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Progress Report

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Space Nuclear Safety Program

September 1983

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S. E. Bronisz

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SPACE NUCLEAR SAFETY PROGRAM

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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. Safety-Verification Impact Tests

1. SVT-1 and SVT-2. These two General-Purpose Heat Source (GPHS) modules have been designated for impact tests in the minimum-gamma mode. The four impact assemblies were aged together in the 6-in. vacuum furnace for 200 h at an iridium clad temperature of 1287°C . The aged assemblies are stored in a helium atmosphere in the furnace until needed for the impact tests. There were no anomalies during the aging period.

2. SVT-3. The accumulated aging exposure time for this test assembly was 1829 h on October 1, 1983. Continuous monitoring of the helium released from the aging assembly has not indicated any period of vent plugging during September. The last plugging occurred at 689 h.

3. SVT-4. On October 1, 1983, the accumulated exposure time for this test assembly reached 892 h. Continuous monitoring of the helium released from the aging assembly has not indicated any evidence of vent plugging thus far in the aging exposure.

4. SVT-5. This test module was shipped to Los Alamos as a complete assembly. The impact assemblies were removed from the aeroshell and inspected. No ab-

normalities were detected. The 90-day aging exposure of the impact assemblies was initiated on September 7, 1983. The aging will be completed at a clad temperature of 1187°C . On October 1, 1983, the accumulated exposure time was 395 h.

B. Nominal Impact Test (NIT-1)

This test assembly consisted of a partial GPHS impacted at conditions corresponding to nominal, orbital-decay reentry parameters. The impact assembly and the insulation components were aged at a clad temperature of 1287°C for 38 days. The aging time was slightly greater than the 30-day exposure of test DIT-5, which was impacted at 110% of the nominal orbital-decay reentry velocity.

The components of the NIT-1 test assembly are identified in Table I. Except for the ablation simulation machined from the aeroshell, all the components were made to specifications.

Radiographic inspection indicated that the plutonia fuel pellet in HF-450 was intact following the aging exposure but had a fine axial crack following the simulated reentry pulse to 1375°C (clad). There was a major fracture at an angle of approximately 50° to the axis across the corner of the plutonia pellet in fueled clad HF-438 before the impact test.

TABLE I. NIT-1 Test Assembly Components

Components	Service No.
Aeroshell	Special
Impact shell	PGL-099
Insulation body	C60-1
Insulation cup 1	P-12-2-22
Insulation cup 2	P-12-12-29
Fueled clad (closure end)	HF-450
Vent cup	S32-1
Shield cup	S30-6
Fueled clad (blind end)	HF-438
Vent cup	MERR40-2
Shield cup	MERR40-3

The impact test was conducted in a "face-on" orientation ($\alpha = 0^\circ$) against a hardened steel target at a clad temperature of 975°C and a velocity of 53.9 m/s.

Upon recovery, the aeroshell of the test assembly had a zig-zag longitudinal crack on the impact face and longitudinal cracks on both sides. The back side was apparently undamaged. The aeroshell closure cap was not retained in the aeroshell body. The impact shell was cracked longitudinally down the center of the impact face for about 90% of its length and had a slightly bowed profile. Its closure cap was retained by the threads of the shell body.

The fueled clads removed from the graphite impact shell were unbroken (Fig. 1). HF-450 was evenly deformed, with no severe local deformations (its fuel pellet was intact after the reentry-heating exposure). The overall deformation of HF-438 was similar, but it also had an area of severe local strain on the impact face caused by fuel push-through (its fuel pellet was cracked). The clad deformations are listed in Table II.

The total plutonium content of the impact shell, determined by radiochemical analysis of the ash resulting from combustion of all the graphite, was 9.3 mg. This amount seems to be rather high considering the absence of failures of the iridium cladding and lack of any visible effusate from the vent assemblies.

