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Space Nuclear-Safety Program

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SPACE NUCLEAR SAFETY PROGRAM

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Compiled by
S. E. Bronisz

ABSTRACT

This technical monthly report covers studies related to the use of $^{238}\text{PuO}_2$ in radioisotope power systems carried out for the Office of Special Nuclear Projects of the US Department of Energy by Los Alamos National Laboratory. Most of the studies discussed here are ongoing. Results and conclusions described may change as the work continues.

I. GENERAL-PURPOSE HEAT SOURCE

A. Early Compatibility Tests (D. Pavone)

1. ECT-1. In the first early compatibility test (ECT-1), an impact assembly was aged at 1310°C for 6 months and impacted at 930°C and 59.3 m/s. Both fueled clads failed. As reported previously,^{1,2} the grain sizes of the vented cups of each assembly were nonuniform. In the areas of the failures, the grains were quite large, but at other locations, duplex structures of large and small grains were present.

Five additional samples of the vent cups were examined by Auger electron spectroscopy (AES). Absorbed-current images taken in the AES unit are shown in Fig. 1. Three of the samples were taken from HF-2, the capsule that failed more severely, and two were taken from capsule HF-5. The samples were chosen on the basis of their grain sizes. The results of the AES analyses are listed in Table I. The four large-grained and moderately large grained samples contained sulfur, but the single fine-grained sample and the four fine-grained samples examined in October² did not contain sulfur. Three of the larger grained samples contained carbon and all four of them contained oxygen. Only a single occurrence of carbon and oxygen was found in the fine-grained samples.²

Thorium concentrations at grain boundaries are usually reduced during high-temperature exposures to $^{238}\text{PuO}_2$. The column of Th/Ir intensity ratios (Table I) shows that thorium depletion occurred in the ECT-1 samples, too. The sample with the largest grains (HF-2-IFF-1) had the greatest depletion, but there is no general correlation between thorium depletions and grain size.

Photomicrographs of sections through the vent assemblies of the two ECT-1 capsules are shown in Fig. 2. In each case, there was a reaction zone around the vent exit. The zones were 4.8 and 6.3 mm in diameter on HF-2 and HF-5, respectively.

At higher magnification (Fig. 3), we can see that the reaction proceeded along grain boundaries and along crystallographic planes in the grains. Analysis of the reaction products with the electron microprobe showed that the lighter gray phase was primarily iridium and silicon with small amounts of copper. The darker gray phase contained iridium, plutonium, silicon, and copper.

Nonmetallic deposits with the characteristic spacing of the holes in the floating membrane that separated the capsules during aging were present on the exterior surface near the vent in capsule HF-2. A section through one of these deposits (Fig. 4) showed it to consist of two phases. The lighter material contained plutonium, calcium, titanium, aluminum, and oxygen. The darker phase

TABLE I. Supplemental AES Results for ECT Iridium Cups (LR234-1 and -3)^a

Location	Th ₆₅ /Ir ₂₂₉	P ₁₂₀ /Ir ₂₂₉	C ₂₇₀ /Ir ₂₂₉	O ₅₁₀ /Ir ₂₂₉	Si ₉₂ /Ir ₂₂₉	S ₁₅₀ /Ir ₂₂₉	B or Cl ₁₈₀ /Ir ₂₂₉	Th ₆₅ /Ir ₅₄
ECT/HF-2, BSF-1^b (fine grain)								
Interior edge	0.18	---	---	---	---	---	---	0.02
Center	0.41	---	---	---	---	---	---	0.04
Exterior edge	0.74	---	---	---	---	---	---	0.08
Average	0.44							0.05
ECT/HF-2, BSF-2 (very large grain)								
Interior edge	0.63	---	---	0.88	---	0.15	---	0.06
Center	0.46	---	---	---	---	---	---	0.05
Exterior edge	0.04	---	---	0.49	---	0.22	---	Trace
Average	0.34			0.46		0.12		0.04
ECT/HF-2, IFF-1^c (very large grain)								
Interior edge	Trace	---	0.50	0.55	---	0.09	---	Trace
Center	0.09	---	0.49	0.77	---	Trace	---	0.02
Exterior edge	0.20	---	0.51	0.71	---	0.20	---	0.02
Average	0.10		0.50	0.68		0.10		0.01
ECT/HF-5, VFF-2^d (moderately large grain)								
Interior edge	0.19	---	1.90	0.84	---	---	---	0.04
Center	0.54	---	1.92	1.29	---	0.15	---	0.07
Exterior edge	0.38	---	1.65	1.81	---	---	---	0.06
Average	0.37		1.82	1.31		0.05		0.06
ECT/HF-5, VFF-1 (moderately large grain)								
Interior edge	0.36	---	---	0.67	---	0.02	---	0.04
Center	0.54	---	0.59	---	---	---	---	0.05
Exterior edge	0.95	---	0.65	1.57	---	0.11	---	0.09
Average	0.62		0.41	0.75		0.04		0.06

^aBlank columns indicate element not detected.

