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General-Purpose Heat Source: Research and Development Program

Process Evaluation, Fuel Pellet GF-47

M. A. H. Reimus
T. G. George

Los Alamos
NATIONAL LABORATORY
Los Alamos, New Mexico 87545

GENERAL-PURPOSE HEAT SOURCE: RESEARCH AND DEVELOPMENT PROGRAM

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by

M. A. H. Reimus and T. G. George

ABSTRACT

The general-purpose heat source (GPHS) provides power for space missions by transmitting the heat of ^{238}Pu decay to an array of thermoelectric elements. Because the potential for a launch abort or return from orbit exists for any space mission, the heat source must be designed and constructed to survive credible accident environments. Previous testing conducted in support of the Galileo and Ulysses missions has documented the response of the GPHS heat source to a variety of fragment-impact, aging, atmospheric reentry, and Earth-impact conditions. Although heat sources for previous missions were fabricated by the Westinghouse Savannah River Company (WSRC), GPHS fueled-clads required for the Cassini mission to Saturn will be fabricated by Los Alamos National Laboratory (LANL). This evaluation is part of an ongoing program to determine the similarity of GPHS fueled clads and fuel pellets fabricated at LANL to those fabricated at WSRC. Pellet GF-47, which was fabricated at LANL in late 1994, was submitted for chemical and ceramographic analysis. The results indicated that the pellet had a chemical makeup and microstructure within the range of material fabricated at WSRC in the early 1980s.

I. INTRODUCTION

The general-purpose heat source (GPHS) is a modular component of the radioisotope thermoelectric generators (RTGs) that will provide power for NASA's Cassini mission to Saturn. An RTG generates electric power by using the heat of ^{238}Pu a-decay to create a temperature differential across a thermoelectric array. Each RTG is loaded with 18 GPHS modules, and each GPHS module (Fig. 1) contains four $^{238}PuO_2$ fuel pellets that provide a total thermal output of 250 W. Each fuel pellet is encapsulated in a vented, DOP-26 iridium alloy shell. Two capsules are held in a fineweave-pierced fabric (FWPF) graphite impact shell (GIS), and two GISs are contained within a FWPF aeroshell.

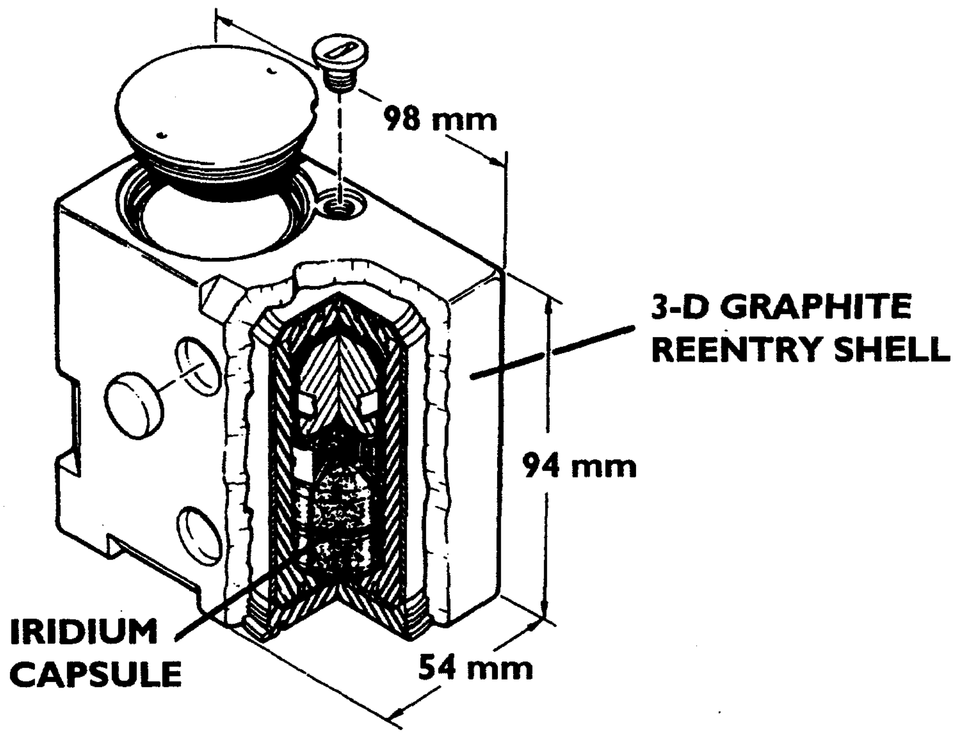

Fig. 1. GPHS module.

