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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. Phosphorus-Effects Experiment (D. Pavone)

Four General-Purpose Heat Source (GPHS) fueled clads were aged at 1310°C for 6 months. The samples differed in the composition and treatment of the fuel in the iridium alloy (DOP-26) clads. After aging, the appearances of the areas around the vent exits of the capsules varied considerably. PEF-44 (nominal composition, vacuum outgassed fuel pellet) had a small amount of effusate deposited around the exit, PEF-45 (nominal composition, not outgassed) had no visible effusate, PEF-48 (phosphorus doped, outgassed) had a moderate quantity of effusate around the vent and a clear hexagonal pattern of deposits away from the vent, and PEF-49 (high cationic impurities, phosphorus doped, and outgassed) had a large mound of deposited material covering the vent exit.

Metallographic examinations of sections taken through the vents are shown in Figs. 1-4. The vent hole in PEF-44 (Fig. 1) was full of deposits, even though only a small amount of a two-phased, nonmetallic material

was present on the outer surface. The filter element also contained deposits. There were deposits in the vent hole of PEF-45 (Fig. 2), but none on the outer surface. The filter element entrances contained nonmetallic deposits. There were substantial quantities of nonmetallic deposits in the vent hole of PEF-48 (Fig. 3), as well as a layer of iridium-tungsten that had been vapor deposited there. We do not know if the closed appearance of the hole is real or just a result of the sample having been cut skew to the vent-hole axis. There were appreciable quantities of nonmetallic deposits in the vent structure and evidence of grain-boundary attack in the capsule and in the vent plates. The vent hole of PEF-49 (Fig. 4) was relatively free of deposited material, but there was significant evidence that the capsule wall had been attacked. The filter element was well filled with nonmetallic material.

The various deposits found in these cross-sections were analyzed by electron microprobe, and results are listed in Table I. The terms "hot side" and "cold side" refer to the circumferential position of the vent that faced toward or away from the second aging assembly, respectively. The letter designations refer to the various types of deposits observed.

Fig. 1. Metallographic cross section through the vent of PEP-44 (50X).

Fig. 2. Metallographic cross section through the vent of PEP-45 (50X).

Fig. 3. Metallographic cross section through the vent of PEF-48 (50X).

Fig. 4. Metallographic cross section through the vent of PEF-49 (50X).

TABLE I. Constituents of Vent Deposits in the Iridium Clads Used in the Phosphorus Effects Experiment

Capsule		Major Elements	Minor Elements
PEF-44			
Vent Entrance	(a)	Ca-Pu-Mg-O-Si	Fe-Ti-Cr
Cold Side	(b)	Pu-O	
Center	(a)	Si-O	
	(b)	Ca-Si-Pu-Mg-O	Al-Ti-Cr-Mn
Vent Entrance	(a)	Ca-Si-Pu-Mg-O	Ti-Fe-Mn
Hot Side			
Clad Exterior	(a)	Al-Ca-Si-Pu-O	Ti
	(b)	Ca-Si-Pu-O	
PEF-45			
Vent Entrance	(a)	Pu-O	
Cold Side	(b)	Si-O	Al-Ca-Pu-Mg-Ti
Vent Hole	(a)	Ca-Al-O	Pu
	(b)	Ca-Al-O	Pu-Si
	(c)	Ca-O-Ta	Al-Si-Pu-Th-U
Vent Entrance	(a)	Pu-O	Ca-Mg-Mn-Ti-Cr
Hot Side	(b)	Pu-Si-O	
PEF-48			
Vent Entrance	(a)	Ca-Pu-P-O	Si-Mg
Cold Side	(b)	Pu-P-O	Ca-Si-Al-Fe-Mn-Ni-Mg
	(c)	Si-O	Pu-Mg-P-Ca-Al-Ti
Center	(a)	Pu-P-Ca-O	W-Si-Ti-Fe-Mg-Al-Cr-Mn
	(b)	Si-O	
Vent Entrance	(a)	Pu-P-Ca-O	Mg
Hot Side	(b)	Si-O	Fe-Mg-Mn-Ca-Ti
	(c)	W-O	Si-Ca-Pu-Mg-Fe-Mn-Ti-Al
Vent Hole	(a)	Pu-O	Si-Ca-P
	(b)	Ir-W-Pu	Ca-O
	(c)	Ir-W	O
	(d)	Pu-O	Si-Ca-P-Ti
Clad Exterior	(a) ^a	Ir	Pu-O
	(b) ^a	Ir	Si-Pu-O
	(c)	Pu-O	Al-Ca-Si
	(d)	Al-Ca-O	Pu
	(e)	W	Pu-O
Clad Interior	(a)	Cr-O	Fe-Mg-Al
	(b)	Si-O	

