Monthly Progress Report
Heat Source Technology Programs
October through December 1996

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*Compiled by
T. G. George*

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MONTHLY PROGRESS REPORT
HEAT SOURCE TECHNOLOGY PROGRAMS
OCTOBER THROUGH DECEMBER 1996

Compiled by

T. G. George

ABSTRACT

This quarterly report describes activities performed in support of Cassini fueled-clad production and studies related to the use of $^{238}\text{PuO}_2$ in radioisotope power systems carried out for the Office of Special Applications of the U.S. Department of Energy (DOE) by Los Alamos National Laboratory (LANL). Most of the activities described are ongoing; the results and conclusions described may change as the work progresses.

I. HEAT SOURCE AND FEED POWDER SHIPMENTS

A shipment of 4.3 kg of ^{238}Pu was received from the Westinghouse Savannah River Company (WSRC) at the end of September. Calorimetry of the shipment began immediately and was completed by mid-October.

All available 5320 casks were returned to WSRC at the end of November.

In mid-December, shipping and receiving personnel at Los Alamos National laboratory (LANL) were informed that the delivery of Light-Weight Radioisotope Heater Units (LWRHUs) designated for the Cassini spacecraft to the Kennedy Space Center (KSC) would be required before 15 April 1997. A total of ten 9968 casks will be required for the shipment.

II. IRIIDIUM HARDWARE SHIPMENTS AND INVENTORY

No shipments of iridium hardware occurred in this quarter. The LANL inventory of

GPHS hardware, as of 31 December 1996, is shown in Table I.

Table I. LANL Inventory of GPHS Iridium Hardware as of 31 December, 1996

Type of Hardware	Category	Number of Items in Inventory
PICS*	Prime	76
PICS	Restricted Use	25
Type II weld shields	Prime	111

*PICS = postimpact containment shell.

III. FACILITIES

A. General

The laboratories used for fuel processing, fuel form fabrication, and encapsulation welding were available for routine operations throughout October and November. However, both months were occupied by preparations for a comprehensive shutdown/cleanout inventory in the $^{238}\text{PuO}_2$ processing laboratories. The inventory was conducted on 19 November.

In December, all work within the Plutonium Facility was curtailed to accommodate replacement of the Facility Control System (FCS) computer and operating system.

B. 7-in. Launcher Facility

We continued to work to close out findings resulting from the Department of Energy (DOE) operational readiness review (ORR) of the 7-in. launcher. By the end of the quarter, work had been completed on upgrading the facility fire protection system.

C. Fuel Processing Laboratories

Work on the installation of a third hot-press continued through mid-November. Design change packages (DCPs) for the next phase of installation work were prepared and submitted to facility configuration management. By the end of November, the DCPs had been reviewed by configuration management and craftworkers had been scheduled to bore a series of 4-in. diameter holes through the concrete floor to accommodate radio-frequency (RF) cable hookups for the furnace portion of the new hot-press.

At the end of December, a work order and a DCP were submitted to configuration management for removal of gloveboxes GB216 and GB218 from the primary $^{238}\text{PuO}_2$ fuel processing laboratory.

D. Aqueous Fuel Processing

By the end of December, conceptual work had been completed on a process flowsheet and footprint for $^{238}\text{PuO}_2$ aqueous recovery operations.

IV. GENERAL-PURPOSE HEAT SOURCE (GPHS) PROGRAM

A. Cassini Heat Source Production

By mid-September, all of the capsules required to meet Cassini production and testing requirements had been shipped to EG&G Mound Applied Technologies (EG&G MAT) in Miamisburg, Ohio. A summary of Cassini GPHS production is presented in Table II.

A summary of impurity analyses for residues and scrap items generated during Cassini production was submitted to DOE NE-50 in mid-December.

B. Production Support Activities: Weld Evaluation Study

Metallographic examination of example welds and unacceptable production welds continued throughout the quarter.

C. Safety Test Program

1. RTG/Thin-Fragment Test

The objective of the RTG/thin-fragment test was to provide information on the response of GPHS capsules, modules, and a loaded radioisotope thermoelectric generator (RTG) to conditions that might be experienced as a result of potential on- or near-pad accidents resulting from launch vehicle failure.

An RTG/thin-fragment test was conducted at the Sandia National Laboratories, Albuquerque (SNLA) rocket sled track on 26 March. In this test, a thin fragment was accelerated down the track where it impacted a simulated RTG section that contained GPHS modules loaded with simulant-fueled (UO_2 - ^{235}U depleted) GPHS capsules. The impact velocity was measured at 306 m/s, and the temperature of the module stack within the RTG was 1090°C.