Because the potential for a launch abort or return from orbit exists for any space mission, the GPHS heat source has been designed and constructed to survive credible accident environments. Previous testing conducted in support of the Galileo and Ulysses missions has documented the response of the GPHS heat source to a variety of fragment-impact, aging, atmospheric reentry, and Earth-impact conditions. Heat sources for previous space exploration missions were fabricated by the Westinghouse Savannah River Company (WSRC); GPHS fueled clads required for the Cassini mission to Saturn will be fabricated by Los Alamos National Laboratory (LANL). The evaluation described in this report is part of an ongoing program to determine the similarity of GPHS fueled clads and fuel pellets fabricated at LANL to those fabricated at WSRC in the early 1980s.

II. BACKGROUND

GPHS fuel pellet GF-47 was produced early in LANL's Cassini production effort. The pellet was fabricated from LANL fuel lots 375 and 369. Fuel lot 375 was composed of material that had been low-fired at 1100°C for 6 h in an argon atmosphere saturated with water vapor ($H_2{}^{16}O$); fuel lot 369 was high-fired at 1600°C for 6 h in a similar atmosphere. Additional information on the fuel lots used for fuel pellet GF-47 is presented in Table I.

TABLE I. Fuel Lots Used in Pellet GF-47				
LANL Lot #	**WSRC Container**	**WSRC Run #**	**WSRC Precipitation Date**	**% of Pellet GF-47**
369	1959	41-2	3/6/94	40.0
375	1998	38-3	2/9/94	60.0

After granule seasoning, the high- and low-fired fuel fractions used in pellet GF-47 were mixed and then loaded into a hot press. The pellet was hot-pressed on 8 November 1994 and sintered first at 1000°C for 6 h and then at 1527°C for an additional 6 h. After sintering, the pellet was placed in storage pending completion of non-destructive test (NDT) operations. The results of these tests are summarized in Table II.

Unfortunately, before the pellet could be encapsulated it cracked into several pieces and could not be loaded into a GPHS clad/vent set. The interior of the pellet was subsequently sampled for chemical analysis and ceramography; the pellet remains were returned to WSRC for reprocessing.

Table II. Pellet GF-47 Attribute				
Sintered Length (mm)	**Sintered Diameter (mm)**	**Weight (g)**	**Density* (%, theoretical)**	**O/Pu Ratio**
27.850	27.440	151.114	80.9	1.983

*Calculated on the basis of a theoretical density of 11.46 g/cc

III. RESULTS

A. Chemical Analyses

One sample from the interior of pellet GF-47 was submitted for DC-arc spectroscopy. The results of analyses performed on two aliquots split from this sample are listed in Table III. Because of the analytical workload associated with ongoing Cassini production, a phosphorus analysis of pellet GF-47 (which would have required an additional sample) was not requested.

Table III. GF-47 Impurity Levels (ppm)			
Element	Calculated Levels*	Measured Levels	
		Run 1	Run 2
Aluminum	141	89	110
Boron	5	<5	<5
Beryllium	1	<1	<1
Calcium	15	7	7
Cadmium	10	<10	<10
Chromium	114	80	84
Copper	4	1	1
Iron	221	400	410
Magnesium	26	39	40
Manganese	11	17	19
Molybdenum	20	<20	<20
Sodium	50	<50	<50
Nickel	61	61	55
Phosphorus	21	— Not Analyzed —	
Lead	10	<10	<10
Silicon	277	810	850
Tin	5	<5	<5
Zinc	5	<5	<5
Total Impurities	976	1610	1682

* Calculated on the basis of chemical analyses of granules from fuel lots 369 and 375, weighted to reflect the percentage of each lot in the fuel pellet.

