9.90 | 4.2 | 3.3x10 ⁻¹² | 3.3x10 ⁻¹⁰ |
| B1 | 2.02 | 7.8 | 1.3x10 ⁻¹² | 6.3x10 ⁻¹⁰ |
| B2 | 2.03 | 9.1 | 1.5x10 ⁻¹² | 7.3x10 ⁻¹⁰ |
| B3 | 10.25 | 8.0 | 6.6x10 ⁻¹² | 6.5x10 ⁻¹⁰ |
| B4 | 10.45 | 7.7 | 6.5x10 ⁻¹² | 6.2x10 ⁻¹⁰ |
| C1 | 2.23 | 17.5 | 3.1x10 ⁻¹² | 1.4x10 ⁻⁹ |
| C2 | 2.31 | 17.0 | 3.2x10 ⁻¹² | 1.4x10 ⁻⁹ |
| C3 | 9.89 | 16.7 | 1.3x10 ⁻¹¹ | 1.3x10 ⁻⁹ |
| C4 | 10.67 | 16.4 | 1.4x10 ⁻¹¹ | 1.3x10 ⁻⁹ |

B. Aquatic Environments

1. Aqueous Release Rates As A Function of PuO₂ Particle Size. Three sets of size ²³⁸PuO₂ particles, each having a different size distribution, are 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 1450 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) exhibit no discernible discrepancies. Each plot shows increasing dissolution of $^{238}\text{PuO}_2$ with C-1 > B-1 > A-1. Integral dissolution rate plots show 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 $\sim 2 \times 10^7$ 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 "large" 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 Pu^{+4} , 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 1853 days (Table II-8). Its release rate is $14 \text{ nCi/m}^2\text{-s}$, up 8% from last month.

The tidal simulation aquarium, using the 19-W source HPZ-186-4, has been under way for 744 days. The source is on sand in a stainless steel tray that is lowered into the water and subsequently raised above the surface twice every 24 h. 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 $60 \text{ nCi/m}^2\text{-s}$, up 7% from last month's value.

TABLE II-8

SUMMARY OF
Pu RELEASE RATE FROM PPO IN AQUATIC ENVIRONMENTS

| <u>Sample</u> | <u>Power (W)</u> | <u>Immersion (days)</u> | <u>Water</u> | <u>Temperature (C)</u> | <u>Release Rate (nCi/m²-s)</u> |
|---------------|------------------|-------------------------|--------------|------------------------|---|
| HPZ-60-2 | 2.5 | 2060 | Fresh | 10 | 130 |
| HPZ-111-1 | 25 | 1759 | Fresh | 10 | 400 |
| HPZ-59-4 | 25 | 1853 | Sea | 10 | 14 |
| HPZ-174 | 18 | 748 | Sea | 10 | 7 |
| HPZ-186-4 | 19 | 744 | Sea (Tidal) | 10 | 60 |

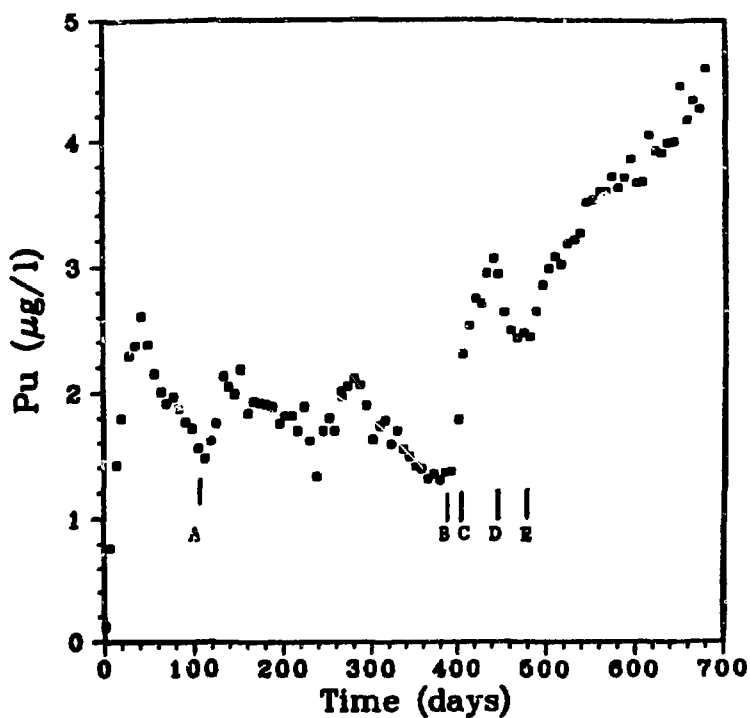


Fig. II-1.
Plutonium concentration in the tidal simulation experiment.