The post-test examination and analyses of the impacted converter section and simulant-fueled capsules were documented in previous monthly reports. During this quarter, work focused on documenting the entire RTG/thin-fragment test series in a formal topical report. In October, comments on a rough draft of the report were received from various reviewers and incorporated into the final version of the report. The report was submitted for printing on 19 November 1996, and was published as report number LA-13220 in early December.

In the second half of the quarter we also worked to produce a video summary of the RTG/thin-fragment test series. A final version of the video was expected to be distributed in January 1997.

Table II. Cassini Production as of 30 September 1996

Activity	Number of Units
^{238}Pu introduced into LANL process line (cumulative, kg)^a	38.6
GPHS	
Clads shipped to EG&G MAT ^b	235
Clads welded	284
Fuel pellets pressed	315
LWRHU	
Capsules shipped to KSC ^c	0
Capsules loaded into aeroshells	180
Capsules welded to date	181
Fuel pellets pressed to date	206

^a From program start in January 1994.

^b EG&G Mound Applied Technologies.

^c Kennedy Space Center.

2. Production Qualification Test Program

The purpose of the production qualification test program was to determine whether heat sources fabricated at LANL had impact properties equivalent to those of heat sources fabricated at Savannah River and evaluated during the Galileo/Ulysses test program. Four GPHS capsules fabricated at LANL in 1994 were selected for use in the first production qualification test (PQ-1).

In the PQ-1 test, a GPHS module loaded with capsules FC0018, FC0022, FC0031, and FC0038, was aged for 200 hours at a temperature of 1287°C (clad surface). The module was then thermally treated to simulate a "min. gamma" atmospheric reentry. In an impact test conducted on 12 July, the module was launched against a steel target at 54 m/s and at 919°C.

Postimpact evaluation did not reveal any evidence of a fuel release. The impacted module was relatively intact, and capsule deformations were relatively modest. The capsule strains and preliminary posttest observations have been documented in previous monthly reports.

When capsules FC0018 and FC0022 were defueled, noncontiguous areas of a thin white deposit were observed on the exterior surfaces of both fuel pellets. Similar deposits and some adjacent discoloration were observed on the clad interiors and on the weld-shields. Small sections for microprobe and SEM analyses were removed from both weld-shields and transported to the analytical facility at LANL's Chemistry and Materials Research (CMR) building.

In mid-October, efforts to perform microprobe and scanning electron microscope (SEM) analyses of the weld-shields were suspended because of the extremely high alpha contamination of the metal samples. The samples were subsequently returned to the Plutonium Facility (PF-4), mounted, polished, and returned to the CMR for analysis. Unfortunately, alpha contamination of mounts and polished sample surfaces were again too high to permit either microprobe or SEM analysis. Consequently, the mounted samples were returned to PF-4 for a second time.

Throughout the month of November, work focused on the evaluation of various coating techniques and formulations of metal-bearing mounting material as potential masks for the high level of alpha contamination on the sample surfaces. By the end of the quarter, we had not identified any remedial actions that would effectively block alpha radiation while at the same time permitting accurate SEM or microprobe analysis.

3. Bare Clad Impact Tests

Two bare clad impact tests (BCIs) were performed at the end of November. Both tests were part of the ongoing program to determine the similarity of GPHS capsules produced at LANL for the Cassini mission to capsules produced at WSRC in the early 1980s for the Galileo and Ulysses missions.

a. BCI-33

On 20 November, GPHS capsule FC0334 was impacted against a steel plate at 55 m/s and 1093°C. Prior to impact, the capsule was pretreated in a vacuum at 1287°C (clad surface temperature) for 200 h.

Postimpact examination of the catch-tube contents revealed that the exterior of capsule FC0334 was discolored. Isolated areas of yellow discoloration were observed on the capsule vent cup, and the entire trailing face of the capsule was covered by a dark brown discoloration. The interior surface of the tantalum impact can was discolored by isolated regions of blue and purple discoloration, and a green deposit was visible on the tungsten foil used to line the capsule support cradle. No plutonia fines were observed on the interior of either the tantalum impact can or the catch tube.

Subsequent examination of capsule FC0334 revealed that the impact face of the capsule was centered approximately 15 degrees from the weld start. The capsule was breached by two transverse cracks located on the cup radii, at the edges of the impact face. The capsule strains are listed in Table III.

Comparison of the preimpact fuel pellet weight, and the weight of the plutonia recovered from capsule FC0334 indicated that a maximum of 0.214 g of plutonia may have been released.