B. Ceramography

One of the samples obtained from the interior of pellet GF-47 was submitted for ceramographic examination. The sample, which had an approximate weight of 0.5 g, was mounted in epoxy loaded with alumina particles (Fig. 2). After the epoxy had cured, the mounted sample was abraded with a 45-mm diamond wheel and then ground with a series of SiC papers. Initial polishing was completed using an automated turntable and 5-mm and then 1-mm diamond pastes. Final polishing was completed on a vibratory table using a slurry of 0.03-mm alumina.

After sample preparation, the as-polished pellet fragment was submitted for examination (Fig. 3). Following this examination the fragment was etched using a solution of three acids: 95% HBr, 3% HCl, 2% HF (Fig. 4).

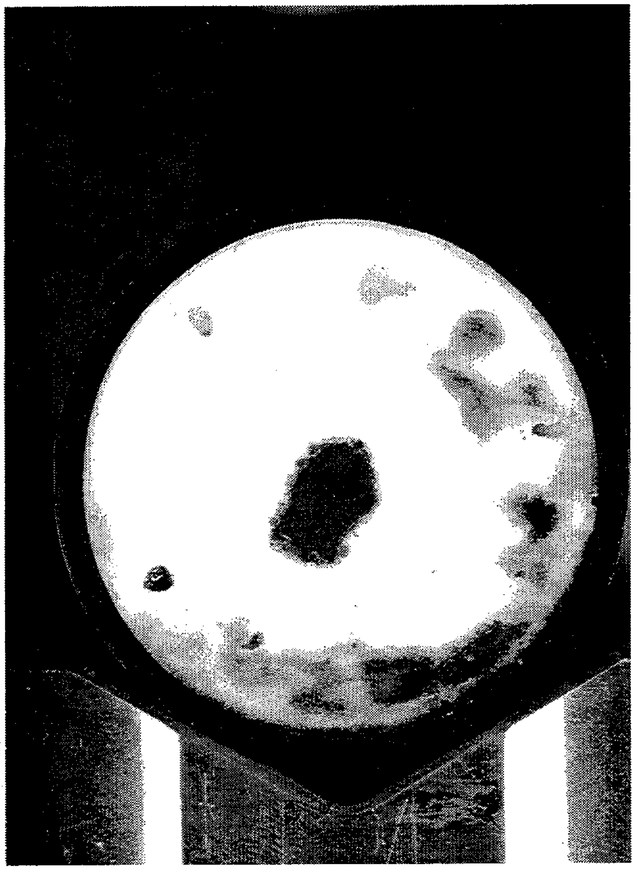

Fig. 2. A fragment from the interior of pellet GF-47, mounted in epoxy for ceramographic examination. (NMT-9 Neg. 95-49)