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 μg to 26.2 μg 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 Pu on or in the sand.

The 18-W pellet HPZ-174 has been immersed in seawater at 10°C for 748 days. The release rate is $7 \text{ nCi/m}^2\text{-s}$, unchanged in the last 2 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 2060 days of immersion is $130 \text{ nCi/m}^2\text{-s}$, down 13% from last month's value. This is the second highest release rate exhibited in this set of PPO pellets.

The fastest release rate is shown by the 25-W pellet, HPZ-111-1, immersed for 1759 days at 10°C . Its average release rate is $400 \text{ nCi/m}^2\text{-s}$, down 3% from the value observed last month. The highest release 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 rate 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.

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- μm 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 μg which represents a release rate per unit surface area of $22.1 \text{ pg/m}^2\text{-s}$. This number can be compared to the release rates seen in the aquatic environment experiments which vary from 0.59 to 36 $\text{ng/m}^2\text{-s}$. The analysis of the second part of the experiment in which the source was submerged indicated the release of 49.3 μg . Calculations based on the same surface area and a 30-minute submergence yields a release rate of $16.8 \text{ }\mu\text{g/m}^2\text{-s}$.

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.

Water Samplers. Two water samplers 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, and their contents are currently being processed for plutonium analysis.

C. Analytical Studies of PuO₂-Soil Agglomeration
(Purchase Order LP9-3589K; LFE Environmental)

Marcel Nathans of LFE Environmental is now reporting progress on a bimonthly basis. The next progress report should be received next month.

Cumulative costs on this purchase order through October 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.

III. SYSTEMS SUPPORT

A. Stirling Isotope Power System

The accumulated exposure time for the 800°C test assembly was 16 714 h as of December 1, 1979. Failure of the furnace temperature control system resulted in a temperature excursion to 1095°C for a period of 4.5 h.

B. Improved Multi-hundred Watt (MHW) Tests

The status of the test series is given in Table III-1. The exposure of assembly ID-2 has been terminated. The helium signal produced by assembly ID-1 indicated a vented condition throughout this reporting period.

TABLE III-1
STATUS OF THE IMPROVED MHW TESTS

| <u>Ref. No.</u> | <u>Assembly Id.</u> | <u>Scheduled Aging</u> | <u>Status as of 12/01/79</u> |
|-----------------|---------------------|--------------------------------------|------------------------------|
| IC-1 | Pt-3008/F1 | 30 days at 1200°C (PICS) + impact | Completed |
| IC-2 | Pt-3008/F3 | 30 days at 1200°C (PICS) | Completed |
| ID-1 | MHFT-61 | 6-12 months at 1330°C (PICS) | 8178 h exposure |
| ID-2 | Pt-3008/F2 | 6-12 months at 1200°C (PICS) | 8974 h exposure |

C. Safety Verification Tests (Savannah River (SR) Fuel Spheres)

The catastrophic impact failure of the iridium shell of MHFT-65 at a fuel push-through was reported in the two previous monthly reports. This month we continued the examination.

1. Grain Boundary Chemistry. We report here the results of our investigation of grain boundary chemistry of MHFT-65 and several other iridium shells by Auger Electron Spectroscopy (AES). Six specimens were examined from MHFT-65, three from each hemisphere. The specimens were thoroughly scrubbed and cleaned to decontaminate them as much as possible. The results of the AES investigation are listed in Table III-2. AES spectra were taken near the inside (in contact with $^{238}\text{PuO}_2$ fuel), center, and outside. The spot size of the beam was $\sim 150 \mu\text{m}$. The thorium level at the grain boundaries appears to be low for DOP-26 Ir-0.3% W. Phosphorus was detected in very large concentrations and sulfur was found in some of the fractured cross sections. The sulfur peaks were also very intense, indicating very high concentrations of sulfur at the grain boundaries. We were not able to establish a specific trend of thorium and phosphorus concentration from the inside to the outside of the post-impact containment shell (PICS). Table III-3 lists the averages of all readings at the inside, center, and outside. These results suggest that the center of the specimens are higher in thorium and lower in phosphorus.