^bBSF indicates backside fracture.

^cIFF indicates impact-face fracture.

^dVFF indicates vent-face fracture.

contained calcium, aluminum, plutonium, silicon, oxygen, and a minor amount of titanium.

There were deposits in the vent structures of the capsules. Iron, chromium, and oxygen were in a deposit at the vent entrance in HF-2, and a deposit containing calcium, iron, silicon, plutonium, and small amounts of titanium, manganese, and magnesium was found in the filter element of the HF-5 capsule.

The microstructure of the iridium capsule wall near the vent assembly was made up primarily of very large grains (Fig. 5). The presence of large grains below the reaction layer strongly suggests that a vaporized impurity from the fuel passed through the vent, was reduced by the graphite, and mobilized the iridium grain boundaries. No AES analyses were made of these large grains, so we don't know if sulfur was present, as it was in the other large-grained areas.

The effect of sulfur contamination on grain growth and ductility of the iridium alloy has not been determined. It would not be astonishing if sulfur enhanced the grain growth and reduced the ductility of iridium because phosphorus is known to do both.

2. ECT-2. The aging of the second early compatibility test, ECT-2, was started. This test assembly is the same design as that used in ECT-1, an impact assembly in a graphite block. The fueled clads were numbered 8007-HF-8 and 8007-HF-9. In each case, the shield cup is from the same LR234 iridium lot used to make the vent cups in ECT-1; thus the ECT-2 exposure should allow us to determine whether the aberrant grain structure was inherent in the ECT-1 LR234 cups or was caused by fuel impurities. The aging conditions for the second test will be 1310°C and 90 days. On February 1, the total time at temperature reached 504 h.

B. Phosphorus Effects Experiment (D. Pavone)

The fuel was removed from the four fueled clads, and the analyses have been started. There was only a moderate amount of $^{238}\text{PuO}_2$ deposited on the inner surfaces of the capsules (Fig. 6). The longitudinal cut visible at the back of each capsule locates the side of the capsule facing the opposite capsule in the aging assembly.

The fuel pellets appeared normal except for PEF-49, which was two-toned gray (Fig. 7). The microstructures of samples taken from the surface and core of each pellet are shown in Figs. 8-15. Pellets PEF-44 and -45 (nominal composition outgassed and unoutgassed, respectively) were finer grained (Figs. 8-11) than pellet PEF-48 (Figs. 12 and 13), which had been doped with phosphorus and outgassed. Only pellet PEF-49 contained visible impurity phases (Figs. 14 and 15). These tended to occur near the external surfaces (Fig. 14) or near surfaces formed by internal cracks (Fig. 15). PEF-49 will be etched after it has been examined with the electron microprobe to determine the constituents of the impurity phase.

The bulk chemical analyses of the fuel pellets are listed in Table II. Only those elements present in amounts exceeding the detection limit of the emission spectrograph are listed. The phosphorus was determined by a colorimetric method. The results show that the four pellets used in this study were significantly purer than those made by the Savannah River Plant's production process. Of particular interest is the low iron content in

PEF-49. This pellet was doped with iron, and a Spark-Source Mass Spectrometry (SSMS) analysis of a piece of the as-fabricated pellet found 10,000 ppm iron. It will be interesting to see if the effusate mound at the vent exit of PEF-49³ is iron-rich; it will be analyzed by SSMS at Oak Ridge National Laboratory.

C. Overpressure Tests (R. W. Zocher)

The general-purpose heat source overpressure tests in the Sandia National Laboratories' shock tube were delayed because of problems with the furnaces used to preheat the module. The furnace elements are a standard commercial design for use in an oxidizing environment. During attempts to run them in the nitrogen atmosphere that will be used in the shock tube to protect the graphite, the furnaces failed, presumably from the formation of a volatile chromium nitride. This problem was mitigated by oxidizing the furnace elements before assembling the furnace.

In the first attempt to complete the 1.3-MPa (200-psi) test, the cover plate that protects the lowered furnace from the blast was activated before the furnace was dropped. That ended the attempt to complete the test. The problem was in the sequence wiring.

A second attempt to complete the test was successful. The recovered module's only apparent damage was a chipped corner. The module will be used for a test at 5.2 MPa (800 psi).

