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Progress Report



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

December 1979

University of California

LOS ALAMOS SCIENTIFIC LABORATORY
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## December 1979

Compiled by

**W. J. Maraman**



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SPACE NUCLEAR SAFETY AND FUELS PROGRAM

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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. The changes in the schedule from the previous one were caused by the Department of Energy's decision to test both the CBCF-3 insulation concept and the pyrolytic graphite (PG) insulation in impact and vibration.

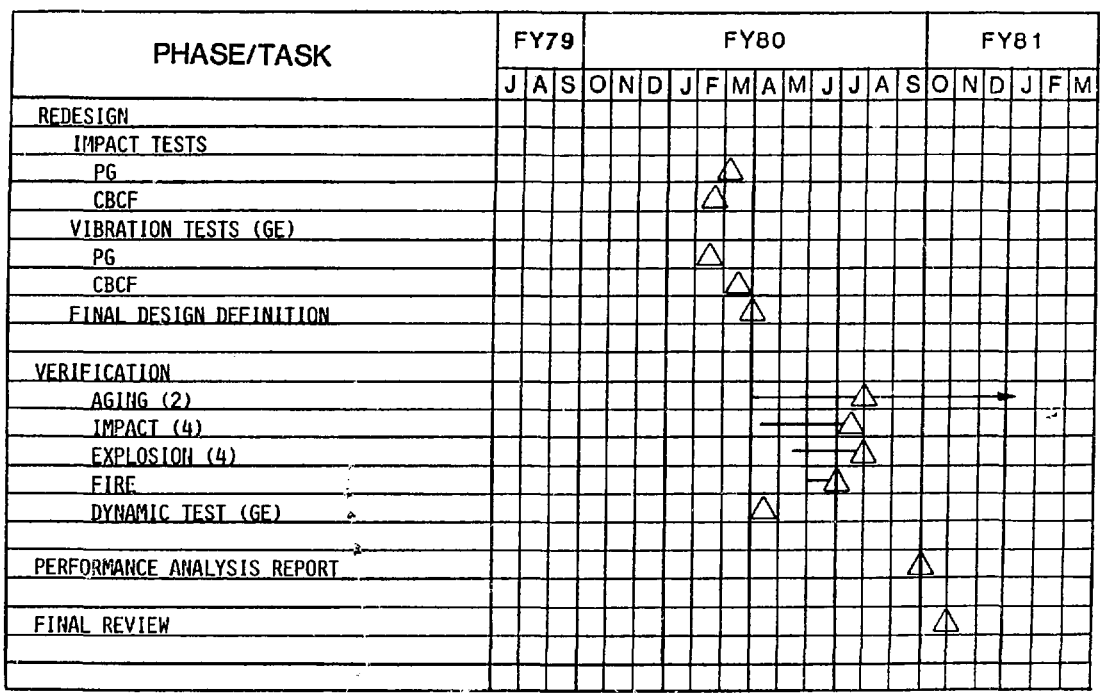



Fig. I-1.
GPHS Project Schedule.

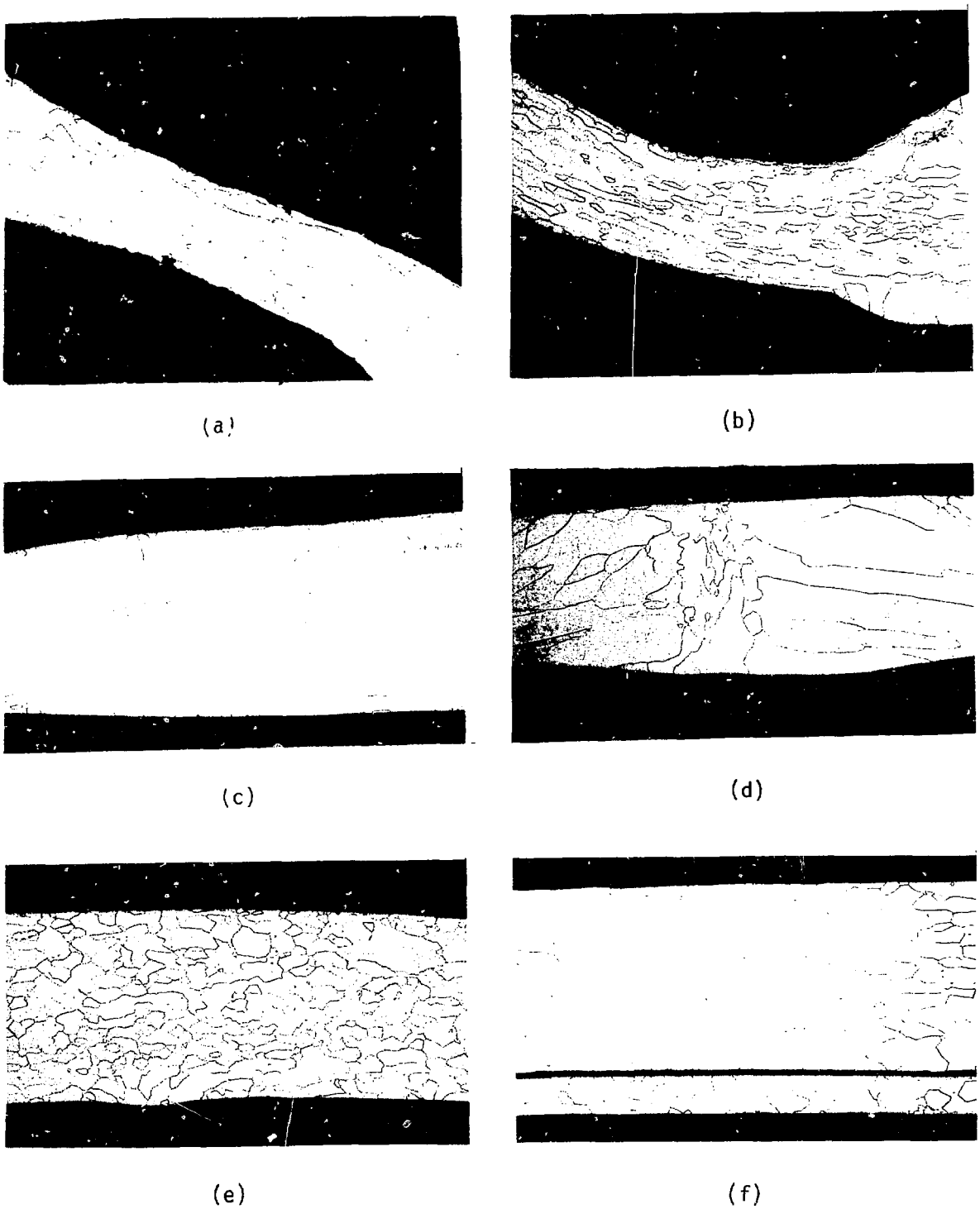



Fig. I-4.
Photomicrographs of capsule IRG-44 after impact at 58 m/s and 965°C following a heat pulse to 1740°C. 50X.

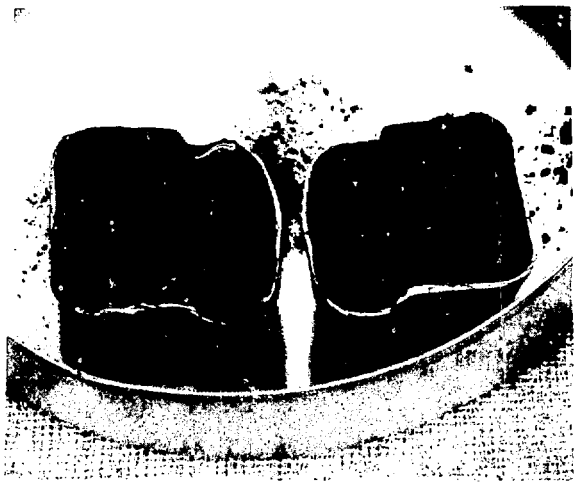


Fig. I-5.
The fuel failure that caused the severe local deformation of IRG-44 is visible in both halves of the section.