Oak Ridge National Laboratory (ORNL) has shown that thorium in the grain boundaries can be depleted by annealing in an oxidizing atmosphere, and that the thorium levels will increase again if this treatment is followed by a vacuum anneal. We annealed a piece of MHFT-65 in vacuum for 1 h at 1500°C and examined the grain boundary chemistry. The AES results are listed in Table III-4 and summarized in Table III-3. We found that the thorium level increased from an average of 0.29 to an average of 0.54 and phosphorus decreased from 0.56 to 0.44.

The grain boundary chemistries of three additional iridium shells were also determined by AES. The results are listed in Table III-5 and summarized in Table III-6. The thorium levels in all PICS were lower than those typically found in DOP-26. The recent investigation of lots L-1 and L-2 gave an average thorium level of 0.54. Table III-6 shows that the phosphorus level in MHFT-65 is abnormally high. We also find that MHFT-64 and MHFT-65 contain very high sulfur levels. A comparison to other MHW shells that were impacted and analyzed is presented in Table III-7. Although phosphorus has been found in many of the doped Ir-0.3% W PICS, the level in MHFT-65 is unusually high. Sulfur was found only in MHFT-64 and -65 both of which contained SR fuel. Table III-7 shows that the grain size for DOP-26 PICS is typically smaller (more grains/tickness) than HD and undoped Ir-0.3% W PICS for an equivalent heat treatment. In all cases the grain sizes of the PICS are larger than those predicted by ORNL for vacuum heat treatment.

These results suggest that MHFT-65 possessed an abnormal grain boundary chemistry (high levels of phosphorus and sulfur). We cannot, however, blame the catastrophic failure of MHFT-65 on the phosphorus and sulfur contamination. It is quite likely that the large deformations imposed by the severe fuel push-through in MHFT-65 would have fractured the iridium even if it were not contaminated. We are currently conducting some biaxial punch tests to evaluate the influence of phosphorus on impact ductility. These tests should aid us in the interpretation of the MHFT-65 impact failure.

TABLE III-2
AES RESULTS FOR MHFT-65

| Specimen | Treatment | Location | Auger Intensity Ratios | | | | | |
|------------|---|---------------|--|--|--|--|--|--|
| | | | Th ₆₅ / _r 229 | P ₁₂₀ / _r 229 | C ₂₇₀ / _r 229 | O ₅₁₀ / _r 229 | Si ₉₂ / _r 229 | S ₁₅₀ / _r 229 |
| MHFT-65-1 | DOP-26 Iridium 6 month age in FSA 1210° C GIS | Edge | 0.203 | 0.662 | 2.46 | 1.32 | 0 | 0 |
| | | Inside | | | | | | |
| | | 150µm area | | | | | | |
| MHFT-65-2 | "-" | Center | 0.307 | 0.487 | 1.04 | 0.85 | 0 | 0 |
| | | Edge | | | | | | |
| | | Outside | 0.025 | 1.407 | 1.70 | 1.02 | 0 | 0 |
| | | Edge | | | | | | |
| | | Inside | 0 | 1.36 | 1.67 | 2.17 | 0 | 0 |
| | | Center | 0.487 | 0.63 | 0.41 | 0.73 | 0 | 0 |
| MHFT-65-A2 | "-" | Edge | | | | | | |
| | | Outside | 0.476 | 0.44 | 0.48 | 0.79 | 0.12 | 0 |
| | | Edge | | | | | | |
| | | Inside | 0.153 | 0.328 | 1.00 | 0.50 | 0 | 0.85 |
| | | Center | 0.362 | 0.142 | 0.85 | 1.06 | Trace | 0.67 |
| | | Edge | | | | | | |
| MHFT-65-A3 | "-" | Outside | 0.372 | 0.585 | 1.54 | 1.38 | Trace | 0.81 |
| | | Edge | | | | | | |
| | | Inside | 0.022 | 0.283 | 0.78 | 0.89 | 0 | 2.36 |
| | | Center | 0.271 | 0 | 1.03 | 1.39 | 0 | 2.90 |
| | | Edge | | | | | | |
| | | Outside | 0.177 | 0 | 0.95 | 1.32 | 0 | 3.83 |
| MHFT-65-B2 | "-" | Edge | | | | | | |
| | | Inside | 0.526 | 0.702 | 0.39 | 0.64 | 0 | 0 |
| | | Center | 0.618 | 0.836 | 0.32 | 0.79 | 0 | 0 |
| | | Edge | | | | | | |
| | | Outside | 0.137 | 1.103 | 0.68 | 0.75 | 0 | 0 |
| | | Edge | | | | | | |
| MHFT-65-B3 | "-" | Inside | 0.113 | 0.356 | 0.76 | 0.56 | 0 | 0.95 |
| | | Center | 0.374 | 0.281 | 0.44 | 0.84 | 0 | 0 |
| | | Edge | | | | | | |
| | | Outside | 0.530 | 0.455 | 0.73 | 0.96 | 0 | 0 |
| | | Edge | | | | | | |
| | | Inside | 0.113 | 0.356 | 0.76 | 0.56 | 0 | 0.95 |

