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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 (GPHS)

A. Safety-Verification Impact Tests (D. Pavone and C. Frantz)

1. SVT-1. The first fueled impact test in the Safety-Verification test program was completed. A full GPHS module was subjected to the aging, reentry, and impact environments characteristic of a minimum-gamma return from an unsuccessful launch attempt. Both prime fueled clads survived the test, but the two secondary fueled clads failed.

The components of the GPHS module used for the SVT-1 impact are listed in Table I. All of the graphite components were made to specification except the aeroshell, which was machined to simulate the degree of ablation that would occur during a minimum-gamma reentry. The prime fueled clads met all requirements of the specification, but the secondary clads contained ultrasonically detected defects that prevented them from being classified as prime.

The impact assemblies were aged simultaneously in a graphite furnace fixture for 200 h at an iridium clad temperature of 1287°C in a vacuum of $<1 \times 10^{-6}$ torr.

Following aging, the impact assemblies were subjected to a simulated reentry temperature pulse to 1580°C . The impact test was performed in a "face-on" orientation ($\alpha = 0$) at a clad temperature of 919°C and a velocity of 54.4 m/s.

The recovered impact assembly is shown in Fig. 1. The module stayed together in spite of the nearly full-length longitudinal cracks on the impact face (Fig. 1a) and on the side faces and the failed area in the center of the back face (Fig. 1b).

When the impact assemblies were removed from the aeroshell, they had the typical bowed profile that results from their being supported at the ends and being free to move into and crush the weak insulation (Fig. 2). In both cases, the impacted sides were cracked longitudinally, but the caps were still retained.

No failures occurred in either of the prime fueled clads, HF-232 and HF-238, and only modest, localized deformation occurred in the iridium cladding (Fig. 3). However, both of the secondary fueled clads failed. There was a longitudinal crack on the impact face of the shield cup of HF-261 as well as a longitudinal crack in both cups on the back face (Fig. 4). The cracks appear to have resulted from movement of a major segment of the fuel

TABLE I. Components of the SVT-1 Module

Component	Serial No.
Aeroshell	PAL-012
Prime assembly	
Fueled clads	HF-232, HF-238
Impact shell	PGL-013
Insulation sleeve	C-40-4
Insulation disks	D-8-2-24, P-8-2-15
Secondary assembly	
Fueled clads	HF-261, HF-410
Impact shell	PGL-014
Insulation sleeve	C-31-3
Insulation disks	P-8-2-2, P-12-1-15

pellet (Fig. 4c) during the impact. That such a shift occurred is shown in Fig. 5. A major diametral fracture plane with ends terminating at the opposed cracks in the iridium clad is easily visible.

The other secondary fueled clad, HF-410, contained a single crack (Fig. 6). The crack appeared to have originated at the shield cup's radius. One end propagated axially through the weld and into the vented cup; the other end extended across the end of the shield cup. The crack appears to be associated with a fuel shift (Fig. 6c), but the shift is not as obvious as the one in HF-261 (Fig. 4c).

The overall strains sustained by the SVT-1 fueled clads are listed in Table II. Of particular interest is that HF-232 and HF-261 had similar strains, but only HF-261 failed, indicating the importance of the mode of fuel failure on the impact response.

Samples were removed from the failed fueled clads for metallographic examination and for Auger electron spectroscopy (AES) analysis. A typical section and a section through the impact-face crack in HF-261 are shown in Fig. 7. The fracture occurred in the reverse-bend region near a rather sharp, ductile bend in the capsule wall (Fig. 7a). The etched microstructure (Fig. 7b) indicates that the fracture occurred in a layer of relatively large, columnar grains on the interior surface of the clad. This columnar layer may not have been the direct cause of the failure, but as the crack below the failure indicates, it was particularly susceptible to cracking when subjected to tensile bending stresses. Elsewhere, the microstructure consisted of relatively fine, elongated grains (Fig. 7c).

Sections through the larger crack on the back of HF-261 and through a typical region in the vented cup are shown in Fig. 8. The relative radial displacement of the two sides of the failure (Fig. 8a) demonstrates the effects of the fuel fragment movement. The two sides of the crack do not match, indicating that the failure consisted of two or more closely spaced cracks. The failure is typically brittle, although the local thickness reduction was 4%. The microstructure in the region of the failure and away from it (Fig. 8b) consisted of fine, somewhat elongated grains.

Sections through the crack and through a typical region in the shield cup of HF-410 are shown in Fig. 9. The radial displacement of the sides of the failure demonstrates the effects of the fuel fragment push-through. The fracture mode was brittle intragranular, even though there was a 9% thickness reduction in the failure region. The microstructure was composed of fine equiaxed grains.