Metallographic examination of the iridium cups that made up the fueled clads revealed generally fine-grained microstructures with a slight tendency toward larger grains near the interior surfaces. Figure 2 illustrates

typical transverse cross-section microstructures. Several small cracks, similar to that shown in Fig. 2b, were observed in the metallographic cross sections examined. Their depth was limited to 2 to 3 grains, or about 100 μm . The average grain sizes of the iridium clads in both axial and transverse cross sections are listed in Table III.

Except for the HF-438 vent cup, which had finer and more asymmetric grains, the microstructures were similar. In all cases, the grain sizes were finer than those we saw in the past.

A metallographic cross section of HF-438 taken across the weld and approximately normal to the fuel fragment push-through on the impact face is shown in Fig. 3. The reduction in thickness in the weld is about 13%, but there is no evidence of failure.

The results of Auger electron spectroscopy analysis (AES) of the grain boundaries of the iridium clads are presented in Table IV. The thorium/iridium intensity ratios observed are about 50% less than those normally observed in the DOP-26 alloy. Carbon and oxygen were detected in all samples. Low levels of sulfur were found in five of the samples and boron or chlorine was present in two samples. In view of the relatively low level of thorium segregation at the grain boundaries, it is rather surprising that the grain size was not larger than that observed.

Ceramographic examination of plutonia samples of both fuel pellets indicated no abnormalities. The phosphorus content of pellet HF-438 was 12 ppm and that of HF-450 was 8 ppm.

II. LIGHTWEIGHT RADIOISOTOPIC HEATER UNIT (LWRHU)

A. Seawater Exposure (C. Land and J. Patterson)

An LWRHU, RHU-9-07, was held at 69 MPa (10,000 psi) for 663 days in 10°C simulated seawater to determine what effect the deep-ocean environment would have on it. The exposure had no apparent effect on the unit.

The seawater in the test chamber was sampled weekly, while the pressure was maintained, and analyzed for plutonium. The results of the weekly analyses are given in Table V, where they are compared with those for the analyses of the seawater used in the ambient-pressure experiment reported last month.¹ In both cases, the plutonium in individual samples varied considerably. Early in the exposure of the high-pressure LWRHU, a sharp spike in the release occurred that was not seen in

TABLE II. Postimpact Deformations of NIT-1 Fueled Clads

Location	Strain (%)	
	HF-438	HF-450
Diameter	+8.7	+8.9
Height	-9.0	-7.5
Length	+4.3	+4.6

TABLE III. Grain Sizes of Iridium Cups Used in NIT-1

Fueled Clad	Cup	Direction	Average Intercept (μm)	Grains per 640- μm Thickness
HF-438	Vent	Transverse	26.9	23.7
		Axial	32.5	19.7
	Shield	Transverse	38.7	16.5
		Axial	40.7	15.7
HF-450	Vent	Transverse	37.7	17.0
		Axial	37.2	17.2
	Shield	Transverse	34.1	18.8
		Axial	37.8	16.9

the release from the low-pressure LWRHU. Near the end of the exposure, there was a marked increase in the plutonium contents of the samples from the low-pressure experiment; no such trend occurred in the high-pressure experiment.

Of special interest is the observation that there was no sharp increase in the plutonium contents of the samples taken after the pressure was released from RHU-9-07. When heat sources containing larger amounts of $^{238}\text{PuO}_2$ were pressurized and subsequently depressurized, relatively large amounts of plutonium were released to the test chamber.² The fact that this did not occur in the current test of the 1-W LWRHU proves that the primary release mechanism is boiling of the water in the larger, hotter capsules, not the decompression of that water.

The as-recovered LWRHU is shown in Fig. 4. The surface showed minor salt deposits that were particularly evident at the ends of the Z-axis rods. There was no salt deposit around the joint of the closure, because the cap had been glued in place, as is that of a production LWRHU.

There were 154 μg of plutonium in the graphite, much more than the 10 μg of plutonium in the water immediately before the pressure was released, and nearly 5 times the amount found in the graphite from the low-pressure sample (RHU-10-15).