TABLE III-3
SUMMARY OF AES RESULTS ON MHFT-65

| <u>MHFT-65 (6 Months At 1210°C GIS)</u> | <u>Th₆₅/Ir₂₂₉</u> | <u>P₁₂₀/Ir₂₂₉</u> | <u>C₂₇₀/Ir₂₂₉</u> | <u>O₅₁₀/Ir₂₂₉</u> |
|--|---|---|---|---|
| Inside (One of Six Specimens) | 0.17 | 0.62 | 1.18 | 1.01 |
| Center - " - | 0.40 | 0.40 | 0.68 | 0.94 |
| Outside - " - | 0.29 | 0.67 | 1.01 | 1.04 |
| MHFT-65 (6 Months + 1 H at 1500°C Reanneal) | 0.540 | 0.44 | 0.54 | 0.94 |

TABLE III-4
AES RESULTS OF MHFT-65 REANNEALED IN VACUUM

| Specimen | Treatment | Location | Auger Intensity Ratios | | | | | |
|----------------|--|----------------|---|---|---|---|---|---|
| | | | <u>Th₆₅/Ir₂₂₉</u> | <u>P₁₂₀/Ir₂₂₉</u> | <u>C₂₇₀/Ir₂₂₉</u> | <u>O₅₁₀/Ir₂₂₉</u> | <u>Si₉₂/Ir₂₂₉</u> | <u>S₁₅₀/Ir₂₂₉</u> |
| MHFT-65 A4 | DOP-26 Iridium 6 month age in FSA at 1210°C GIS + 1h at 1500°C C in vacuum | Edge | 0.120 | 0.280 | 0.55 | 1.04 | 0 | 1.07 |
| | | Inside | | | | | | |
| | | 150.0m area | 0.638 | 0.215 | 0.35 | 1.23 | 0 | 0 |
| | | Center | 0.535 | 0.208 | 0.63 | 0.90 | Trace | 0 |
| MHFT-65- B4 | "- | Edge | | | | | | |
| | | Inside | 0.441 | 0.591 | 0.63 | 0.80 | 0 | 0 |
| | | Center | 0.823 | 0.652 | 0.43 | 0.76 | 0 | 0 |
| | | Edge | | | | | | |
| | | Outside | 0.688 | 0.671 | 0.65 | 0.88 | 0 | 0 |