The results of AES analysis of the grain-boundary chemistry of the iridium samples from both failed fueled clads are tabulated in Table III. The samples were selected from areas of the capsule adjacent to the fractures. The results showed high concentrations of sulfur contaminant in one sample of the shield cup of HF-261 and in one sample of the vent cup of HF-410. Smaller amounts were observed in the second sample of HF-410. Because of the size of the sulfur peak and its proximity to an iridium peak, a sulfur/iridium intensity ratio could not be estimated for two of the samples. The thorium/iridium intensity ratios are generally lower than normal except for one sample of HF-261. Carbon and oxygen were detected in nearly all locations in variable quantities. It is unlikely that sulfur impurities caused the failures because both cups of HF-261 failed but only one sample of the

TABLE II. Overall Impact Deformation of the Fueled Clads in SVT-1

Fueled Clad	Strain, %	
	Diametral	Axial
HF-232	13.5, -11.1	5.8
HF-238	9.6, - 7.3	3.8
HF-261	11.7, -11.1	5.6
HF-410	9.5, - 9.6	1.4

TABLE III. Results of AES Analysis of the Iridium Clads of HF-261 and HF-410

Specimen	Location	TH_{65}	S_{150}	C_{270}	O_{510}
		Ir_{229}	Ir_{229}	Ir_{229}	Ir_{229}
HF-261 Vent cup-1	Int. edge	0.41	nd ^a	0.68	0.93
	Center	0.35	nd	0.57	1.04
	Ext. edge	0.44	nd	0.53	1.07
	Average	0.42	nd	0.59	1.01
HF-261 Vent cup-2	Int. edge	0.64	nd	0.20	0.28
	Center	0.78	nd	nd	0.39
	Ext. edge	0.56	nd	0.28	0.39
	Average	0.66	nd	0.16	0.22
HF-261 Shield cup-1	Int. edge	0.63	nd	nd	0.55
	Center	0.56	nd	nd	0.38
	Ext. edge	0.65	nd	nd	0.34
	Average	0.61	nd	nd	0.42
HF-261 Shield cup-2	Int. edge	0.27	v.s. ^b	1.16	1.24
	Center	0.24	v.s.	0.95	1.12
	Ext. edge	0.32	v.s.	0.92	0.85
	Average	0.28	v.s.	1.01	1.07
HF-410 Vent cup-1	Int. edge	0.35	0.23	0.60	1.03
	Center	0.31	0.20	0.52	1.00
	Ext. edge	0.29	0.21	0.46	1.02
	Average	0.32	0.21	0.53	1.02
HF-410 Vent cup-2	Int. edge	0.25	v.s.	0.85	1.32
	Center	0.34	v.s.	1.02	1.13
	Ext. edge	0.45	v.s.	1.00	1.23
	Average	0.35	v.s.	0.96	1.23
HF-410 Shield cup-1	Int. edge	0.37	nd	0.34	0.79
	Center	0.50	nd	0.20	0.79
	Ext. edge	0.42	nd	0.37	0.78
	Average	0.43	nd	0.30	0.79
HF-410 Shield cup-2	Int. edge	0.40	nd	0.24	0.64
	Center	0.74	nd	nd	0.36
	Ext. edge	0.50	nd	0.18	0.51
	Average	0.55	nd	0.14	0.50

^and not detected.

^bv.s. very strong.

shield cup contained grain-boundary sulfur, and the shield cup of HF-410 failed but only the vent cup contained sulfur.

2. SVT-2. We completed the aging of this test assembly, which will be used for an impact test in the minimum-gamma mode. It is being stored in a helium atmosphere until it can be reentered and impacted.

3. SVT-3. The scheduled 90-day aging exposure was completed on October 21, 1983. No evidence of vent plugging was observed beyond a short, 0.7-h period at 689 h of exposure. The impact assemblies were reentered (orbital decay), and the module was assembled and impacted at 975°C and 54.43 ± 0.22 m/s.

4. SVT-4. By November 1, 1983, the accumulated exposure time for this test assembly was 1615 hours. Continuous monitoring of the helium released from the assembly has not indicated any evidence of vent plugging.

5. SVT-5. As of November 1, 1983, the accumulated exposure time for this assembly was 1116.5 h. Helium release is not being monitored during the aging exposure.