## 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 2374 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 four 32-mm summer rains increased this month (Table II-2). The average water volume was 5.0 ℓ, compared with 3.4 ℓ last month, while the average plutonium content changed to 1.67 from 0.99 μCi. The plutonium collected in the dehumidifier condensate changed from 0.4 to 0.5 μCi/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 8 days. The average of the measured volumes for four 32-mm rains is slightly higher, but the average plutonium content is a factor of two lower than last month (Table II-3). The average volume increased from 3.0 to 3.3 ℓ, and the average plutonium content decreased from 0.69 to 0.36 μCi. The collection rate of plutonium by the dehumidifier condensate increased from 0.05 to 0.24 μCi/wk.

TABLE II-1

TERRESTRIAL ENVIRONMENTAL CHAMBER EXPERIMENTS

| Experiment Number | Description | Date Started |
|---|---|---|
| 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 third

TABLE II-2

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

| No. | Date | Vol. Collected (ℓ) | Pu Found (μCi) |
|---|---|---|---|
| 328 | 11/19 | 5.3 | 1.79 |
| 329 | 11/26 | 5.2 | 1.86 |
| 330 | 12/3 | 4.2 | 1.23 |
| 331 | 12/10 | 5.3 | 1.81 |
| Average | | 5.0 | 1.67 |

TABLE II-3

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

| No. | Date | Vol. Collected (ℓ) | Pu Found (μCi) |
|---|---|---|---|
| 320 | 11/19 | 3.9 | 0.30 |
| 321 | 11/26 | 3.1 | 0.34 |
| 322 | 12/3 | 3.0 | 0.32 |
| 323 | 12/10 | 3.2 | 0.47 |
| Average | | 3.3 | 0.36 |

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 8 days. Liquid scintillation analysis and volume measurement of one 81-mm rain gave results of 0.04 μCi of plutonium in 2.8 ℓ, compared with 0.06 μCi in 4.2 ℓ last month. The collection rate for plutonium in the dehumidifier condensates was 0.11 μCi/wk, compared with 0.27 μCi/wk last month.

The finer fraction of MHFT-27, 34 g of particles of PPO with diameters between 0.01 and 6mm, 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 0.008 μCi of plutonium in 2.6 ℓ of water, lower in plutonium than the 0.012 μCi of plutonium in 3.5 ℓ of water for the last rain. The dehumidifier condensates collected 0.07 μCi/wk of plutonium, higher than the 0.01 μCi/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 eight days into the sixth winter. The soil in this 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 four 34-mm rains the average values for both compartments are higher this month (Table II-4). In the inner compartment the average volume was 1.48 ℓ and the average plutonium content was 0.109 μCi, compared with 1.13 ℓ and 0.095 μCi last month. In the outer compartment the average volume increased from 9.4 to 11.2 ℓ, while the plutonium content increased from 0.10 to 0.13 μCi. The rate of plutonium collection by the dehumidifier condensate changed from 0.9 to 0.7 μCi/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

TABLE II-4

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

| | | Inner | | Outer | |
|---|---|---|---|---|---|
| No. | Date | Vol (ℓ) | Pu (μCi) | Vol (ℓ) | Pu (μCi) |
| 237 | 11/19 | 2.25 | 0.155 | 10.0 | 0.120 |
| 238 | 11/26 | 1.65 | 0.171 | 11.0 | 0.120 |
| 239 | 12/3 | 1.10 | 0.085 | 11.0 | 0.125 |
| 240 | 12/10 | 0.92 | 0.023 | 12.6 | 0.140 |
| Average | | 1.48 | 0.109 | 11.2 | 0.126 |

are also on loam in a similarly compartmented tray. Liquid scintillation analyses and measured volumes of percolated rainwater from four rains show an increase in the average volume, but a decrease in plutonium content for the inner compartment, while the outer compartment averages changed very little (Table II-5). The average for the outer compartment volumes and plutonium contents are 5.1 ℓ and 0.142 μCi, respectively, compared with 5.7 ℓ and 0.138 μCi of plutonium last month. The inner compartment averages are 3.0 ℓ and 0.036 μCi, compared with 2.2 ℓ and 0.056 μCi last month. The collection rate for plutonium in the dehumidifier condensate changed from 0.2 to 0.4 μCi/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-50 ON COMPARTMENTED SOIL)

| | | | Inner | | Outer | |
|---|---|---|---|---|---|---|
| No. | Date | Rainfall (mm) | Vol (ℓ) | Pu (μCi) | Vol (ℓ) | Pu (μCi) |
| 238 | 11/19 | 26 | 2.7 | 0.048 | 5.5 | 0.176 |
| 239 | 11/26 | 27 | 3.2 | 0.040 | 4.8 | 0.137 |
| 240 | 12/3 | 27 | 3.1 | 0.033 | 5.5 | 0.110 |
| 241 | 12/10 | 27 | 2.9 | 0.024 | 4.5 | 0.143 |
| Average | | | 3.0 | 0.036 | 5.1 | 0.142 |

TABLE II-6

Pu TRANSPORT FROM $PuO_2$ PARTICLES

| Column | Elution Duration: 1011 days Total Vol (ℓ) | 1011 days Total Pu (ng) | 1136 Total Vol (ℓ) | 1136 Total Pu (ng) |
|---|---|---|---|---|
| 7 | 74.1 | 29.7 | 82.0 | 31.1 |
| 8 | 64.2 | 26.1 | 71.5 | 28.0 |
| 9 | 34.0 | 38.6 | 38.3 | 40.6 |

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, $\sim$ 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 $PuO_2$ particles on its soil surface, with a particle size distribution mainly in the 40- and 60-μm range, column 8 contains 2.6 mg $PuO_2$ particles with an average diameter of 12 μm, and 9 contains 2.6 mg with an average diameter of 7 μm. 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 1136 days. Measurements made after 1011 and 1136 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 $PuO_2$ 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 $HClO_4$

| Experiment Number | $PuO_2$ (mg) | Percent Dissolved | Dissolution Rate (g/s) | Dissolution Rate (g/s-g) |
|---|---|---|---|---|
| A1 | 2.02 | 4.6 | $7.3\times10^{-13}$ | $3.6\times10^{-10}$ |
| A2 | 2.05 | 4.2 | $6.8\times10^{-13}$ | $3.3\times10^{-10}$ |
| A3 | 10.19 | 4.3 | $3.5\times10^{-12}$ | $3.4\times10^{-10}$ |
| A4 | 9.90 | 4.3 | $3.3\times10^{-12}$ | $3.3\times10^{-10}$ |
| B1 | 2.02 | 7.9 | $1.3\times10^{-12}$ | $6.2\times10^{-10}$ |
| B2 | 2.03 | 9.2 | $1.5\times10^{-12}$ | $7.2\times10^{-10}$ |
| B3 | 10.25 | 8.3 | $6.7\times10^{-12}$ | $6.5\times10^{-10}$ |
| B4 | 10.45 | 8.0 | $6.5\times10^{-12}$ | $6.3\times10^{-10}$ |
| C1 | 2.23 | 18.1 | $3.2\times10^{-12}$ | $1.4\times10^{-9}$ |
| C2 | 2.31 | 17.1 | $3.1\times10^{-12}$ | $1.3\times10^{-9}$ |
| C3 | 9.89 | 17.2 | $1.3\times10^{-11}$ | $1.3\times10^{-9}$ |
| C4 | 10.67 | 16.7 | $1.4\times10^{-11}$ | $1.3\times10^{-9}$ |

## B. Aquatic Environments

1. Aqueous Release Rates As A Function of $PuO_2$ Particle Size. Three sets of sized $^{238}PuO_2$ particles, each having a different size distribution, are suspended in 1M $HClO_4$ 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 1485 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.