TABLE II. Emission Spectrographic Analysis of Fuel Pellets Used in the Phosphorus Effects Experiment

Elements ^a	Impurity Content (ppm)			
	PEF-44	PEF-45	PEF-48	PEF-49
Silicon	35	30	50	170
Aluminum	60	60	65	190
Calcium	15	15	15	200
Iron	30	20	25	7
Tantalum	100	100	100	<100
Titanium	10	7	10	8
Manganese	1	1	2	1
Phosphorus ^b	5	5	20	35

^aElements not listed were below detection limits in all four pellets.

^bPhosphorus was determined by a colorimetric method.

D. Impact Working Group (R. W. Zocher)

After the failures in the DIT-5 test,⁴ the Department of Energy established a working group to determine the reason for the failures. The initial organizing meeting was held in December at Fairchild Industries, and the first working meeting was held at Los Alamos on January 11-13, 1983.

Four possible explanations of the DIT-5 failures were identified:

- the test components were defective;
- the pre-impact conditioning was incorrect and degraded the materials;
- the test conditions were incorrect, and the sample was overstressed; or
- the module design provided insufficient impact protection.

The working group found that the components were not defective and that the pre-impact conditioning was correct. They also found that the impact velocity of 60.5 m/s was slightly higher than the nominal range of 58 ± 2 m/s, but that this deviation was not the causal factor in the failures. They concluded that the response of the module in the DIT-5 impact was in the nominal range of expectation and that perhaps 50% of the fueled clads in modules impacted in the flat-on orientation would fail.

II. LIGHT-WEIGHT RADIOISOTOPE HEATER UNIT (R. E. Tate)

Forty-two of the latest batch of 102 pyrolytic-graphite insulator sleeves were rejected for use in flight units. The most prevalent defect was insufficient stand-off height. Mound Facility is examining the possibilities of replacing or reworking the deviant items. This problem will not affect the scheduled delivery date.

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(a) HF-2-BSF-1.

(b) HF-2-BSF-2.

(c) HF-2-IFF-1.

(d) HF-5-VF-1.

(e) HF-5-VFF-2.

Fig. 1. Absorbed-current AES images of ECT-1 iridium clads.

(a) HF-2.

(b) HF-5.

Fig. 2. Photomacrographs of cross sections of the vent assemblies of the ECT fueled clads showing the extent of reaction on the exterior surfaces. Both $\sim 7X$.

(a) HF-2.

(b) HF-5.

Fig. 3. Intermetallic reaction products on the exterior of the vent assemblies of the ECT fueled clads. The light-gray phase contained primarily iridium and silicon with small amounts of copper. The medium-gray phase contained plutonium in addition to the iridium, silicon, and copper. Both 250X.

Fig. 4. Nonmetallic deposits on the exterior of the vent assembly of fueled clad HF-2. The light-gray phase contained plutonium, calcium, titanium, aluminum, and oxygen. The dark-gray phase contained calcium, aluminum, plutonium, silicon, oxygen, and a minor amount of titanium. 250X.

(a) HF-2.

(b) HF-5.

Fig. 5. Typical microstructures of the iridium cladding in the vicinity of the vent of the iridium fueled clads of the ECT test. Both 50X.

(a) PEF-44.

(b) PEF-45.

(c) PEF-48.

(d) PEF-49.

Fig. 6. Photographs of the interior of the Savannah River Laboratory phosphorus experiment fueled clads.

Fig. 7. Photograph of the plutonium pellet of PEF-49. Note that some areas had a lighter gray appearance than the bulk of the pellet.

(a) 10X.

(b) 100X.

Fig. 8. Microstructure of the surface region of plutonia pellet PEF-44.

(c) 250X.

(a) 10X.

(b) 100X.

(c) 250X.

Fig. 9. Microstructure of the core region of plutonia pellet PEF-44.

(a) 10X.

(b) 100X.

Fig. 10. Microstructure of the surface region of plutonia pellet PEF-45.

(c) 250X.

(a) 10X.

(b) 100X.

(c) 250X.

Fig. 11. Microstructure of the core region of plutonia pellet PEF-45.

(a) 10X.

(b) 100X.

Fig. 12. Microstructure of the surface region of plutonia pellet PEF-48.

(c) 250X.

(a) 10X.

(b) 100X.

(c) 250X.

Fig. 13. Microstructure of the core region of plutonia pellet PEF-48.

(a) 10X.

(b) 100X.

Fig. 14. As-polished microstructure of the surface region of plutonia pellet PEF-49.

(c) 250X.