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Progress Report

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Space Nuclear-Safety Program

November 1982

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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. DIT-4 Impact (F. W. Schonfeld)

The fourth design iteration test, DIT 4, of the General-Purpose Heat Source (GPHS) was intended to provide information about the possible effects on impact behavior of using motor-driven abrasive cleaning instead of manual abrasive cleaning. The test details and initial results were reported in the September report. The post-mortem examination of the iridium capsules was completed this month. A previously undetected failure was discovered.

1. Weld Microstructures. The internal surface of the overlap portion of the closure weld of capsule SRP-394 is shown in Fig. 1. A small centerline crack can be seen just beyond the submergence of the weld. A section through this region is shown in Fig. 2. The grain structure of the weld (Fig. 2b) is not unusual, but the crack and porosity (Fig. 2a) are certainly undesirable. The single-pass weld region of SRP-394 was uncracked and essentially free of porosity. Etching revealed, as usual, a much better microstructure than in the overlap region (Fig. 3).

The weld overlap region of the interior of capsule SRP-398 is shown in Fig. 4. The typical longitudinal

cracks formed in the reverse-bend region can be seen in the right of the photograph. A section through the overlap region (Fig. 5) revealed more porosity than usual. The pores are concentrated away from the surface and are particularly prevalent at the overlap interface. Etching revealed a generally satisfactory microstructure in this region (Fig. 5b). The microstructure of the single-pass weld region (Fig. 6) also contained more porosity than is usually seen in Savannah River welds, but the microstructure near the centerline was quite good. A single large pore whose revealed diameter was 45% of the capsule's wall thickness was found near the edge of the single-pass region (Fig. 7).

2. Cup Microstructures. The center sections cut from the two capsules are shown in Fig. 8. Both sections display the general pattern of deformation typical of the GPHS module impacted in the face-on orientation. SRP-398 was more seriously deformed than SRP-394; it was in the closure end of the impact shell. We have observed this location-dependent deformation in previous tests and believe that it occurs because the impact shell's cap pops off at an early stage of the impact and without the cap the closure end of the impact shell does not provide as much protection.

The inside surface of the area of SRP-394 that contained the majority of the cracks is shown in Fig. 9.

This piece was cut from the reverse-bend region. A section through it (Fig. 10) shows that one of the cracks penetrated 80% of the wall. The section also shows a region (upper left) where the capsule wall was deformed between the target and a piece of fuel, demonstrating again the importance of stress state to the deformation of iridium.

The principal cracks visible in Fig. 10 are shown at higher magnification in Fig. 11. The crack morphology has the expected brittle intergranular mode. What is unexpected is the range of grain sizes seen in these etched sections. The large grains near both the inner and outer surfaces should not be present after the relatively mild 30-day, 1310°C aging treatment. The 930°C impact temperature was well below the nominal recrystallization temperature of the DOP-26 iridium alloy, so the grain structures were present before the impact and represent the response of the cup to the aging treatment.

During examination of the pieces of capsule SRP-394, a small crack was found in the radius of the nonvented iridium cup (Fig. 12). The crack goes through the wall and is, technically, a failure. However, it was only 2 μ m wide and could have released only a minimal amount of fuel. The grain structure in this region (Fig. 12b) is significantly different from those in the reverse-bend region.

The cracks in the reverse-bend region of capsule SRP-398 had the usual brittle, intergranular morphology (Fig. 13), and the deepest crack penetrated 65% of the wall thickness. The grain structure is normal for the heat treatment.

B. DIT-5 Impact (R. W. Zocher, F. W. Schonfeld)

A fifth design iteration test, DIT-5, was completed. The purpose of the test was to compare the impact response of a GPHS fueled clad that had been welded with a new four-pole oscillator to the impact response of a fueled clad welded with the previously used two-pole oscillator. The two fueled clads, supplied by Savannah River and designated SRP-423 (two-pole) and SRP-444 (four-pole), were aged for 30 days at 1310°C in an impact shell, radiographed, placed into a partial aeroshell that had been machined 1.27 mm on all faces, and impacted in the face-on orientation at 930°C and 60.5 m/s (slightly above the normal range of 58 ± 2 m/s). The tests were identical in all aspects to DIT-4 discussed above except for the source of the fueled clads and the velocity of the impact.

Both clads in DIT-5 were cracked by the impact. SRP-444 failed extensively. None of the failures was related to the welding technique.

The post-aging radiographs of the DIT-5 impact assembly showed that both fuel pellets were cracked (Fig. 14). The aging treatment did not change the degree of cracking appreciably. The recovered test object is shown in Fig. 15. A corner of the aeroshell was broken off, the aeroshell cap came out, and the impact shell was cracked, but the fueled clads remained in the graphite.

The impact assembly is shown in Fig. 16. The degree of cracking on the impact face (Fig. 16a) and the typical bowed conformation (Fig. 16b) are easily visible as are the thermocouple holes in the side of the shell. The cap was partially released by the impact, which also caused the circumferential crack visible on the back of the shell (Fig. 16c).