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 | 2092 | Fresh | 10 | 130 |
| HPZ-111-1 | 25 | 1796 | Fresh | 10 | 390 |
| HPZ-59-4 | 25 | 1883 | Sea | 10 | 16 |
| HPZ-174 | 18 | 772 | Sea | 10 | 7 |
| HPZ-186-4 | 19 | 776 | Sea (Tidal) | 10 | 59 |

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 $^{238}PuO_2$ 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 "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 $Pu^{+4}$, 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 1883 days (Table II-8). Its release rate is 16 $nCi/m^2$-s, up 14% from last month.

The tidal simulation aquarium, using the 19-W source HPZ-186-4, has been under way for 776 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 hours. Figure II-1 is a plot of plutonium concentration in μg/ℓ 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 59 nCi/m$^2$-s, unchanged 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 μg to 26.2 μg with an average of 42 ± 12 μg. 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 772 days. The release rate is 7 nCi/m$^2$-s, unchanged in the last 4 months. This is the same system in which we maintained a live sea cucumber for 230 days.

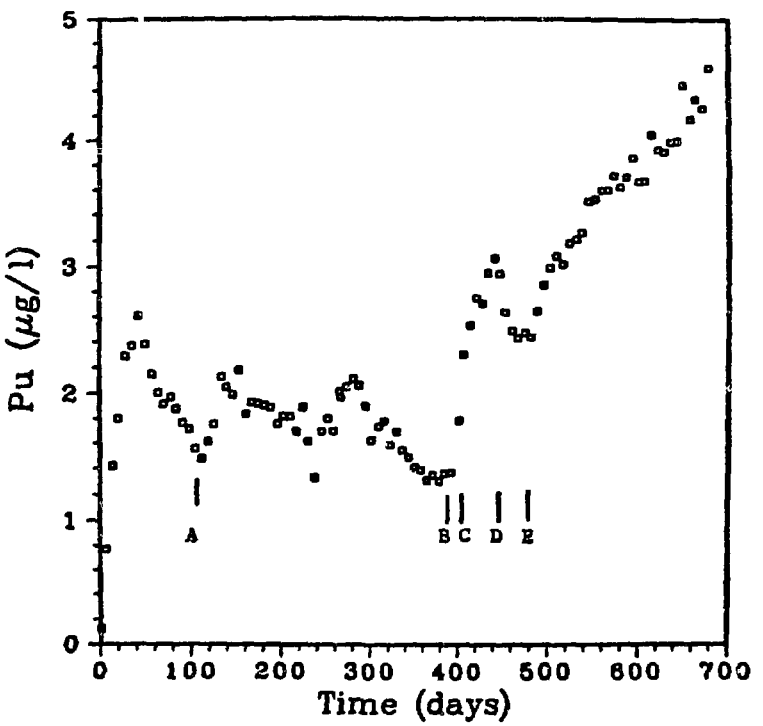



Fig. II-1.
Plutonium concentration in the tidal simulation experiment.

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 2092 days of immersion is 130 nCi/$m^2$-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 1796 days at 10°C. Its average release rate is 390 nCi/$m^2$s, down 3% 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.

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 $^{238}PuO_2$, 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 $^{238}PuO_2$ on the interior surfaces. This was accomplished using a $HNO_3$-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 pg/$m^2$-s. This number can be compared to the release rates seen in the aquatic environment experiments which vary from 0.59 to 36 ng/$m^2$-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 μg/$m^2$-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, two chambers leaked at the center seal. At the present time, two types of gasket material are being tested to seal the chambers.

d. 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.

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 $HNO_3$-$HClO_4$. 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 resultant recovery was 96 ± 10%. The two aliquots from the water sampler produced 230 d/m and 13 d/m. It can be suggested that one aliquot was contaminated or that some plutonium was lost during ion exchange in the other aliquot. The two remaining aliquots will be processed using lanthanum floride precipitation. After completion of the tests on the first samples, the second sampler will be processed.

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.

## III. SYSTEMS SUPPORT

### A. Stirling Isotope Power System

As of January 1, 1980 the accumulated exposure time for the 800°C test assembly was 17 458 h.

### B. Improved Multi-hundred Watt (MHW) Tests

The exposure of test ID-1 (MHFT-61) was terminated on December 26, 1979 after 8834 h of exposure. Aging of all four of the assemblies in theis series has been completed. Examination of assembly Pt-3008/$F_2$ is pending. MHFT-61 will be impacted before its examination.

C. Galileo

As part of the continuing effort to identify the source of the phosphorous impurity that has been detected as a grain boundary impurity in iridium by Auger analysis of the iridium shells of test MHW fueled sphere assemblies (FSA), and attempt to detect phosphorous impurity in the Savannah River (SR) plutonia fuel spheres on hand (MHFT-67 through -72) at Los Alamos Scientific Laboratory (LASL) is being conducted.

The technique being used involves recording of the $\gamma$-ray spectrum from the individual FSAs, and searching for $\gamma$-rays reuslting from the $(\alpha,\gamma)$ reaction on $^{31}P$. This method is attractive because it is nondestructive and could be applied to the units that have been fabricated for the Galileo mission Radioisotopic Thermonuclear Generators (RTG).

To date, results obtained for MHFT-67 indicate less than 25 ppm phorphorus in the plutonia sphere of this assembly.

Last month we presented Auger Electron Spectroscopy (AES) results on impurities in recent iridium shells, including MHFT-65. We now have analyzed a total of six separate specimens of post-impact containment shells (PICS) MHFT-62, -64, and -65 and five specimens of MHFT-66. The most recent results on MHFT-62 through -65 are presented in Table III-1. The results presented last month are updated in Table III-2 to reflect the additional work. In Table III-3 we compare these results to other shells that are part of the MHW impact series. The average thorium concentrations are similar to previous PICS, but still less than that expected from as-annealed sheet. The phorphorus levels were unusual only in MHFT-65. All four iridum shells of the current series, all of which contained SR fuel, contained sulfur at the grain boundaries. The levels in MHFT-62 and -66 were low, whereas those in MHFT-64 and -65 were high. In all cases the presence of sulfur was sporadic. The results in Table III-2 fail to demonstrate any distinct gradient of impurities through the thickness of these shells. The AES results suggest the following conclusions: (1) The level of phosphorus was unusually high only for MHFT-65, (2) we cannot determine the source of phorphorus from these results, (3) the SR fuel appears to be the source for the sulfur contamination, and (4) sputtering experiments showed that thorium, phosphorus, and sulfur segregation were all within a few atomic layers of the grain boundary.

We also have previously summarized AES results from MHW iridium shells that were not impacted. Table III-4 contains the most recent results in this category. These results are listed with the previous data in Table III-5. It is apparent that phosphorus contamination is not a new problem. As shown in Table III-3 and -5, phosphorus has been found in all types of Ir-0.3% W including Engelhardt, undoped ORNL, DOP-4, and DOP-26.