Table III. BCI Capsule Strains

<u>Test</u>	<u>Capsule</u>	<u>Strain (%)</u>				
		<u>Axial</u> <u>max.</u>	<u>Diametral</u>			
			<u>Vent Cup</u>		<u>Shield Cup</u>	
			<u>Min.</u>	<u>Max.</u>	<u>Min.</u>	<u>Max.</u>
BCI-33	FC0334	19.6	- 13.8	10.9	- 14.8	10.8
BCI-34	FC0336	15.9	- 15.9	12.7	-14.4	12.2

b. BCI-34

On 25 November, GPHS capsule FC0336 was impacted against a steel plate at 47.6 m/s and 1090°C. Prior to impact, the capsule was pretreated in vacuum at 1287°C (clad surface temperature) for 200 h. Although the desired impact velocity for this test was 55 m/s, an unexpected amount of friction between the 7-in. gun projectile (used to accelerate the capsule) and the gun barrel reduced the velocity at impact.

Postimpact examination of the catch-tube contents revealed that capsule FC0336 was discolored on the areas that had contacted the tungsten foil used to line the support cradle. In addition, the capsule was loosely bonded to the lid of the tantalum impact can; surfaces of the tantalum can adjacent to the capsule were discolored with a thin white deposit. No plutonia fines were observed on the interior of either the tantalum impact can or the catch tube.

Examination of capsule FC0334 revealed that the impact face of the capsule was centered approximately 10 degrees from the weld start. The capsule was breached by a transverse crack located on the vent cup radius at the edge of the impact face and by a weld failure that extended from approximately 30 to 185 degrees (with the weld start at 0 degrees). However, the portion of the weld-shield under the weld failure was not breached, and effectively prevented any significant release of plutonia.

Comparison of the preimpact fuel pellet weight and the weight of the plutonia recovered from capsule FC0336 indicated that a maximum of 0.517 g of plutonia may have been released.

D. Research and Development

1. High-Silicon Fuel Characterization (HSC) Study

The purpose of the high-silicon fuel study was to determine the effect of fuel impurities on the response of the GPHS to conditions that were baselined during the Galileo/Ulysses test program. Four fuel pellets fabricated and encapsulated at LANL in 1993 and 1994 were selected for use in the first two half-module impact tests in this test series.

The two pellets selected for the first test, HSC-1, were fabricated from low-enrichment (<82% ^{238}Pu) fuel and contained relatively high concentrations of silicon (905 and 1059 ppm). The GPHS capsules containing these pellets were loaded into a graphite impact shell (GIS), which was designated GIS A. This GIS was then paired with another GIS (designated GIS B) and was loaded into a GPHS module.

The two pellets selected for the second HSC test had calculated silicon contents of 415 and 605 ppm. The GPHS capsules containing these pellets were loaded into a GIS (designated GIS D), paired with another GIS (designated GIS C), and then loaded into a GPHS module for heat treatment.

Both GPHS modules were treated in a module reduction and monitoring facility (MRMF) vacuum system. During the MRMF treatment, each module was placed in a vacuum chamber that was evacuated and then backfilled with argon. Each module was maintained at a constant temperature during the MRMF treatment. The module containing GIS A stabilized at 920°C. The module containing GIS D stabilized at 862°C. In each case, the MRMF treatment continued until a fuel stoichiometry of approximately 1.98 O/Pu was achieved.

After MRMF treatment, each module was soaked at 1074°C (module surface temperature) in a vacuum for an extended time period; this treatment temperature was calculated to give a fuel temperature of 1461°C. After the aging treatment, the modules were removed from the aging furnaces and GIS A and GIS D were loaded into GPHS half-modules for impact testing. Both impact tests (HSC-1 and HSC-2) were performed in mid-1995; the results of these tests have been presented in previous monthly reports.

The remaining two GISs (GIS B and GIS C) were reinserted in the aging furnace and heat treatment continued at 1074°C. GIS B was removed from the aging furnace during the first week of November, 1995, after a total of 235 days at temperature. The GIS was then placed in a helium-atmosphere glovebox for storage. GIS C was removed from the furnace on 8 January, 1996, after a total of 262 days at temperature. After removal from the aging furnace, GIS C was placed in storage with GIS B. The fuel capsules in GIS B

had silicon contents of 1127 and 776 ppm; the GIS C capsules had silicon contents of 336 and 325 ppm.

2. Aqueous Recovery Operations

Dissolution and precipitation operations were suspended at the beginning of October to accommodate the November shutdown/cleanout inventory.

3. Investigation Of Urania/Plutonia Simulants

In this quarter we completed the postmortem examination of the HfO_2 -fueled capsule (SC0131) that was impacted on 17 May at 974°C and 56.1 m/s. Metallographic examinations of wall sections removed from the capsule vent cup, weld, and shield cup were completed at the end of October. The average vent and shield cup grain sizes are listed in Table IV.