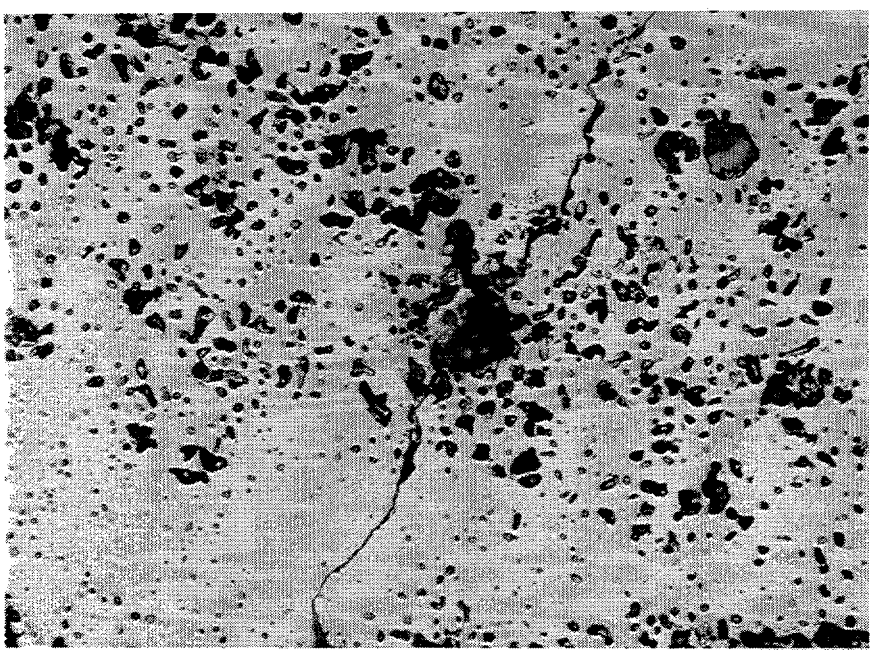

Fig. 3. Representative microstructure of the GF-47 pellet fragment; as polished, at 375×. (NMT-9 Neg. 95-76)

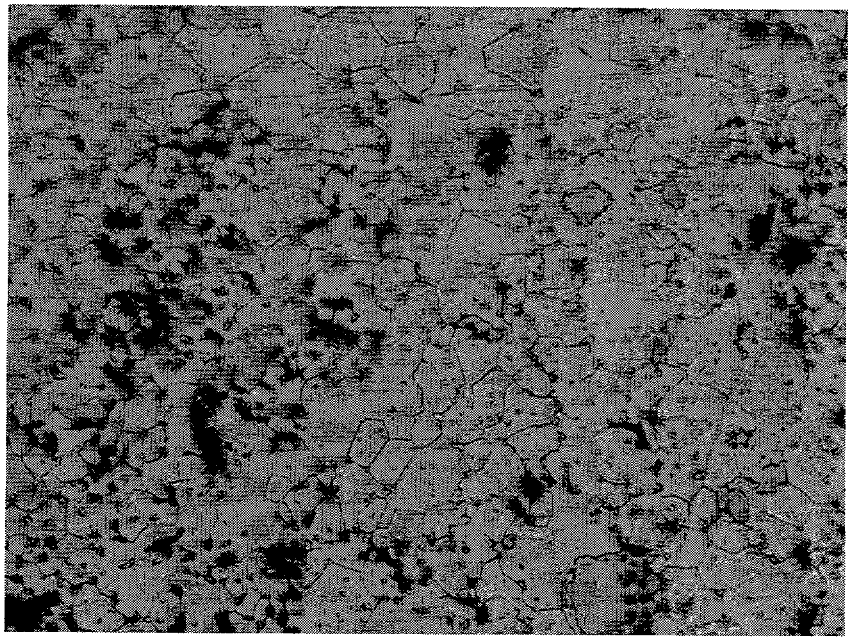

Fig. 4(a)

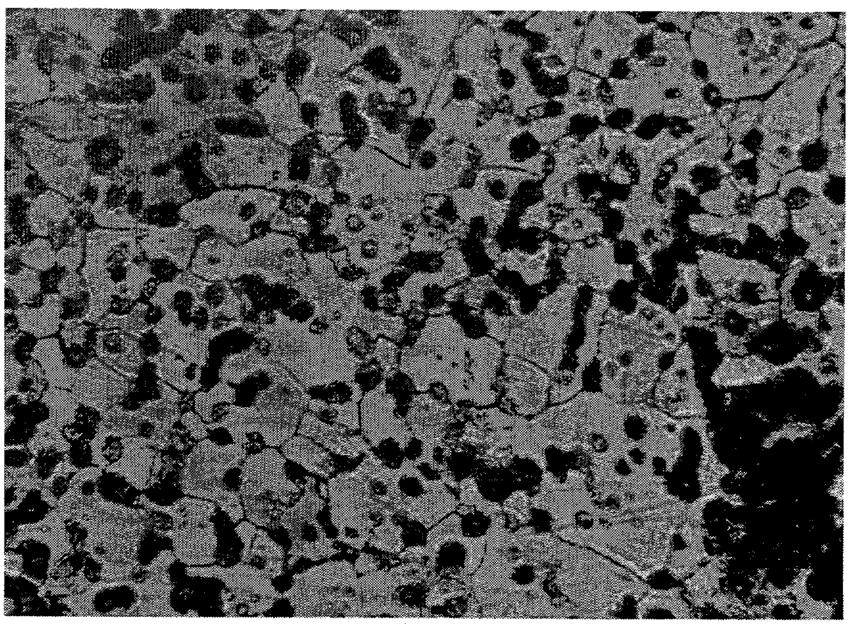

Fig. 4(b)