Although phosphorus contamination is often found in Ir-0.3% W PICS, it has not been demonstrated conclusively that phosphorus segregation to grain boundary causes a loss in ductility. We have previously demonstrated that we can dope Ir-0.3% W with phosphorus by heating in the presence of $P_2O_5$ at a temperature of ~1500°C. In order to determine the effect of phosphorus on iridium ductility we annealed a tensile specimen and a biaxial punch test specimens at 1500°C for 19 h with $P_2O_5$ in graphite. Control specimens of the same materials were annealed at 1575°C for 19 h in vacuum in an attempt to duplicate the grain size. AES results on grain boundary chemistry confirmed the presence of phosphorus in the doped specimens. The results, listed in Table III-6, show that the phosphorous levels in the test specimens were approximately half of those found in MHFT-65. The grain sizes were 7.8 grains per thickness for the tensile specimen (lot OLMF-8) and 8.7 grains per thickness for the biaxial disk (lot L-30). The control tensile specimens exhibited 7.4 grains per thickness and the control disk had

# TABLE III-1

## AES RESULTS ON MHFT-62, -64 AND -65

| Specimen | Treatment | Location | Auger Intensity Ratios $Th_{65}/Ir_{229}$ | $P_{120}/Ir_{229}$ | $C_{270}/Ir_{229}$ | $O_{510}/Ir_{229}$ | $Si_{92}/Ir_{229}$ | $Si_{150}/Ir_{229}$ | $Th_{65}/Ir_{54}$ |
|---|---|---|---|---|---|---|---|---|---|
| MHFT-65C | DOP-26 Ir-0.3% W 6 months age in FSA at 1210°C GIS | Edge Inside | 0.15 | 0.79 | 1.50 | 0.60 | 0 | 0.40 | 0.020 |
| | | Center | 0.47 | 0.16 | 0.60 | 1.02 | 0 | Trace | 0.084 |
| | | Edge Outside | 0.55 | 0.28 | 0.84 | 1.21 | 0 | 0.27 | 0.093 |
| MHFT-65D | -"- | Edge Inside | 0.27 | 0.73 | 0.58 | 0.58 | 0 | 0 | 0.041 |
| | | Center | 0.83 | 0.84 | 0.23 | 0.50 | 0 | 0 | 0.110 |
| | | Edge Outside | 0.53 | 1.42 | 0.42 | 0.53 | 0 | 0 | 0.072 |
| MHFT-64-B3 | DOP-26 Ir-0.3%W Aged 30 days in FSA at 1210°C GIS | Edge Inside | 0.34 | 0 | 0.68 | 0.47 | 0 | Trace | 0.056 |
| | | Center | 0.56 | 0.19 | 0.32 | 0.98 | 0 | 0 | 0.076 |
| | | Edge Outside | 0.35 | 0 | 1.5 | 3.4 | 0 | 0.3 | 0.064 |
| MHFT-62-B3 | MDR Ir-0.3%W Aged 30 days in FSA at 1210°C GIS | Edge Inside | 0.24 | 0 | 0.57 | 0.71 | 0 | 0.17 | 0.038 |
| | | Center | 0.52 | 0 | 0.57 | 1.13 | 0 | Trace | 0.074 |
| | | Edge Outside | 0.35 | 0 | 0.88 | 0.82 | 0 | Trace | 0.075 |
| MHFT-64-A2 | Same as 64-B3 | Edge Inside 150um area | 0.40 | Trace | 1.45 | 1.58 | 0 | 0.25 | 0.105 |
| | | Center | 0.69 | 0.21 | 0.51 | 0.86 | 0 | 0 | 0.098 |
| | | Edge Outside | 0.32 | 0.14 | 0.94 | 0.89 | 0 | 0 | 0.131 |
| MHFT-64 B2 | Same | Edge Inside | 0.33 | 0 | 0.58 | 0.83 | 0 | 0.19 | 0.051 |
| | | Center | 0.67 | 0.23 | 0.23 | 0.65 | 0 | 0 | 0.094 |
| | | Edge Outside | 0.47 | 0.16 | 0.64 | 0.93 | 0 | 0.15 | 0.075 |
| MHFT-64-A3 | Same | Edge Inside | 0.46 | 0.40 | 0.38 | 0.60 | 0 | 0 | 0.051 |
| | | Center | 1.06 | 0.31 | 0 | 0.20 | 0 | 0 | 0.121 |
| | | Edge Outside | 0.71 | 0.34 | 0.13 | 0.33 | 0 | 0 | 0.097 |
| MHFT-62-A2 | Same as 62-B3 | Edge Inside | 0.02 | 0 | 0.54 | 0.54 | 0 | 0 | 0.002 |
| | | Center | 0.55 | 0 | 0.28 | 0.91 | 0 | 0 | 0.070 |
| | | Edge Outside | 0.54 | 0 | 0.45 | 0.98 | 0 | 0 | 0.075 |
| MHFT-62-B2 | Same | Edge Inside | 0.19 | 0 | 0.48 | 0.50 | 0 | 0 | 0.023 |
| | | Center | 0.76 | 0 | 0.20 | 0.50 | 0 | 0 | 0.072 |
| | | Edge Outside | 0.52 | 0 | 0.33 | 0.64 | 0 | 0 | 0.062 |
| MHFT-62-A3 | Same | Edge Inside | 0.04 | 0 | 0.74 | 0.85 | 0 | 0.52 | 0.005 |
| | | Center | 0.49 | 0 | 0.48 | 1.09 | 0 | 0.24 | 0.072 |
| | | Edge Outside | 0.54 | 0 | 0.73 | 1.02 | 0 | 0.41 | 0.084 |
| MHFT-66-A2 | HD Ir-0.3% W Aged 30 days in FSA at 1210°C GIS | Edge Inside 150um area | 0.30 | 0 | 0.44 | 0.70 | 0 | 0 | 0.037 |
| | | Center | 0.43 | Trace | Trace | Trace | 0 | 0 | 0.048 |
| | | Edge Outside | 0.63 | 0.25 | 0.97 | 1.14 | 0 | 0.36 | 0.095 |
| MHFT-66-B2 | | Edge Inside | 0.32 | 0 | 0.92 | 0.71 | 0 | 0.83 | 0.046 |
| | | Center | 0.37 | 0 | 0.82 | 1.02 | 0 | 0.62 | 0.047 |
| | | Edge Outside | 0.58 | 0.08 | 1.65 | 1.68 | Trace? | 1.07 | 0.088 |
| MHFT-66-A3 | | Edge Inside | 0.29 | 0 | 0.36 | 0.61 | 0 | 0 | 0.035 |
| | | Center | 0.68 | 0 | 0.31 | 0.61 | 0 | 0 | 0.080 |
| | | Edge Outside | 0.42 | 0.19 | 0.38 | 0.55 | 0 | 0 | 0.053 |

## TABLE III-2

## AES RESULTS ON RECENT MHW PICS

| | | $Th_{65}/Ir_{229}$ | $P_{120}/Ir_{229}$ | $S_{150}/Ir_{229}$ |
|---|---|---|---|---|
| MHFT-62* | Inside | 0.21 | 0 | $0.3^{(2)**}$ |
| | Center | 0.48 | 0 | $0.2^{(2)}$ |
| | Outside | 0.41 | 0 | $0.3^{(2)}$ |
| MHFT-64 | Inside | 0.37 | 0.09 | $0.6^{(4)}$ |
| | Center | 0.63 | 0.16 | $0.7^{(1)}$ |
| | Outside | 0.41 | 0.11 | $0.5^{(3)}$ |
| MHFT-65 | Inside | 0.20 | 0.66 | $1.2^{(4)}$ |
| | Center | 0.52 | 0.43 | $1.8^{(2)}$ |
| | Outside | 0.45 | 0.72 | $0.9^{(3)}$ |
| MHFT-66 | Inside | 0.30 | 0 | $0.83^{*(1)}$ |
| | Center | 0.53 | 0.03 | $0.62^{*(1)}$ |
| | Outside | 0.52 | 0.18 | $0.72^{(1)}$ |

** Numbers in parentheses indicate number of specimens that exhibited a sulfur peak.

* All specimens except 66 represent the results of six specimens. MHFT-66 results are from five specimens.