No salt was seen inside the capsule. There was no evidence of interaction between the fuel and the seawater (Fig. 5). The capsule was also unaffected by its immersion in the seawater (Fig. 6). A small amount of undercutting was present in the closure weld (Fig. 6b), but not enough to affect the capsule's integrity.

III. HELIUM RELEASE (D. Peterson)

The temperature dependencies of the helium fraction released as measured in runs 25, 29, 38, and 40 are plotted in Fig. 7. These runs involved relatively small $^{238}\text{PuO}_2$ samples (<1 g) exposed to a 227°C/min thermal ramp. Less than 10% of the total inventory was actually released during the ramps because of the fast pulse and the low upper temperature limit of only 1500°C. The line shown in the figure represents the helium release behavior predicted by applying a simple diffusion model. Parameters used in the model were obtained by averaging the results of more than 30 experiments. The agreement between the calculated and experimental data appears quite good considering the variations in fuel microstructure and the simplifications used in the model.

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TABLE IV. Results of AES Analysis of the Iridium Clads of NIT-1

Specimen	Location	Th ₆₅	C ₂₇₀	O ₅₁₀	S ₁₅₀	B or Cl ₁₈₀
		Ir ₂₂₉	Ir ₂₂₉	Ir ₂₂₉	Ir ₂₂₉	Ir ₂₂₉
HF-450	Int. edge	0.38	0.12	0.48	0.02	
	Center	0.57	0.22	0.60	0.02	
Vent cup - 1	Ext. edge	0.42	0.37	0.95	0.02	
	Average	0.46	0.24	0.68	0.02	
HF-450	Int. edge	0.40	0.71	1.12	nd	0.34
	Center	0.34	0.36	0.89	0.09	0.13
Vent cup - 2	Ext. edge	0.44	0.71	1.02	nd	0.21
	Average	0.39	0.59	1.01	0.03	0.23
HF-450	Int. edge	0.26	0.97	1.30	nd	
	Center	0.23	0.67	1.32	nd	
Shield cup - 1	Ext. edge	0.29	0.65	1.41	nd	
	Average	0.26	0.76	1.34	nd	
HF-450	Int. edge	0.23	1.16	0.96	0.11	0.08
	Center	0.32	0.99	1.26	0.09	nd
Shield cup - 2	Ext. edge	0.32	1.19	1.54	0.04	0.10
	Average	0.29	1.11	1.25	0.08	0.06
HF-438	Int. edge	0.30	0.55	0.78	nd	nd
	Center	0.35	0.72	1.06	nd	nd
Vent cup - 1	Ext edge	0.23	1.00	1.05	nd	nd
	Average	0.29	0.76	0.96	nd	nd
HF-438	Int. edge	0.34	0.76	0.91	nd	nd
	Center	0.25	0.75	0.91	nd	nd
Vent cup - 2	Ext. edge	0.21	0.79	0.88	nd	nd
	Average	0.27	0.77	0.90	nd	nd
HF-438	Int. edge	0.20	0.25	0.84	0.06	
	Center	0.35	0.27	0.67	nd	
Shield cup - 1	Ext. edge	0.35	0.66	0.99	0.13	
	Average	0.30	0.39	0.83	0.06	
HF-438	Int. edge	0.33	0.18	0.90	0.07	
	Center	0.24	0.28	0.66	nd	
Shield cup - 2	Ext. edge	0.35	0.38	0.98	0.06	
	Average	0.31	0.28	0.85	0.04	

TABLE V. Plutonium Contents in Weekly Seawater Samples (ng/l)

