LA-9839-PR
Progress Report

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Space Nuclear Safety Program

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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Compiled by
S. E. Bronisz

Los Alamos Los Alamos National Laboratory
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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 may change as the work continues.

I. GENERAL-PURPOSE HEAT SOURCE

A. Early Compatibility Test (D. Pavone)

Four additional samples from the vent cups of HF-2 and HF-5, the fueled clads used in the first early compatibility test (ECT-1), were examined by Auger electron spectroscopy (AES). The results are listed in Table I and adsorbed-current images of the surfaces analyzed are shown in Fig. 1. The thorium/iridium (Th/Ir) intensity ratios varied considerably, as was the case in prior samples from these clads.* One sample contained sulfur; another, boron or chlorine.

The grain sizes (Fig. 1) are mixed, but moderately large. There appears to be no correlation of thorium concentration with grain size nor of the presence of sulfur with grain size. At this stage of the post-mortem examination of the ECT-1 fueled clads, there is no unequivocal explanation of the variable microstructures seen in the two cups made from iridium sheet LR 234.

*S. E. Bronisz, Compiler, "Space Nuclear Safety Program, January, 1983," Los Alamos National Laboratory report LA-9813-PR (June 1983).

B. Phosphorus Effects Experiment (D. Pavone)

Metallographic examinations of the aged archive rings, the empty control capsule, and the fueled clads used in this experiment were completed. In each case where fuel was present, the grain size was larger than either the aged archive sample or the control cups (Table II). Large grains were observed near the interior surfaces of every fueled clad (Figs. 2-10). The microstructures of two cups, P701-4 (Fig. 7) and PR715-2 (Fig. 9), were extremely variable and reminiscent of the microstructures of the vent cups on ECT-1.

C. Overpressure Tests (C. Seabourn, R. Tate, and R. Zocher).

A second overpressure test was completed at 5.1 MPa (735 psi) in the Sandia National Laboratories' 0.57-m shock tube. The aeroshell was broken, but the impact assemblies remained within it. Post-mortem examination revealed that none of the iridium clads had been breached. No exterior cracks were observed on any of the

clads. Based on the absence of interior cracks on the sectioned clad (IRG-117) it is presumed no interior clad cracks were produced in any of the tested capsules.

As the initial test in the General-Purpose Heat Source (GPHS) explosion test series, module PAL-002 was exposed to a 1.4 ± 0.07 MPa (200 ± 10 psi), 8.4 ± 1.4 kPa-s (1.22 ± 0.20 psi-s) blast wave at $1091 \pm 5^\circ\text{C}$. The module penetrated 0.8 m into the vermiculite-filled catch box. The only external damage consisted of pitting on the surface facing the blast wave and a small chip off one corner. The latter damage probably occurred when the module hit one of the dummy modules with which it was surrounded. A radiographic examination showed that neither the capsules nor the impact shells had been damaged or displaced.

Module PAL-002 was then exposed to a 5.1 ± 0.21 MPa (735 ± 30 psi), 22.1 ± 2.8 kPa-s (3.2 ± 0.4 psi-s) blast, while at $1093 \pm 5^\circ\text{C}$. The module was driven 1 m into the vermiculite. The recovered module is shown in Fig. 11. The aeroshell was broken on a broad face that had been oriented parallel to the blast direction. The lower impact shell was broken (Fig. 12) and fueled clad IRG-118 was visible. The upper impact shell appeared undamaged (Fig. 13).

The four fueled clads removed from the impact shells are shown in Fig. 14. After the two tests, the capsules exhibited light overall deformations. Capsule IRG-115 was determined to be the clad with the most (-2.5%) general deformation. It had been in the undamaged impact shell. Capsule IRG-118, which had been in the broken impact shell with capsule IRG-117, had the greatest local deformation. IRG-118 was opened. The inner clad surface was crack-free. The condition of the UO_2 fuel simulant is shown in Fig. 15. It was broken into relatively large pieces by the sequenced explosion tests.

II. HELIUM RELEASE (D. Peterson)

All of our $^{238}\text{PuO}_2$ helium release experimental results were analyzed by means of the simple diffusion model this month. The heating rates had ranged from 12 to $454^\circ\text{C}/\text{min}$ and the maximum temperature of the heating ramps had been 1600°C . When the calculated effective diffusion constants, D' , were plotted against the reciprocal of the absolute temperature, they tended to fall into four distinct linear regions. An illustration of this is shown in Fig. 16, where the $227^\circ\text{C}/\text{min}$ heating rate results are plotted.

The four regions can be related to the microstructure of the PuO_2 . At the lowest temperatures (Region 1 in Fig. 16), the release of helium occurs by bulk diffusion modified by grain-boundary trapping. At higher temperatures (Region 2), diffusion along grain boundaries is the rate-controlling mechanism. At the higher temperatures in Region 2, the helium arrival rate is so great that grain-boundary bubbles become interconnected, so that in Region 3 the release is controlled by bulk diffusion to the now open grain boundaries. At the highest temperatures (Region 4), the intragrain bubbles also become mobile.