TABLE III-5
AES RESULTS FOR MHFT-62, -64 AND -66

| Specimen | Treatment | Location | Auger Intensity Ratios | | | | | |
|-----------|--|----------|------------------------|----------------------|----------------------|----------------------|----------------------|----------------------|
| | | | Th_{65}/I_r 229 | P_{120}/I_r 229 | C_{270}/I_r 229 | O_{510}/I_r 229 | Si_{92}/I_r 229 | S_{150}/I_r 229 |
| MHFT-62-1 | HDR Iridium Aged 30 days in FSA at 1210°C GIS | Edge | | | | | | |
| | | Inside | 0.787 | 0 | 1.13 | 1.13 | 0 | 0 |
| | | Center | 0.060 | 0 | 0.34 | 1.13 | 0 | 0 |
| MHFT-62-2 | "- | Edge | | | | | | |
| | | Inside | 0 | 0 | 0.52 | 0.81 | 0 | 0 |
| | | Center | 0.480 | 0 | 0.22 | 0.62 | 0 | 0 |
| MHFT-66-1 | HD Iridium Aged 30 days in FSA at 1210°C GIS | Edge | | | | | | |
| | | Inside | 0.221 | 0 | 0.49 | 0.69 | 0 | 0 |
| | | Center | 0.638 | 0.162 | 0.19 | 0.38 | 0 | 0 |
| MHFT-66-2 | "- | Edge | | | | | | |
| | | Inside | 0.379 | 0 | 0.75 | 1.05 | 0 | 0 |
| | | Center | 0.528 | 0 | 0.52 | 1.47 | 0 | 0 |
| MHFT-64-1 | DOP 26 Iridium Aged 30 days in FSA at 1210°C GIS | Edge | | | | | | |
| | | Inside | 0.195 | 0 | 1.28 | 1.05 | 0 | 0.95 |
| | | Center | 0.371 | 0 | 1.10 | 1.62 | 0 | 0.88 |
| MHFT-64-2 | "- | Edge | | | | | | |
| | | Inside | 0.486 | 0.135 | 0.68 | 0.89 | 0 | 0 |
| | | Center | 0.393 | 0 | 0.36 | 1.12 | 0 | |
| | | Edge | | | | | | |
| | | Outside | 0.218 | 0 | 0.51 | 0.91 | 0 | 0 |

TABLE III-6
SUMMARY OF AES RESULTS OF RECENT PICS

| <u>PICS #</u> | <u>Type of Ir</u> | <u>Treatment</u> | <u>Th₆₅/Ir₂₂₉</u> | <u>P₁₂₀/Ir₂₂₉</u> | <u>Si₁₅₀/Ir₂₂₉</u> | <u>C₂₇₀/Ir₂₂₉</u> | <u>O₅₁₀/Ir₂₂₉</u> |
|---------------|-------------------|---------------------------|---|---|--|---|---|
| MHFT-62 | HDR | 30 Days At 1210°C GIS | 0.30 | 0 | 0 | 0.82 | 0.86 |
| -64 | DOP-26 | 30 Days At 1210°C GIS | 0.34 | 0.02 | 0.87 | 0.84 | 1.14 |
| -65 | DOP-26 | 6 Months At 1210°C GIS | 0.29 | 0.56 | 0.69 | 0.96 | 1.00 |
| -66 | HD | 30 Days At 1210°C GIS | 0.46 | 0.09 | 0 | 0.48 | 0.89 |

TABLE III-7
SUMMARY OF AES RESULTS FOR LIVE (PuO₂) FSAs THAT WERE IMPACTED

| <u>Specimen</u> | <u>Ir Type</u> | <u>Fuel</u> | <u>Treatment</u> | <u># grains/ thickness</u> | <u>Auger Intensity Ratios</u> | | | | <u>Impact Failure</u> |
|-----------------|----------------|-------------|------------------------------------|--------------------------------|---------------------------------|------------------|-------------------|---------------|---|
| | | | | | <u>Th/ Ir₂₂₉</u> | <u>P/ Ir</u> | <u>Si/ Ir</u> | <u>Others</u> | |
| MHFT-40 | WC-N | MF | 2009h at 1214° GIS ^a | 1-2 | 0 | 0 | 0 | -- | Push-through |
| MHFT-47 | WE | " | 720h at 1214° GIS | 1-3 | 0 | 0 | 0 | -- | Push-through |
| MHFT-55 | HD | " | 777h at 1214°C GIS | 4.5 | 0.32 | 0.03 | 0 | -- | Push-through |
| 56 | HD | " | 777 at 1214°C GIS | 2.5 | 0.26 | 0.11 | 0 | -- | Hoop + Fingerprint |
| 57 | HD | " | 4000h at 1215°C GIS | 3.8 | 0.15 | 0.06 | 0.05 | -- | Possible Fingerprint |
| 58 | HD | LASL | 737h at 1215°C GIS | 4.5 | 0.27 | 0.03 | 0 | -- | No |
| 59 | HD | LASL | 737h at 1210°C GIS | 5 | 0.39 | 0 | 0.12 | -- | No |
| 60 | HD | LASL | 4423h at 1210°C GIS | 1-2 | 0 | 0 | 0 | -- | Hoop |
| 62 | HDR | SR | 720h at 1210°C GIS | 12.6 | 0.30 | 0 | 0 | -- | Push-through + Fingerprint |
| 64 | DOP-26 | " | 720h at 1210°C GIS | 13 | 0.34 | 0.02 | 0 | Hi S | Fingerprint |
| 65 | DOP-26 | " | 4400h at 1210°C GIS | 5.6 | 0.29 | 0.56 | 0 | Hi S | Large Push- Through + Fingerprint |
| 66 | HD | " | 720h at 1210°C GIS | 7.5 | 0.46 | 0.09 | 0 | -- | Fingerprint |