Sampling Date	Ambient-Pressure RHU-10-15	High-Pressure RHU-9-07	Sampling Date	Ambient-Pressure RHU-10-15	High-Pressure RHU-9-07
	Aquarium	HTF		Aquarium	HTF
11/06/81	RHUs placed in seawater		10/21	0.6	0.8
11/12	0.2	0.4	10/28	0.6	0.8
11/18	0.2	0.4	11/04	0.8	0.9
11/25	0.2	0.4	11/10	0.7	1.4
12/02	0.2	2.0	11/17	0.8	0.7
12/09	0.2	10.0	11/24	1.0	0.9
12/15	0.2	5.5	12/02	0.8	0.3
12/22	0.3	2.5	12/09	0.5	0.4
12/30	0.2	1.2	12/16	1.6	0.4
01/06/82	0.3	1.3	12/22	0.8	1.3
01/13	0.5	1.7	01/06/83	0.7	0.5
01/20	0.3	1.4	01/13	0.7	0.7
01/28	0.3	1.7	01/21	0.8	0.4
02/04	1.2	1.6	01/27	0.8	1.1
02/11	1.2	1.3	02/03	0.7	0.4
02/18	1.0	1.1	02/10	0.6	0.6
02/25	0.9	1.2	02/17	0.6	0.6
03/04	0.6	0.8	02/25	0.6	
03/11	0.8	1.1	03/03	0.7	0.5
03/18	0.8	1.2	03/11	0.5	0.6
03/25	0.7	1.6	03/18	0.6	0.4
04/01	0.9	0.9	03/31	0.5	0.4
04/08	0.9	0.9	04/08	0.5	0.3
04/15	1.4	0.9	04/14	0.5	1.2
04/22	1.2	0.9	04/21	0.5	1.0
04/29	1.1	1.0	04/28	0.5	1.0
05/06	1.1	1.0	05/05	0.6	0.8
05/13	1.0	1.0	05/16	0.6	1.3
05/20	1.1	1.2	05/20	0.7	0.6
05/27	1.1	0.4	05/26	0.7	0.6
06/03	0.9	0.9	06/02	0.8	0.5
06/10	0.9	0.9	06/09	0.8	0.7
06/17	0.5	0.4	06/16	0.9	0.4
06/24	0.4	0.4	06/23	1.0	0.4
07/01	0.4	0.4	07/01	1.1	0.4
07/08	0.7	0.4	07/07	1.3	0.3
07/15	0.9	0.4	07/13	2.0	0.4
07/22	0.8	0.4	07/21	2.0	0.3
07/29	0.7	0.4	08/05	2.5	0.4
08/05	0.7	0.3	08/08	RHU removed from aquarium	
08/12	0.6	0.9		2.3	0.9
08/19	0.6	1.0	08/11		0.3
08/26	0.5	0.4	08/18		0.5
09/02	0.5	0.9	08/26		1.0
09/09	0.6	1.0	08/31		
09/16	0.8	0.8		10:10 a.m.	HTF pressure relieved
09/23	0.9	0.8		11:40 a.m.	0.8
09/30	0.7	1.2		2:00 p.m.	0.6
10/07	0.8	0.8	09/02		0.4
10/15	0.7	1.1	09/07		RHU removed from HTF

(a)

(b)

(c)

(d)

Fig. 1. The fueled clads in NIT-1 were deformed but unbroken after the impact ($\sim 1.5X$): (a) HF-450 impact face; (b) HF-450 profile; (c) HF-438 impact face; (d) HF-438 profile.

(a)

(b)

(c)

(d)

Fig. 2. Typical iridium microstructures of the iridium cladding of NIT-1 (100X): (a) HF-450 vent cup; (b) HF-450 shield cup; (c) HF-438 vent cup; (d) HF-438 shield cup.

Fig. 3. Weld microstructure of fueled clad HF-438 at the location of the fuel fragment push-through (cross section not normal to the weld bead) (50X).

Fig. 4. RHU-9-07 was unaffected by seawater immersion (2X).

(a)

(b)

Fig. 5. The fuel in RHU-9-07 was unaffected by the high-pressure immersion: (a) the microstructure was uniform (10X); (b) there were no deposits in the pores (250X).

Fig. 6. The RHU-9-07 Pt-30Rh capsule was unaffected by high-pressure seawater immersion (10X).

Fig. 7. Temperature dependence of helium fraction released from plutonia exposed to 227°C/min ramp in runs 25 (x), 29 (○), 38 (Δ), and 40 (●). Solid curve represents results calculated from simple diffusion model.

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