The results from this experiment and from the other experiments cited show that the effectiveness of grain-boundary trapping is reduced by reductions in the heating rate and by decreasing the PuO_2 grain size. The high-temperature bubble mobility was observed in nearly every experiment, indicating that the release of the helium from $^{238}\text{PuO}_2$ fuel during a reentry event is more complex than previously postulated.

TABLE I. Supplemental AES Results for ECT-1 Iridium Capsules

Location	Th ₄₉ /Ir ₂₂₉	P ₁₂₀ /Ir ₂₂₉	C ₂₇₆ /Ir ₂₂₉	O ₅₁₀ /Ir ₂₂₉	Si ₉₂ /Ir ₂₂₉	S ₁₃₀ /Ir ₂₂₉	B or Cl ₁₈₀ /Ir ₂₂₉	Th65/Ir ₅₄
ECT-1/HF5								
VFF-1 ^a								
Interior edge	0.10	---	1.08	0.98	---	0.22	---	0.02
Center	0.15	---	0.91	1.25	---	---	---	0.03
Exterior edge	0.11	---	---	1.54	---	0.07	---	0.02
Average	0.12	---	0.66	1.26	---	0.10	---	0.02
ECT-1/HF2								
Wall								
Interior edge	0.25	---	---	0.47	---	---	---	0.05
Center	0.53	---	0.22	0.69	---	---	---	0.09
Exterior edge	0.48	---	0.30	0.72	---	---	---	0.08
Average	0.42	---	0.17	0.63	---	---	---	0.07
ECT-1/HF2								
VF-2 ^b								
Interior edge	0.08	---	0.37	0.45	---	---	0.10	0.01
Center	0.57	---	0.27	0.68	---	---	0.07	0.09
Exterior edge	0.56	---	0.82	1.15	---	Trace	1.52	0.09
Average	0.40	---	0.49	0.76	---	Trace	0.56	0.06
ECT-1/HF2								
IFF-2 ^c								
Interior edge	0.64	---	---	0.62	---	---	---	---
Center	0.72	---	---	0.65	---	---	---	---
Exterior edge	0.45	---	---	0.56	---	---	---	---
Average	0.60	---	---	0.61	---	---	---	---

^aVFF = Vent face fracture

^bVF = Vent face

^cFF = Impact face fracture

TABLE II. Grain Size of Iridium Cups in SRL Fuel Impurity Experiment

	Longitudinal									
	Transverse		Interior		Center		Exterior		Mean	
	l'ept μm	grains 640 μm	l'ept μm	grains 640 μm	l'ept μm	grains 640 μm	l'ept μm	grains 640 μm	l'ept μm	grains 640 μm
PEF-44										
L254-7										
Archive	21.7	29.5	47.1	13.6	50.6	12.6	46.4	13.8	48.0	13.3
Archive aged	40.8	15.7	57.2	11.1	64.6	9.9	65.5	9.8	62.4	10.3
Cup	66.1	9.7	196.5	3.3	103.3	6.2	100.4	6.4	133.4	4.8
L256-8										
Archive	24.5	26.2	42.9	14.9	38.4	16.7	46.4	13.8	42.6	15.0
Archive aged	42.3	15.1	55.8	11.5	67.5	9.5	71.7	8.7	65.0	9.8
Cup	70.1	9.1	167.4	3.8	98.3	6.5	110.2	5.8	125.3	5.1
PEF-45										
L255-6										
Archive	25.0	25.3	58.4	10.9	43.5	14.7	54.7	11.7	52.2	12.3
Archive aged	44.1	14.5	88.6	7.2	65.5	9.8	70.6	9.1	74.9	8.5
Cup	64.8	9.9	137.8	4.6	100.9	6.3	110.8	5.8	116.5	5.5
L247-R2										
Archive	21.1	30.3	49.9	12.8	36.5	17.6	41.9	15.3	42.8	15.0
Archive aged	37.1	17.3	50.2	12.7	50.2	12.7	55.8	11.5	52.1	12.3
Cup	63.3	10.1	182.3	3.5	81.8	7.8	86.9	7.4	117.0	5.5
PEF-48										
N502-3										
Archive	25.5	25.1	67.8	9.4	36.5	17.5	38.5	16.6	47.6	13.4
Archive aged	38.2	16.7	63.7	10.1	57.2	11.2	53.2	12.0	58.0	11.0
Cup	55.9	11.4	128.4	5.0	88.3	7.2	106.6	6.0	107.8	5.9
P701-4										
Archive	27.8	23.0	53.8	11.9	51.4	12.5	57.5	11.1	54.2	11.8
Archive aged	45.6	14.0	71.7	8.9	74.1	8.6	82.2	7.8	76.0	8.4
Cup	74.0	8.6	188.3	3.4	91.1	7.0	301.3	2.1	193.5	3.3
PEF-49										
P714-5										
Archive	31.6	20.2	77.0	8.3	53.8	11.9	78.8	8.1	69.9	9.2
Archive aged	43.9	14.6	72.9	8.8	79.3	8.1	90.4	7.1	80.9	7.9
Cup	70.3	9.1	217.3	2.9	120.2	5.3	125.6	5.1	154.4	4.1
PR715-2										
Archive	23.8	26.9	39.4	16.2	40.8	15.7	53.8	11.9	44.7	14.3
Archive aged	38.2	16.7	52.6	12.2	74.1	8.6	77.9	8.2	68.2	9.4
Cup	62.4	10.4	131.3	4.9	104.3	6.1	115.3	5.6	117.0	5.5
T-40										
PR719-6										
Archive	26.5	24.1	41.3	15.5	41.3	15.5	45.2	14.2	42.6	15.0
Archive aged	40.1	16.0	52.6	12.2	56.5	11.3	70.6	9.1	59.9	10.7
Cup-Circum.	46.1	13.9	70.6	9.1	64.6	9.9	63.7	10.1	66.3	9.7
-Axial	46.2	13.9	79.3	8.1	58.7	10.9	98.3	6.5	78.8	8.1
NR520-2										
Archive	25.0	25.6	48.4	13.2	39.0	16.4	45.8	14.0	44.4	14.4
Archive aged	37.9	16.9	55.1	11.6	56.5	11.3	60.3	10.6	57.3	11.2
Cup-Circum.	34.6	18.5	82.2	7.8	62.8	10.2	71.8	8.7	72.3	8.9
-Axial	38.5	16.6	86.9	7.4	71.7	8.9	83.7	7.6	80.8	7.9