^aCorresponds to PICS temperature of 1330°C

2. Phosphorus in MHFT-65. An effort to identify the source of the phosphorus impurity which was observed in the metallographic examination of the intact vent assembly is being conducted.

Routinely, the GIS debris from an impact test is analyzed for plutonium by ignition of the recovered debris in oxygen, dissolution of the resulting ash, and analysis of the solution for plutonium by radio-chemical techniques. The residue obtained by taking an aliquot of this solution to dryness with nitric acid and ignition in oxygen at 900°C was analyzed for phosphorus by emission spectroscopy. The residue, which in addition to PuO₂ and ash from the GIS contained an undetermined quantity of oxidized dust from the abrasive cut-off wheel and tantalum powder, was found to contain 0.17% phosphorus.

The plutonium analysis indicated 1.85 g of plutonium (2.1 g of PuO₂) to be present in the GIS debris. Therefore a minimum of 3.6 mg of phosphorus was present in the debris of the GIS.

If the source of the phosphorus was the GIS (nominally 120 g) this would be equivalent to a minimum phosphorus content of 30 ppm in the GIS.

3. Phosphorus-thorium. The element phosphorus has been found in the grain boundaries of several MHW iridium shells. The source of this element has not been established. The compound ThP is known to be very stable. It does not vaporize significantly below 2000°C, and it has a melting point approaching 3000°C. It is possible that the thorium dopant in the iridium reacts with the phosphorus present to form ThP in the grain boundaries which in turn reacts with the fuel oxygen to form ThO₂ which then dissolves into the fuel, effectively depleting the thorium content of the iridium. The phosphorus released could form IrP or PuP compounds which provide further driving forces for the reaction.

IV. SAFETY ENGINEERING - LIGHT-WEIGHT RADIOISOTOPIC HEATER UNIT (LWRHU)

A test plan was written for vibration testing of assembled LWRHU components. The primary objective is to evaluate the integrity and stability of two LWRHU insulator candidate materials, PG, and CBCF-3, in the vibration environment anticipated at Galileo launch. Prototype heat source assemblies have been built from MF production drawings which differ from the final product only in that the "fuel" pellets are depleted uranium dioxide. No vibration levels or resonances have as yet been established for the LWRHU performance. As an alternative an initial test will be conducted at 0.1 g²/Hz for 1 minute to evaluate degradation at that level. Assuming no significant deterioration is observed a subsequent test at twice that level will be conducted. Further tests at higher levels may be conducted to establish a failure limit.

One result of building these assemblies is measured values of the LWRHU weights; 32.8 g with CBCF-3 insulator and 40.0 g with PG insulator.

Three Pt-30 Rh capsules were loaded with UO₂ simulant pellets and transferred for vibration testing.

Two weld development capsules were welded and then sectioned for metallographic examination. The first capsule was welded with DC arc. The second capsule was welded in an identical manner, but the arc was pulsed.

V. FUEL PROCESSING

A. Processing

During November, 1 lot (0.25 kg) of Savannah River Plant (SRP) feed powder was processed at the Los Alamos Scientific Laboratory. The lot was ^{16}O -exchanged and then ball-milled for 40 h and slugged and screened to form $<125\text{-}\mu\text{m}$ granules. The granules were thermally seasoned and set aside for the fabrication of GPHS pellets.

B. Residues and Shipments

Six SR shipping containers were loaded with plutonium oxide-containing residues for shipment to SR for recycle. These containers comprise residue shipment RS-238-62 (1.00 kg ^{238}Pu).