Fig. 4. Representative microstructure of the GF-47 pellet fragment; etched, (a) at 250× and (b) at 500×. (NMT-9 Negs. 958-81, 958-82)

IV. DISCUSSION

The physical attributes of pellet GF-47 are generally similar to those of pellets pressed at WSRC in the 1980s to support the Galileo and Ulysses missions. However, because the pellets fabricated at LANL for the Cassini mission had slightly greater weight (nominally ≈ 151.0 g) and the same dimensions as those fabricated for previous missions, the density of pellet GF-47 was slightly greater (on a basis of theoretical density) than the densities of most pellets fabricated in the early 1980s.

Even with its slightly increased density the microstructure of pellet GF-47 did not appear to be significantly different from the appearance of pellets pressed for the design iteration test (DIT) series conducted in 1983 and 1984.[1–5] Apart from a subtle difference in general appearance, possibly attributable to differences in sample preparation, enrichment, or postpressing history, the microstructure of pellet GF-47 appeared to be within the range of pellet microstructures evaluated previously. It should be noted, however, that the majority of the DIT pellets used for comparison were subjected to additional treatments, such as encapsulation welding, storage, pretest aging (at ≈1287°C for 30 days), and impact testing.

The results of spectroscopic analyses of samples from pellet GF-47 were generally consistent with each other and within the range of calculated impurity contents. The most-significant exceptions were between actual and calculated values for silicon, aluminum, and iron.

The aluminum content of pellet GF-47 was approximately 30% lower than expected. Although this may reflect uncertainties associated with the DC-arc spectroscopy technique, it may also indicate that some fraction of the aluminum is removed during hot pressing and/or sintering. However, the relative proximity of both calculated and actual aluminum values would most likely support the first premise.

The calculated values for both iron and silicon were significantly lower than the analytical results.

In the case of iron, the difference may be the result of contamination by processing equipment or gloveboxes, all of which are composed of stainless steel. However, the actual chromium content of the GF-47 pellet was slightly less than expected, while the nickel content was identical to the the calculated value.

The apparent increase in silicon content between granule seasoning and pellet fabrication would appear to be caused by one of four possibilities:

- The granules were contaminated with silicon between sampling and loading into the hot press die.
- The hot pressing and sintering operations are a source of significant silicon contamination.
- Significant gradients in silicon content exist in the as-received fuel and persist through pellet fabrication.
- Uncertainties associated with DC-arc spectroscopic analyses resulted in the apparent increase.

Because several previous evaluations of LANL fuel-processing operations, conducted by both external and LANL personnel, failed to reveal any potential sources of silicon contamination, it is unlikely that the listed discrepancies in actual and calculated silicon content are related to an in-line source of silicon contamination. Similarly, it is unlikely that any inhomogeneities could persist in the fuel through ball milling. Consequently, it appears that uncertainties associated with the DC-arc analytical technique are the most credible explanation for the apparent difference in actual and calculated silicon contents.

LANL analytical personnel have previously indicated that results obtained by DC-arc spectroscopy, particularly for high-content (>100 ppm) impurities such as silicon, should be considered as having an accuracy of a factor of 2. That is, at a very-high confidence level, the reported elemental value may be as much as twice, or as little as half, the actual amount of impurity in the fuel sample. Although the calculated and actual values for silicon in pellet GF-47 were barely within this range, examination of the granule analyses for fuel lot 375 (which made up 60% of pellet GF-47) revealed an average silicon content of 355 ppm.

V. CONCLUSIONS

No obvious or significant differences appear to exist between the microstructure of pellet GF-47 and the microstructures of pellets pressed at WSRC in the early 1980s. The impurity contents, calculated on the basis of granule analyses, were generally in good agreement with the actual impurity levels detected in pellet GF-47. The most significant discrepancy, silicon, fell within the range of analytical values associated with the DC-arc spectroscopy technique.

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