B. Safety-Verification Launch-Abort Tests (R. Zoehrer)

Los Alamos supported the General Electric Company's experiment in the Defense Nuclear Agency's DIRECT COURSE event. An electrically heated GPHS radioisotope thermo-electric generator (containing one UO_2 -loaded GPHS module) was mounted 11.9 m from the surface of a 554-Mg (609-ton) sphere of ammonium nitrate/fuel oil (ANFO). Our role was to aid in the recovery of the test object after the ANFO exploded.

Following the explosion, the Los Alamos search team staked out the area and began the search. Thirty-five fragments of the generator housing and a single fragment of the aeroshell from the GPHS module were found by visual searching. No iridium or UO_2 was found this way.

The iridium cladding around the UO_2 had been activated in Los Alamos' Omega-West reactor so that it could be detected by personnel from our Advanced Nuclear Technology Group (Q-2), who swept the area with sensitive gamma-radiation detectors. They found 70% of the iridium that had been in the test. The pieces ranged from the size of a pinhead to one piece 20×30

mm. The degree of scatter of the iridium fragments suggested that the generator had disintegrated in mid air.

II. SAFETY TECHNOLOGY

A. Biaxial Testing of Iridium (T. George)

The first two iridium disks were prepared for testing. Each was electroplated with copper onto which was photoetched a grid of circles. The etched surfaces were grit-blasted. The grit blasting formed circles on the iridium where the copper had been etched away, but the regions where this copper remained were protected. After grit-blasting, the remaining copper was removed. The disks were cleaned, and a circumferential hold-down groove was formed. The first two disks were heat-treated to a grain size of 25 μm .

B. Fuel Transport (D. Peterson)

Plutonium and impurities in the PuO_2 fuel appear to be transported by the movement of species other than the simple oxides. We are attempting to make PuPO_4 so we can measure its thermodynamic properties and determine its stability in the fuel and in the presence of iridium.

C. Iridium Grain Growth (D. Peterson)

We consistently observe that the grain growth of the DOP-26 iridium alloy (and its DOP-4 predecessor) exposed to PuO_2 is faster than in the same alloy not exposed to PuO_2 . Thorium is added to iridium to slow the rate of grain growth; it is present initially as ThIr_5 . On exposure to the fuel, the thorium in the iridium becomes depleted. We also observe carbon on the grain boundaries of the iridium alloy. Iridium carbides are not stable (there is no iridium/carbon interaction even after years at high temperature), so the carbon must be present in another form. We postulate that the phase ThIr_3C could form (the carbon coming from the graphite components of the heat source) and then react with the fuel to form ThO_2 and CO, the former dissolving in the fuel and the latter passing off. We are attempting to prepare the ternary compound by arc melting. If we are successful, we will measure its thermodynamic properties.

(a)

(b)

Fig. 1. SVT-1 was cracked by the impact, but it stayed together: (a) impact face; (b) back face (0.6X).

(a)

(b)

(c)

(d)

Fig. 2. The impact assemblies in SVT-1 were cracked and bowed after the impact: (a) PGL-013 impact face; (b) PGL-013 side; (c) PGL-014 impact face; (d) PGL-014 side (0.6K).

HF 232

(a)

HF 232

(b)

HF 238

(c)

HF 238

(d)

Fig. 3. The prime fueled clads in the SVT-1 module were deformed but intact after the impact: (a) HF-232 impact face; (b) HF-232 side; (c) HF-238 impact face; (d) HF-238 side (1.5X).

HF 261

(a)

HF 261

(b)

HF 261

(c)

Fig. 4. HF-261, a secondary fueled clad in the SVT-1 impact, failed: (a) impact face with crack in the shield cup; (b) back face with crack extending into both cups; (c) vent end showing fuel movement associated with the failures (1.5X).

Fig. 5. The fuel pellet in HF-261 had a major diametral fracture plane whose termini were at the opposed cracks in the iridium clad ($\sim 1.5X$).

HF410

(a)

HF410

(b)

HF410

(c)

Fig. 6. HF-410, a secondary fueled clad in SVT-1, failed on impact: (a) impact face; (b) back side; (c) shield cup bottom (1.5X).

Fig. 7. The impact-face crack in the shield cup of HF-261 was a brittle, intragranular fracture: (a) a section shows the failure and a nearby ductile bend (7X); (b) etching revealed an internal layer of columnar grains (50X); (c) the typical microstructure consisted of elongated, relatively fine grains (100X).

(a)

(b)

Fig. 8. The microstructure of the HF-261 vent cup was normal: (a) impact-face failure (50X); (b) typical microstructure (100X).

(a)

(b)

Fig. 9. The shield cup of HF-410 shows brittle failure as a result of the impact (a, 50X), but its microstructure was fine grained and equiaxed (b, 100X).

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