## TABLE III-3

## SUMMARY OF AES RESULTS FOR LIVE ($PuO_2$) IMPACTED FSAs

| Specimen | Ir Type | Fuel | Treatment | #grains/ thickness | $Th/Ir_{229}$ | P/Ir | Si/Ir | Others | Impact failure |
|---|---|---|---|---|---|---|---|---|---|
| MHFT-40 | WC-N | MF | 2009 h at 1214°C GIS* | 1-2 | 0 | 0 | 0 | -- | Push-thru |
| MHFT-47 | WE | MF | 720 h at 1214°C GIS | 1-3 | 0 | 0 | 0 | -- | Push-thru |
| MHFT-55 | HD | MF | 777 h at 1214°C GIS | 4.5 | 0.32 | 0.03 | 0 | -- | Push-thru |
| 56 | HD | MF | 777 h at 1214°C GIS | 2.5 | 0.26 | 0.11 | 0 | -- | Hoop + fingerprint |
| 57 | HD | MF | 4000 h at 1215°C GIS | 3.8 | 0.15 | 0.06 | 0.05 | -- | Possible fingerprint |
| 58 | HD | LASL | 737 h at 1215°C GIS | 4.5 | 0.27 | 0.03 | 0 | -- | No |
| 59 | HD | LASL | [illegible] h at 1210°C GIS | 5 | 0.39 | 0 | 0.12 | -- | No |
| 60 | HD | LASL | 4423 h at 1210°C GIS | 1-2 | 0 | 0 | 0 | -- | Hoop |
| 62 | HDR | SR | 720 h at 1210°C GIS | 12.6 | 0.37 | 0 | 0 | Trace S | Push-thru + fingerprint |
| 64 | DOP-26 | SR | 720 at 1210°C GIS | 13 | 0.47 | 0.12 | 0 | HI S | Fingerprint |
| 65 | DOP-26 | SR | 4400 h at 1210°C GIS | 5.6 | 0.39 | 0.60 | 0 | HI S | Large Push-thru + fingerprint |
| 66 | HD | SR | 720 h at 1210°C | 7.5 | 0.45 | 0.07 | 0 | Trace S | Fingerprint |

*Corresponds to PICS temperature of 1330°C

# TABLE III-4

## AES RESULTS FOR SEVERAL PICS NOT IMPACTED

| Specimen | Treatment | Location | Auger Intensity Ratios $Th_{65}/Ir_{229}$ | $P_{120}/Ir_{229}$ | $C_{270}/Ir_{229}$ | $O_{510}/Ir_{229}$ | $Si_{92}/Ir_{229}$ | $Sn_{428}/Ir_{229}$ | $Fe_{710}/Ir_{229}$ | $Ni_{850}/Ir_{22}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| MHF-165 | WER Iridium Q-2 R Vibration Test 5-7 grains/t | Edge Inside | 0 | 0 | 0.29 | 0.18 | 0 | 0.86 | 0 | 0 |
| | | Center | 0 | 0 | 0.16 | 0.16 | 0 | 0.84 | 0 | 0 |
| | | Edge Outside | 0 | 0 | 0.60 | 0.27 | B or $Cl_{180}/Ir_{229}$ 0 | | 0 | 0 |
| MHF-166 | WE Iridium Q-2R Vibration Test One grain/t | Edge Inside | 0 | 0 | 0.64 | 0.34 | 0.64 | 0 | 0.22 | 0.14 |
| | | Center | 0 | 0 | 0.54 | 0.48 | 0.37 | 0 | 0.19 | 0.18 |
| | | Edge Outside | 0 | 0 | 0.55 | 0.45 | 0.55 | 0 | 0 | 0.32 |
| MHF-161-1 | WER-Iridium Q-2R Vibration Test 2-3 grains/t | Edge Inside | 0 | 0 | 1.47 | 0.63 | 0 | 2.90 | 0 | 0 |
| | | Center | 0 | 0.26 | 1.31 | 0.54 | 0 | 0.35 | 0 | 0 |
| | | Edge Outside | 0 | 0.16 | 1.35 | 0.41 | $Si_{92}/Ir_{229}$ | 0.23 | 0 | 0 |
| MHF-161-2 | WER-Iridium Q-2R Vibration Test 1-2 grains/t | Edge Inside | 0 | 0 | 0.61 | 0.44 | 1.56 | 0 | 0.46 | 0 |
| | | Center | 0 | 0 | 0.13 | 0.55 | 1.68 | 0 | 0.21 | 0 |
| | | Edge Outside | 0 | 0 | 0.34 | 0.29 | 1.00 | 0 | 0 | 0 |
| MHFT-44-A | WC-R Iridium 27,310 h at 1145° C GIS 2-3 grains/t | Edge Inside | 0 | 1.10 | 0.61 | 0 | 0 | 0 | 0 | 0 |
| | | Center | 0 | 0.723 | 0.53 | 0.29 | 0 | 0 | 0 | 0 |
| | | Edge Outside | 0 | 0.797 | 0.81 | 0.36 | 0 | 0 | $Si_{150}/Ir_{229}$ | $Te_{490}/Ir_{22}$ |
| MHFT-44-B | -"- | Edge Inside | 0 | 0.239 | 0.67 | 0.58 | 0 | 0 | 1.20 | 0 |
| | | Center | 0 | 0.210 | 0.66 | 0.59 | 0 | 0 | 1.18 | 0.26 |
| | | Edge Outside | 0 | 0.333 | 0.75 | 0.64 | 0 | 0 | 1.15 | 0.71 |
| MHFT-23-A | El II, lot 46 Environmental sample 2-5 grains/t | Edge Inside | 0.22 | 0 | 0 | 0.82 | B or $Cl_{180}/Ir_{229}$ 0 | 0 | 0 | 0 |
| | | Center | 0 | 0 | 0 | 0 | 0.26 | 0 | 0 | 0 |
| | | Edge Outside | 0.13 | 0 | Trace | 0.62 | 0.18 | 0 | 0 | 0 |
| MHFT-23-B | Same | Edge Inside | 0.27 | 0 | 0.27 | 0.6 | 0.14 | 0 | 0 | 0 |
| | | Center | 0 | 0 | 0 | 0 | Trace | 0 | 0 | 0 |
| | 1-3 grains/t | Edge Outside | 0 | 0 | 0 | 0 | Trace | 0 | 0 | 0 |

## TABLE III-5

## IRIDIUM GRAIN SIZE DATA IN LIVE ($PuO_2$) FSAs THAT WERE NOT IMPACTED

| Specimen | Thermal Treatment | Type of Iridium | Grain Size, No. of grains/ 0.56 mm thickness | Auger Intensity Ratios of Impurities $Th_{65}/Ir_{229}$ | $P_{120}/Ir_{229}$ | $Si_{92}/Ir_{22}$ | Others |
|---|---|---|---|---|---|---|---|
| MHFT-1 | 18h at 1500°C with $PuO_2$ 2145 h at 1370°C GIS[a] | EI, lot 4A | 1-2 | b | | | |
| MHFT-8 | 18 h at 1500°C with $PuO_2$ | EI, 5 | >10 | b | | | |
| MHFT-10 | 18 h at 1500°C with $PuO_2$[c] 1248 h at 1250°C PICS[d] | EI, 11, 13 | 4 | b | | | |
| MHFT-11 | 18 h at 1500°C with $PuO_2$ | EI, 13 | 5 | b | | | |
| MHFT-14 | 18 h at 1500°C with $PuO_2$ 1248 h at 1250°C PICS[d] | EI, 13, 14 | 6.5 | b | | | |
| MHFT-15 | 7395 h at 1268°C GIS[a] in vacuum | EI, 36 | 1-3 | b | | | |
| MHFT-16 | 17339 h at 1018°C GIS[a] in He (1 atm) | EI, 36 | 4-6 | b | | | |
| MHFT-17 | 17713 h at 1018°C GIS[a] in He (1 atm) | EI, 36 | 4-6 | b | | | |
| MHFT-18 | 1332 h at 1180°C GIS[a] in He (1 atm) | EI, 46 | 2-5 | b | | | |
| MHFT-19 | 1105 h at 1268°C GIS[a] in vacuum | EI, 50 | 2-4 | b | | | |
| MHFT-20 | 18293 h at 1268°C GIS[a] in vacuum | EI, 46 | Variable 1-4 | 0 | 0.41 | 0 | B or Cl |
| MHFT-21 | 4508 h at 1268°C GIS[a] in He (1 atm) | EI, 46 | 2-5 | b | | | |
| MHFT-22 | 3330 h at 1268°C GIS[a] in vacuum | EI, 46 | 2-3 | b | | | |
| MHFT-23 | Environmental sample | EI, 46 | 1-5 | 0.1 | 0 | 0 | B or Cl |
| MHFT-25 | 857 h at 1272°C GIS[a] in vacuum (SST Series) | EI, 51 | 1-3 | b | | | |
| MHFT-28 | 738 h at 1272°C GIS[a] in vacuum (SST Series) | EI, 51 | 1-3 | b | | | |
| MHFT-37 | 23,818 h at 1210°C GIS, Reentry | WC | 1-2 | 0 | 0 | 0 | |
| MHFT-44 | 27,310 h at 1145°C GIS | WC-R | 1-4 | 0 | 0.57 | 0 | Hi S |
| MHFT-41 | 2016 h at 1207°C GIS[a] in vacuum (SVT Series) | WC, N | 1-2 | b | | | |
| MHFT-42 | 5440 h at 1210°C GIS[a] in vacuum (SVT Series) | WC, N | 1-2 | b | | | |
| MHF-27 | Q-1 generator tests | EI, 84 | 11 | b | | | |
| MHF-15 | Q-2R Vibratory tests | EI, 70 | 1-3 | Trace | 1.04 | Trace | B or Cl |
| MHF-64 | Q-2R Vibratory tests | WE | 1-3 | 0 | 0.26 | 0.18 | B or Cl |
| MHF-164 | Q-2R Vibratory tests | WER | 1-3 | 0 | 0.32 | 0.10 | B or Cl, Sn |
| MHF-161 | Q-2R Vibratory tests | WER | 1-3 | 0 | 0.07 | 0 | Fe, Sn |
| MHF-165 | Q-2R Vibratory tests | WER | 5-7 | 0 | 0 | 0 | Sn |
| MHF-166 | Q-2R Vibratory tests | WE | 1 | 0 | 0 | 0 | B, Fe, Ni |