The recovered fueled clads used in DIT-5 are shown in Fig. 17. The large failure in SRP-444 is easily visible, but the smaller crack in SRP-423 is barely visible. The impact face and the two ends of capsule SRP-423 are shown in Fig. 18. The longitudinal crack can be seen on the impact face (Fig. 18a).

Capsule SRP-444 is shown in Fig. 19. The major crack and a parallel smaller crack are readily apparent on the impact face (Fig. 19a); the extent of the major crack can be seen in the end views. As in SRP-423, a deposit around the vent can be seen on SRP-444 (Fig. 19b). It is evident from Fig. 19c that the degree of failure in SRP-444 is directly linked to the movement of relatively large blocks of fuel.

The gross strains of the impacted DIT-5 fueled clads listed in Table I suggest that SRP-444 was deformed somewhat more than SRP-423; however, if the crack width were taken into account, there probably would be little difference in the strains.

Table I. DIT-5 Impact Strains

	Capsule 423	Capsule 444
Diametral		
Vent cup	0.085-0.082	0.106-0.109
Weld	0.103-0.085	0.123-0.099
Shield cup	0.085-0.094	0.096-0.095
Length		
Minimum	0.010	0.009
Maximum	0.063	0.056

C. Early Compatibility Test (D. Pavone)

The transverse metallographic cross section of capsule HF-2 was submitted for ion microprobe analysis to attempt to compare the grain boundary chemistry of the large-grain and small-grain regions, particularly as regards the thorium segregation. No correlation between thorium concentration and grain size was found. Other elements detected were hydrogen, sodium, aluminum, potassium, calcium, chromium, manganese, iron, copper, and iridium. The alkali and alkaline earth elements in this list appeared to be concentrated in pores and are believed to be surface contaminants. No correlation was observed between the chromium, manganese, iron, and copper concentrations and grain size.

D. Phosphorous Effects Experiment (D. Pavone)

On December 1, 1982, the accumulated exposure times were 3985 h for the fueled assemblies and 2969 h for the control samples.

E. Explosion Tests (C. M. Seabourn)

Work by Sandia National Laboratories is continuing in preparation for the shock-tube testing of GPHS modules. The shock tube muzzle adaptor and the mechanism to hold the furnace that will be used to preheat the module stack are being fabricated. Delivery is expected by mid December. Calibration explosions have been made to establish explosive weight requirements for the different pressure levels that may be required. As planned, the first test will be at 1.38 MPa (200 psi) (static).

F. Fragment Impact Tests (C. M. Seabourn)

The post-mortem examinations of the three FWPF* graphite plates that had been impacted with 50-cal aluminum alloy bullets¹ were completed. They showed that the collateral damage caused by the penetrating bullets was modest.

*FWPF is Fineweave-Pierced Fabric 3-D carbon/carbon composite, a product of AVCO Systems Division, 201 Lowell St., Wilmington, MA 01887.

Three FWPF plates equivalent in thickness (4.86 mm) and orientation to the side wall of the GPHS module were impacted with 18-g, 50-cal aluminum alloy bullets at initial velocities of 900, 633, and 320 m/s at room temperature. The velocities of the emerging bullets were 833, 580, and 289 m/s, respectively.

The plates were vacuum impregnated with catalyzed epoxy resin and sectioned on hole diameters defined by reference to the z-axis of the FWPF composite; one cut was parallel to the z-axis and the other was perpendicular to it. The sections were mounted, polished, and examined microscopically.

The results are shown for the three tests in Figs. 20, 21, and 22. The areas designated "A" indicate pre-existing imperfections, and the numbered areas indicate cracks that are filled with epoxy resin and believed to have formed as a result of the impact. The distances of the impact cracks from the edges of the respective holes are given in Table II.

Examination of the table and the photomicrographs indicates that there is little difference between the 320- and 632-m/s tests, but that there is significantly less collateral damage in the 900-m/s test. When the actual hole sizes are taken into consideration, the damage radii (defined as the average distance from the center of the hole to the damage shown in Figs. 20, 21, and 22) increase with the bullet velocity from 7.9 to 10.0 to 10.4 mm. The increase is not simply related to the velocity or energy because the bullet is not rigid and it deforms more with increasing velocity.

TABLE II. Distances from Hole Edges to Impact-Formed Defects

Velocity (m/s)	Distance (mm)	
	Parallel z-axis	Perpendicular z-axis
320	0.6	1.3
	4.4	1.0
	0.9	1.5
		1.4
633	1.6	1.8
		1.3
		1.7
900	0.4	0.5

II. HELIUM RELEASE FROM PLUTONIA

(D. E. Peterson)