Table IV. Capsule SC0131 Grain Size Data

<u>Cup</u>	<u>Orientation</u>	<u>Average Grains/Thickness</u> <u>(per 0.635-mm nominal wall thickness)</u>
Vent	Axial	28.4
Vent	Transverse	24.7
Weld-Shield	Axial	26.1
Weld-Shield	Transverse	27.4

4. Accelerated Oxygen Exchange

We continued to investigate methods of accelerating the ^{16}O oxygen exchange process throughout the quarter. Previously, a thick-walled tantalum container was loaded with approximately 150 g of unexchanged $^{238}\text{PuO}_2$ and a small amount of ^{16}O water, and then welded shut. In October, the tantalum vessel was decontaminated, leak-checked, and then loaded into a stainless-steel secondary containment vessel. The assembly was then submitted for neutron emission rate (NER) measurement. The NER measurements indicated that in the 22.5 months that elapsed from the time of container loading, the fuel NER had dropped from 13,000 neutrons/s-g ^{238}Pu to 8,000 n/s-g. This result suggests that long-term storage of fuel in contact with ^{16}O water might preclude the need for oxygen exchange treatment during fuel processing.

Although we had originally expected to complete the ^{16}O exchange by heating the sealed vessel at 400°C for approximately 1 hour in the hot isostatic press (HIP), a number of equipment issues and the shutdown/cleanout inventory delayed processing until after the start of the new calendar year.

E. Project Management

Primavera updates were received from Orbital Sciences Corporation (OSC) at the beginning of each month. Three monthly status reports for the Cassini program were submitted to OSC on schedule.

V. LIGHT-WEIGHT RADIOISOTOPE HEATER UNIT (LWRHU) PROGRAM

A. Cassini Heat Source Production

A total of 180 LWRHU capsules were previously loaded into aeroshells, welded into product cans, and transferred to the PF-4 vault for storage. By the end of December, a final schedule for shipment of the product cans to Kennedy Space Center (KSC) had not been established.

On 18 December, representatives from the Westinghouse Applied Technology Business Area (WATBA) reviewed the Cassini LWRHU heat source fabrication data packages. A subsequent review and approval by representatives from the DOE Albuquerque Operations Office (DOE/ALO) was scheduled for the second week in January, 1997.

B. LWRHU Production Qualification

On 28 June, an LWRHU heat source was impacted against a steel target plate at 53.5 m/s. The test was conducted at ambient temperature in the 7-in. launcher. Preliminary results of the postmortem examination were documented in previous reports.

At the beginning of this quarter, the impacted LWRHU aeroshell was disassembled. There was no discernible evidence of the impact on the surface of the LWRHU capsule.

C. LWRHU Safety Testing: Sequential Impact Tests

In July and August, a series of sequential impact tests were performed on simulant-fueled LWRHUs to determine energy thresholds for the destruction of various LWRHU components. These tests were performed to provide data for the Cassini final safety analysis report (FSAR), and were documented in previous monthly reports. In this quarter, we worked to prepare a topical report that would formally document the results of these tests.

VI. 60-WATT HEAT SOURCE

Heat source 16-040 (containing capsule FC0029) was placed in an aging furnace under vacuum at 650°C on 14 December 1995, and reached thermal equilibrium on 15 December. The heat source was removed from the aging furnace on 9 July 1996, and

was submitted for radiographic examination. On 26 July 1996 the heat source was returned to the furnace for an additional six months of heat treatment at 650°C.

A Primavera update was received from OSC at the beginning of the quarter.

VII. 1-kW SHIPPING CONTAINER

The LANL inventory of 1-kW hardware, as of 31 December 1996 is shown in Table V.

Table V. LANL Inventory of 1-kW Hardware as of 31 December 1996

<u>Type of Hardware</u>	<u>Number of Items</u>
Primary containers	4
Primary lids	2
Secondary containers	2
Secondary lids	2
Product cans	28

VIII. PROGRAMMATIC MEETINGS AND VISITORS

Quality assurance personnel from DOE/NE-50 and WATBA were on site for the first week in October.

Four LANL technical staff members attended the Cassini quarterly program review held on 30 October at OSC in Germantown, Maryland.

On 30 October, three personnel from the DOE Office of the Inspector General toured the 7-in. launcher addition in building PF-4.

On 12 November, three LANL technical staff members attended a meeting on WSRC ²³⁸Pu aqueous recovery capabilities held in Aiken, South Carolina.

On 20 November, a representative from the Defense Nuclear Facilities Safety Board (DNFSB) toured the ²³⁸PuO₂ operations area and was briefed on continued heat source production activities and capability maintenance.

On 19 November, LANL hosted a review of ²³⁸Pu aqueous recovery capabilities at LANL's Plutonium Facility.

On 4 December, three LANL technical staff members attended a review of ORNL ^{238}Pu production and aqueous recovery capabilities, held in Oak Ridge, Tennessee.

On 18 December, representatives from the Westinghouse Applied Technology Business Area (WATBA) reviewed the Cassini LWRHU heat source fabrication data packages.