[a] GIS temperature, IR PICS Temperature was ~50°C higher

[b] Not analyzed

[c] MHFT-10, and MHFT-15 and on, were given anneal of 1 h at 1500°C to Ir hemishells

[d] Temperature not known very accurately

## TABLE III-6

## AES RESULTS ON TENSILE AND BIAXIAL TEST SPECIMENS

| SPECIMEN | Treatment | Location | Auger Intensity Ratios Th /Ir 65 229 | P /Ir 120 229 | C /Ir 270 229 | O /Ir 510 229 | Si /Ir 92 229 | Th /Ir 65 54 |
|---|---|---|---|---|---|---|---|---|
| OLMF-8-61 | 19h at 1500°C with $P_2O_5$ tensile specimen | Edge 150μm area | 0.83 | 0.37 | 5.0 | 3.5 | 0.46 | 0.126 |
| | | Center | 0.95 | 0.36 | 0.14 | 0.28 | 0 | 0.123 |
| | | Edge | 0.75 | 0.42 | 0.35 | 0.60 | 0 | 0.100 |
| L-30-5A | 19h at 1500°C with $P_2O_5$ Biaxial disc (near fracture) | Edge | 0.42 | 0.29 | 0.47 | 0.93 | 0 | 0.059 |
| | | Center | 0.60 | 0.47 | 0.40 | 0.90 | 0 | 0.075 |
| | | Edge | 0.32 | 0.31 | 0.40 | 0.64 | 0 | 0.042 |
| L-30-5B | Same (near edge) | Edge | 0.46 | 0.20 | 1.07 | 1.21 | 0 | 0.079 |
| | | Center | 0.43 | 0.09 | 0.81 | 1.50 | 0 | 0.077 |
| | | Edge | 0.46 | 0.16 | 0.85 | 1.27 | 0 | 0.075 |
| L-30-1A | 19h at 1575°C in vacuum on graphite | Edge | 0.36 | 0 | 0.60 | 1.25 | 0 | 0.052 |
| | | Center | 0.41 | 0 | 0.34 | 0.90 | 0 | 0.053 |
| | | Edge | 0.34 | 0 | 0.62 | 1.07 | 0 | 0.057 |

6.2 grains per thickness. The variation in grain sizes is due to the different lots of DOP-26 Ir-0.3% W. These treatments gave us an excellent comparison for the tensile specimens, but resulted in a substantial grain size difference for the biaxial disks.

The tensile specimens were tested in our Two-Inch Gun Facility at 1350°C and 45 m/s. OLMF-8-61, doped with phosphorus, exhibited a total elongation to failure in a 12-mm gage length of 5.1%. The two control specimens, OLMF-8-64 and -65, broke in the grips after an elongation of ∿7% (see Table III-7). It is not possible to assess the influence of phosphorus on tensile ductility from these results. The biaxial disks were tested at 1440°C and 45 m/s. The phosphorous-doped specimen (L-30-5) and control specimen (L-30-1) are shown in Fig. III-1. The failure strains determined with the aid of an etched circle grid were ∿75% for both specimens. Both specimens necked and exhibited extensive ductility before fracture. The phoshorous-doped spemimen appears less catastrophically fractured. This is simply an artifact of the test. During testing the phosphorous-doped specimen pulled in from the hold-down groove which resulted in less strain per unit penetration. The fracture strains as measured by the circles were the same. As mentioned above, the grain size of the control specimen was somewhat greater than that of the phosphorous-doped specimen, thereby obscuring the comparison somewhat. One can, however, state unequivocably that phosphorus does not cause embrittlement under the test conditions examined here. Further tests to examine the effects of phosphorus on grain growth and ductility are in progress.

TABLE III-7

SUMMARY OF PHOSPHOROUS-DOPING EXPERIMENTS

| Specimen No. | Type of Specimen | Treatment | Grains per thickness | Failure strain | Th /Ir 65 | Th /Ir 229 | P /Ir 120 | P /Ir 229 |
|---|---|---|---|---|---|---|---|---|
| OLMF-8-64 | Uniaxial tensile | 19h at 1575° C in vacuum on graphite | --- | >7.1[a] | --- | | --- | |
| OLMF-8-65 | -"- | -"- | 7.4 | >7.4[a] | --- | | --- | |
| OLMF-8-61 | -"- | 19h at 1500° C with $P_2O_5$ | 7.8 | 5.1[a] | 0.81 | | 0.38 | |
| L-30-1 | Biaxial disc | 19h at 1575° C in vacuum on graphite | 6.2 | ∿75[b] | 0.34 | | 0 | |
| L-30-5 | -"- | 19h at 1575° C with $P_2O_5$ | 8.7 | ∿75[b] | 0.45 | | 0.36 | |

(a) In a 12mm gage length.

(b) Measured by a 1.27mm diam circle.


(a) (b)

Fig. III-1.
Biaxial disk specimens tested at 1440°C and 45 m/s. (a) Control specimen L-30-1, fracture strain of ∿75% and (b) phosphorous-doped specimen L-30-5, fracture strain of ∿75%.

## IV. SAFETY ENGINEERING

### A. Light-Weight Radioisotopic Heater Unit (LWRHU)

1. Vibration testing. Three LWRHU vibration tests were carried out this month on samples with three insulator configurations, one with PG and two with CBCF insulation. The PG design survived the test environment, but both CBCF designs failed at one-fourth of the specified environment.

The random vibration environment for the RHUs was specified by the Jet Propulsion Laboratory (JPL) to be 10 $G^2$/Hz between 100 and 200 Hz, 0.4 $G^2$/Hz between 400 and 1000 Hz with appropriate slopes between those plateaus in the overall range of 20 to 2000 Hz. The test duration was 4 min. The tests were carried out in the LASL WX-3 vibration test facility. The test objects were secured in closely fitting cavities in an aluminum block by compression spring loaded to 44 kg.