(a)

(b)

(c)

(d)

Fig. 1. AES absorbed current images of iridium samples from test assembly ECT-1.
(a) ECT-1/HFS, VFP-1; (b) ECT-1/HF2, wall; (c) ECT-1/HF2, VF-2; and (d) ECT-1/HF2, IFF-2.

(a)

(b)

Fig. 2 Microstructure of iridium sample L254-7 aged 6 mos @ 1310°C, (a) archive ring and (b) fueled cup. 100X.

(a)

(b)

Fig. 3. Microstructure of the iridium sample L256-8 aged 6 mos @ 1310°C, (a) archive ring and (b) fueled cup. 100X.

(a)

(b)

Fig. 4. Microstructure of iridium sample L255-6 aged 6 mos @ 1310°C, (a) archive ring and (b) fueled cup. 100X.

(a)

(b)

Fig. 5. Microstructure of iridium sample L257-R2 aged 6 mos @ 1310°C, (a) archive ring and (b) fueled cup. 100X.

(a)

(b)

Fig. 6. Microstructure of iridium sample N502-3 aged 6 mos @ 1310°C, (a) archive ring and (b) fired cup. 100X.

(a)

(b)

(c)

(d)

Fig. 7. Microstructure of iridium sample P701-4 aged 6 mos @ 1310°C. (a) archive ring and (b), (c), and (d) fueled cup. 100X.

(a)

(b)

Fig. 8. Microstructure of iridium sample P714-5 aged 6 mos @ 1310°C, (a) archive ring and (b) fueled cup. 100X.

(a)

(b)

(c)

(d)

Fig. 9. Microstructure of indium sample PR715-2 aged 6 mos @ 1310°C, (a) archive ring and (b), (c), and (d) fueled cup. 100X.

(a)

(b)

(c)

(d)

Fig. 10. Microstructure of control iridium samples aged 6 mos @ 1310°C, (a) archive PR719-6, (b) capsule cup PR719-6, (c) archive NR520-2, and (d) capsule cup NR520-2. 100X.

Fig. 11. After the sequential 1.4- and 5.1-MPa overpressure tests the PAL-002 aeroshell was broken. Note: The narrow nonclosure surface (up in the figure) was exposed to the blast wave.

Fig. 12. The impact shell containing capsules IRG-117 and IRG-118 was broken after the 5.1-MPa blast. Simulant fueled clad IRG-118 is visible.

Fig. 13. The impact shell containing simulant fueled clad IRG-115 and IRG-116 appeared undamaged after the 5.1-MPa blast.

IRG-115

IRG-116

IRG-117

IRG-118

Fig. 14. Simulant fueled clads from PAL-002 after the sequential overpressure tests at 1.4 and 5.1 MPa.

Fig. 15. The simulant fuel (UO_2) in the clad IRG-118 was broken into relatively large fragments after the two sequenced explosion tests.

Fig. 16. Temperature dependence of the diffusion constant for helium in $^{238}\text{PuO}_2$ subjected to a $227^\circ\text{C}/\text{min}$ heating ramp.

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