Results of all previous measurements of helium release from plutonium dioxide were correlated this month. A simple diffusion model was used to analyze the gas release rates. Examination of the temperature dependence of the diffusion coefficient revealed several breaks in the curve that may be interpreted as changes in the release mechanism. Both the thermal pulse and fuel microstructure appear to influence the release behavior over selected temperature regimes. In the case of a relatively fast thermal pulse of $227^{\circ}/\text{min}$, the release behavior shows four distinct regions. Below 885°C , the release rate is relatively slow with an activation energy of 24.4 ± 1.6 kcal. A definite mass dependence of the absolute value of the diffusion coefficient is evident over this region with lower values associated with higher masses. This behavior is interpreted as being due to bulk diffusion with trapping along the grain boundaries. (The diffusion rate through each grain is higher than that along the boundary.) Over the region 885 - 1170°C , the release rate increases with an associated E_a value of 91.3 ± 3.4 kcal. In this region the helium pressure in the grain boundaries is thought to become so great that bubbles have been initiated, and diffusion along the grain

boundaries is predominating the release behavior. In the range 1170 - 1400°C , the release of helium has an E_a value of 52.9 ± 2.2 kcal. In this range we believe that the rate of release of helium from the grain boundaries is in equilibrium with the rate of helium arriving from the grains and the E_a value represents the energy for purely bulk diffusion. A significant observation is that in the region above 1400°C , the bubbles in the grains and along the grain boundaries appear to become mobile, resulting in a marked increase in the release rate and an E_a value to 142 ± 11 kcal. This mechanism then predominates up to the point of exhaustion of the helium inventory.

Slower thermal ramps, below $120^{\circ}/\text{min}$, show that bulk diffusion predominates even over the low-temperature regions. When the temperature is above 1400°C , helium bubbles within the grains become mobile and diffuse to and along the grain boundaries resulting in an increased E_a value approaching 140 kcal.

REFERENCE

1. S. E. Bronisz (Comp.), "Flight Systems Safety Program, September 1982," Los Alamos National Laboratory report LA-9673-PR (February 1983).

Fig. 1. A small crack was present in the overlap region of the SRP-394 closure weld, 12X.

(a)
As-polished, 50X.

(b)
Etched, 50X.

Fig. 2. A section through the weld overlap region of capsule SRP-394 revealed the presence of porosity in addition to the weld crack.

Fig. 3. The microstructure of the single-pass region of the SRP-394 closure weld was quite good, 50X.

Fig. 4. Several reverse-bend cracks can be seen near the weld submergence point in the interior of capsule SRP-398.

(a)
As-polished, 50X.

(b)
Etched, 50X.

Fig. 5. There was considerable porosity in the overlap region of the closure weld of capsule SRP-398.

(a)
As-polished, 50X.

(b)
Etched, 50X.

Fig. 6. The single-pass region of the SRP-398 closure weld was porous but had a good microstructure.

Fig. 7. A large gas bubble was found near the edge of the single-pass region of the SRP-398 closure weld, 50X.

(a)
SRP-394, 1.5X.

(b)
SRP-398, 1.5X.

Fig. 8. The slices cut from the center sections of the capsules used in the DIT-4 impact show the typical deformation pattern for the face-on impact orientation.

Fig. 9. Capsule SRP-394 was cracked in several places on the interior in the reverse-bend region of the impact face, $\sim 3.8X$.

Fig. 10. A cross section through the reverse-bend region of SRP-394 showed that the cracks penetrated as much as 80% of the wall thickness, $\sim 7X$.

(a)

(c)

(b)

Fig. 11. The cracks in SRP-394 were intergranular and the grain structure was unusual, 50X.

(a)
As-polished, 10X.

(b)
Etched, 50X.

Fig. 12. A small crack penetrated the nonvented capsule of SRP-394.

Fig. 13. The deepest crack on the impact face of capsule SRP-398 penetrated 65% of the wall, 50X.

Fig. 14. Both fuel pellets used in the DIT-5 experiment were cracked before the impact.

Fig. 15. The DIT-5 aeroshell and impact shell were damaged by the impact, but the fueled clads were not released, ~IX.

(a)
Impact surface, 1X.

(c)
Back surface, 1X.

(b)
Side view, 1X.

Fig. 16. The DIT-5 impact shell was cracked and bent by the impact.

Fig. 17. The fueled clads impacted in DIT-5 showed the deformation typical of the face-on impact, ~1X.

(a)
The crack is at the left of the impact face.

(c)
Nonvented end.

(b)
Some effusate is visible around the vent.

Fig. 18. DIT-5 capsule SRP-423 after impact.
All views 1.5X.

(a)
The major failure occurred on the impact face.

(c)
The deformation caused by fuel movement is visible from the nonvented end.

(b)
Fuel effusate was present around the vent.

Fig. 19. Capsule SRP-444 failed in the DIT-5 impact. All views 1.5X.

(a) Parallel to z-axis.

(b) Perpendicular to z-axis.

Fig. 20. The damage caused by the 320-m/s bullet. Both views 20X.

(a) Parallel to z-axis.

(b) Perpendicular to z-axis.

Fig. 21. The damage caused by the 633-m/s bullet. Both views 20X.

(a) Parallel to z-axis.

(b) Perpendicular to z-axis.

Fig. 22. The damage caused by the 900-m/s bullet. Both views 20X.

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