The three types of assemblies tested were:

a. The prime LWRHU design with a five-piece PG insulator (Monsanto Research Corp. drawing AYD 790379).
b. A LWRHU with a three-piece CBCF insulator made with a nominal clearance of 0.1 mm between the Pt-3008 capsule and the insulator.
c. A LWRHU with a three-piece CBCF insulator assembled with minimum clearances.

In the CBCF designs, the end caps had the correct orientation, but the tubes were machined from CBCF plate and did not have the correct orientation for insulation. We do not believe that this deviation, which is made necessary by the unavailability of formal tubes, affected the outcome of the tests.

In the first test, all three designs were vibrated simultaneously. Mindful of the recent experience with some CBCF insulation in vibration tests of GPHS prototypes, we elected to perform this first test at half the JPL levels, i.e. 5 $G^2$/Hz and 0.2 $G^2$/Hz for two minutes along the axis and two minutes perpendicular to it. The PG design survived intact with only a minimum of burnishing where its components rubbed. Both the CBCF insulators were drastically altered. Large holes appeared in the end caps, which lost 60% of their mass, while the tubes were seriously abraded inside and lost 20% of their mass. Figure IV-1 shows the nominal-clearance CBCF assembly after the half-level test and Fig. IV-2 shows the tighter CBCF assembly. The difference in clearances did not make any difference in the catastropic degradation of the CBCF parts at this vibration level.

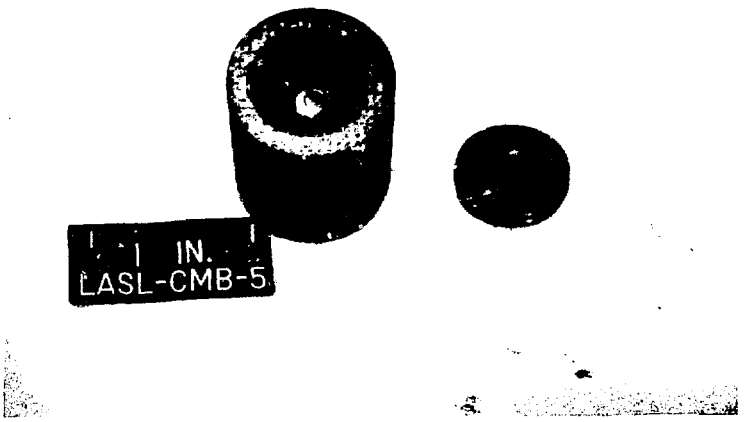
In the second LWRHU vibration test the PG-insulated assembly was tested by itself at 6 $G^2$/Hz and 0.4 $G^2$/Hz. We could not attain 10 $G^2$/Hz without overloading the testing machine. The assembly, shown in Fig. IV-3, survived the test with only minimal marking where the components rubbed.

In the third LWRHU vibration test the two CBCF configurations were tested at one-forth the JPL-recommended levels, 2.5 $G^2$/Hz and 0.1 $G^2$/Hz. The CBCF components were again drastically altered. Smaller holes were eroded in the upper caps, which lost 40% of their mass, while the tubes suffered a 15% mass loss.

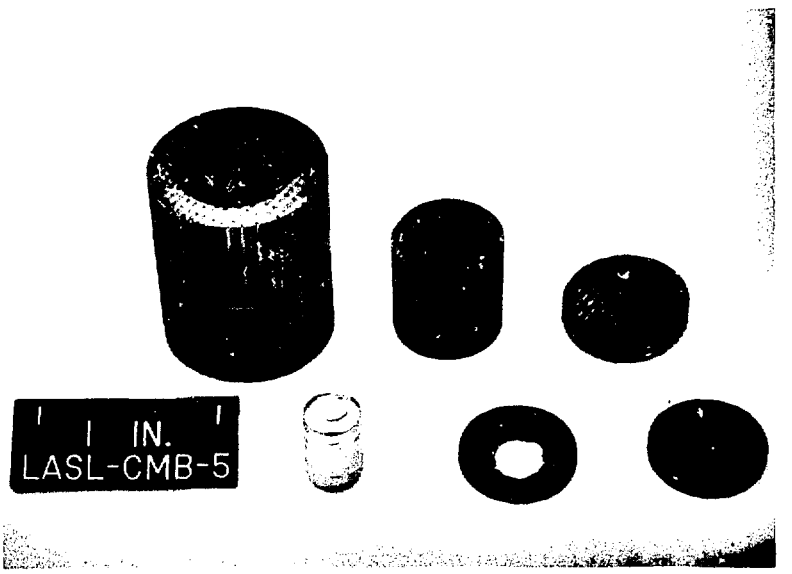
The following conclusions can be drawn from the test results.

a. The vibration environment is a strong constraint on the LWRHU design.
b. PG performs well in the JPL-specified environment, but CBCF performs poorly even in less severe environments.

Thus, we have a satisfactory LWRHU design from the mechanical viewpoint. The impact response reported last month and the vibration response reported

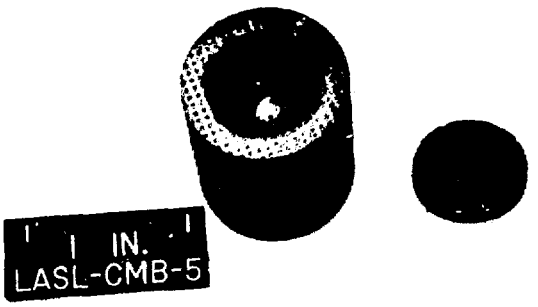



(a)

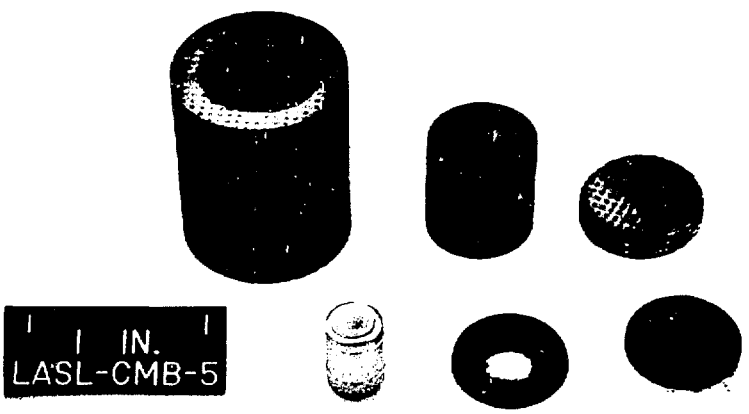



(b)

Fig. IV-1.
The CBCF LWRHU assembly with nominal clearances failed in the half-level vibration test. a) With the aeroshell cap removed the fuel capsule is visible through the top CBCF end cap, and b) after disassembly, the abrasion of the CBCF tube was visible.




(a)




(b)

Fig. IV-2.
The minimum-clearance CBCF LWRHU assembly failed in the half-level vibration test. a) Removal of the aeroshell cap revealed the failure of the top CBCF end cap and b) after disassembly the FWPF parts and the capsule were seen to be undamaged and the inner surface of the CBCF tube was found to be abraded.

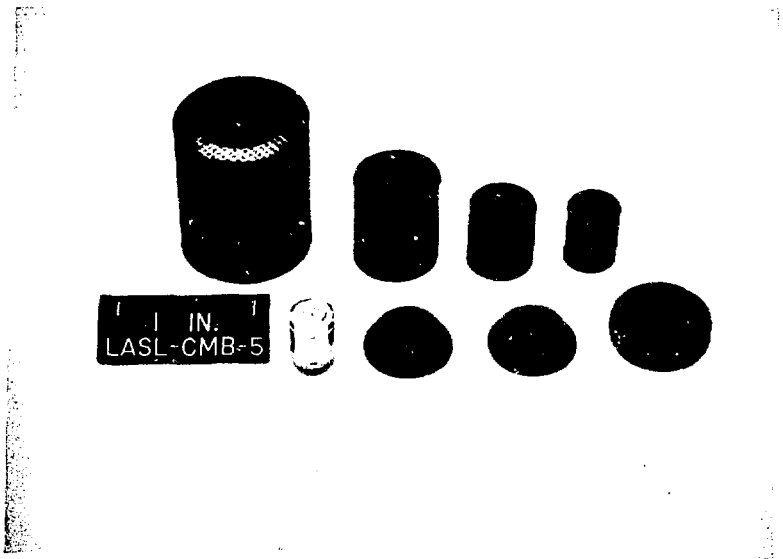



Fig. IV-3.
The PG-insulated LWRHU survived the 6 $G^2$/Ha (100-200 Hz), 0.4 $G^2$/Hz (400-1000 Hz) without damage.

above are proof of this. The thermal design is not known to be in such satisfactory shape. Applied Physics Laboratory has reported that the PG design will withstand side-on reentry, but they have not finished the analysis of the end-on reentry case.

2. Fuel. Three lots of 16-each chamfered RHU production pellets were hot pressed from <125-μm GROG-type feed granules seasoned at 1100°C (60 wt%) and 1600°C (40 wt%). The feed granules were processed from SR Lot 440, Run 906923 (LASL Lot 38) and SR Lot 440, Run 906922 (LASL Lot 39). This material is enriched at 83.5 at.% $^{238}$Pu, thus the fuel pellets have a thermal inventory slightly in excess of 1.1 watts at the time of fabrication.

After the pellets were hot pressed, they were sintered for 6 h at 1527°C in flowing Ar-$H_2{}^{16}O$. Fabrication parameters and pellet dimensions for pellet lot RU3 are listed in Table IV-1. One pellet from this lot, RU3-12, was sampled for spectrochemical analysis. A second pellet, RU3-1, was encapsulated and the neutron emission rate was measured. The neutron emission rate measured in the analytical chemistry group, CMB-1, was 5268 n/s-g $^{238}$Pu. The value measured in the new counter in CMB-11 was 5242 n/s-g $^{238}$Pu, in excellant agreement. The new counter will be used to determine the neutron emission rate for each encapsulated RHU pellet shipped from LASL.

Savannah River feed lot 440 is the first feed, enriched at 83.5 at.% $^{238}$Pu received at LASL. The neutron emission rate for the as-received feed was 18 475 n/s-g $^{238}$Pu. This value is somewhat higher than the values we have measured previously for SR feed enriched at 80 at.% $^{238}$Pu, 17 754 ± 191 n/s-g $^{238}$Pu. However, the neutron emission rate for pellet RU3-1 easily meets the value specified for LWRHU pellets, 6 000 n/s-g $^{238}$Pu.

TABLE IV-1

RHU PRODUCTION PELLET LOT RU3

| | |
|---|---|
| Feed Material | <125-μm $^{238}PuO_2$ granules (Lot 38) seasoned at 1100°C (60 wt%) and 1600°C (40 wt%) |
| Hot Pressing Parameters | 1530°C for 15 min at 19.5 MPa |
| Post-Press Sintering | 6 h at 1000°C plus 6 h at 1527°C in Ar-$H_2{}^{16}O$ |
| Comments | 16 chamfered pellets, feed enriched at 83.5 at.% $^{238}Pu$ |

| | Dimensions | | | |
|---|---|---|---|---|
| Condition | Diam (cm) | Length (cm) | Weight (g) | Density (% TD) |
| As Pressed | 0.631 ± 0.001 | 0.933 ± 0.004 | 2.645 ± 0.002 | 85.9 ± 0.5 |
| Sintered | 0.625 ± 0.001 | 0.927 ± 0.004 | 2.664 ± 0.001 | 87.9 ± 0.5 |

TABLE IV-2

RHU PRODUCTION PELLET LOT RU4

| | |
|---|---|
| Feed Material | <125-μm $^{238}PuO_2$ granules (Lot 39) seasoned at 1100°C (60 wt%) and 1600°C (40 wt%) |
| Hot Pressing Parameters | 1530°C for 15 min at 19.5 MPa |
| Post-Press Sintering | 6 h at 1000°C plus 6 h at 1527°C in Ar-$H_2{}^{16}O$ |
| Comments | 16 chamfered pellets, feed enriched at 83.5 at.% $^{238}Pu$ |

| | Dimensions | | | |
|---|---|---|---|---|
| Condition | Diam (cm) | Length (cm) | Weight (g) | Density (% TD) |
| As Pressed | 0.630 ± 0.000 | 0.938 ± 0.003 | 2.647 ± 0.004 | 85.6 ± 0.5 |
| Sintered | 0.624 ± 0.001 | 0.929 ± 0.003 | 2.665 ± 0.004 | 88.2 ± 0.7 |

TABLE IV-3

RHU PRODUCTION PELLET LOT RU5

| | |
|---|---|
| Feed Material | <125-μm $^{238}PuO_2$ granules (Lot 39) seasoned at 1100°C (60 wt%) and 1600°C (40 wt%) |
| Hot Pressing Parameters | 1530°C for 15 min at 19.5 MPa |
| Post-Press Sintering | 6 h at 1000°C plus 6 h at 1527°C in Ar-$H_2{}^{16}O$ |
| Comments | 16 chamfered pellets, feed enriched at 83.5 at.% $^{238}Pu$ |

| | Dimensions | | | |
|---|---|---|---|---|
| Condition | Diam (cm) | Length (cm) | Weight (g) | Density (% TD) |
| As Pressed | 0.630 ± 0.000 | 0.942 ± 0.004 | 2.649 ± 0.003 | 85.4 ± 0.5 |
| Sintered | 0.624 ± 0.001 | 0.933 ± 0.003 | 2.667 ± 0.003 | 87.8 ± 0.4 |

Fabrication parameters and dimensions for pellet lots RU4 and RU5 are listed in Tables IV-2 and IV-3 . These pellets, together with 14 pellets from lot RU3, will be encapsulated for shipment when the Pt-30Rh assemblies are received.

During December, two Pt-30Rh capsules were welded, employing different power settings. These were sectioned and examined metallographically as part of the weld development effort.

B. Enhanced-Ductility Fuel

Three plutonia pellets doped with 10 mol% $Y_2O_3$ were cold pressed and sintered for 6 h at 1600°C in flowing air. The furnace temperature was programmed so that the heat-up and cool-down cycles each took 24 h. The pellets appeared to be completely crack free, based on external examination. A photograph of the three pellets is shown in Fig. IV-4. The pellets will be tested in compression to see if they have the greater strength and ductility observed in undoped $PuO_{2-x}$.

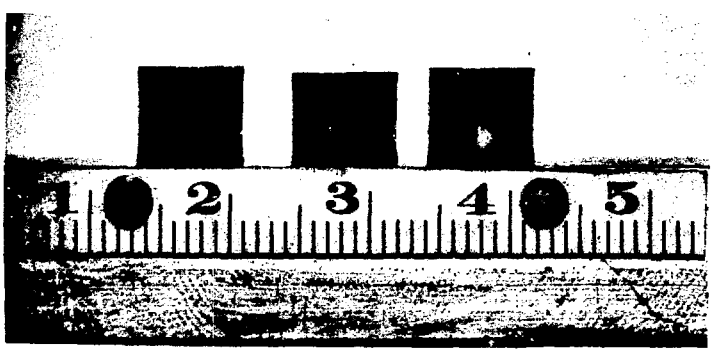



Fig. IV-4.
Three $PuO_2$-10 mol% $Y_2O_3$ pellets made by cold pressing and sintering. The O/M ratio is 1.95.

## V. FUEL PROCESSING

### A. Residues and Shipments

Residue Declaration RS-238-62 (1.0 kg $^{238}Pu$) was submitted for approval. The material will be sent when transport is available.

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C. W. Whitmore, GE, Philadelphia, PA
R. Hemler, GE, Philadelphia, PA
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V. Haley, GE, Philadelphia, PA
E. C. Krueger, Sundstrand, Rockford, IL

F. Schumann, TES, Timonium, MD
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