TABLE I. Continued

Capsule		Major Elements	Minor Elements
PEF-49			
Vent Entrance	(a)	Ca-Pu-P-Si-O	Al-Ti
Cold Side	(b)		
Center	(a)	Pu-Si-O	Ca-Al-Ti
Vent Entrance	(a)	Ca-Pu-P-O	Mg-Al
Hot Side	(b)	Pu-Si-O	Ca-Al-P-Ti
Vent Hole	(a)	Ir	Ti
	(b)	Pu-O	Si-Ca-Al-Ti
	(c)	Ca-Si-Pu-O	Al-Ti
Vent Foil	(a)	Pu-Si-O	Ca-Al-Ti
Clad Exterior	(a)	Pu-O	Ca-Al
	(b) ^a	Ir	Al
	(c) ^a	Ir-Si-Pu	Al-O
	(d)	Ir	W (Approx. 5X bulk)
	(e) ^a	Ir-Al	O

^aIntermetallic reaction product.

A few general observations can be made. Plutonium appears to be transported principally in association with calcium, silicon, and magnesium, and to a lesser extent with aluminum, or as plutonium oxide. Phosphorus was found to transport primarily as a calcium-plutonium-phosphorus-oxygen compound. A phase rich in silicon-oxygen is frequently observed. X-ray dot maps showing the distribution of elements in selected areas are shown, along with a photograph of the deposit (SEM), in Figs. 5-9. Titanium was observed in the iridium grains adjacent to the grain-boundary deposit in PEF-49 (Fig. 9). It is not known yet what role it played in the grain-boundary reaction.

B. Early Compatibility Test II (D. Pavone)

The aging exposure of the second test assembly was terminated after a 90-day exposure at a clad temperature of 1310°C. The components were subjected to post-mortem examinations in the as-aged condition without having been impacted, as the first assembly had been.¹ Photographs of the fueled clads, identified as HF-8 and HF-9, are shown in Fig. 10: both exteriors appeared

bright and metallic; a small ring of gray, metallic material surrounded the vent exit of each.

The shield cups of both fueled clads had been fabricated from iridium alloy sheet LR 234, the same sheet used for the vent cups in the first early compatibility test. The vent cup of HF-8 was fabricated from sheet LR 236 and that of HF-9 from sheet L 237. Both axial and circumferential cross sections of the four cups were prepared metallographically and examined. Typical microstructures are shown in Figs. 11-14. All four cups were fine grained, with larger grains near the interior surfaces. The grain sizes of the four cups are listed in Table II.

The grain sizes are smaller than the 7-8 grains in an equivalent thickness that would be predicted from the earlier Multi-hundred Watt data. Of special interest is the fact that the grains in both shield cups were reasonably fine and uniform, unlike the grains in the vent cups in the first test, even though all were made from the same iridium sheet (LR 234). This indicates that the large, nonuniform grains observed in the earlier vent cups were not an inherent characteristic of the LR 234 sheet, but were environmentally induced.

Fig. 5. Distributions of plutonium, phosphorus, calcium, magnesium, aluminum, and silicon in the vent-filter deposits of PEP-48.

SEM

Ir

W

Pu

Si

Ca

O

Fig. 6. Distributions of iridium, tungsten, plutonium, silicon, calcium, and oxygen in vent-hole deposits of PEF-48 show a strong association of the last four elements.

Fig. 7. Distributions of iridium, plutonium, silicon, and oxygen in the intermetallic reaction product on the exterior of the clad of PEF-48.

Fig. 8. Distributions of calcium, aluminum, silicon, plutonium, phosphorus, and titanium in the vent deposit of PEF 49.

Fig. 9. Distributions of calcium, aluminum, silicon, titanium, plutonium, and oxygen in a nonmetallic deposit in a grain boundary near the vent hole of PEF-49.

(a)

(b)

Fig. 10. Vent-end faces and profile views of the GPHS fueled clads from the second early compatibility test: (a) HF-8 and (b) HF-9 (1.5X).

Fig. 11. Typical microstructures of the vent cup of HF-8: (a) axial cross section, (b) transverse cross section (50X).

Fig. 12. Typical microstructures of the shield cup of HF-8: (a) axial cross section, (b) transverse cross section (50X).

Fig. 13. Typical microstructures of the vent cup of HF-9: (a) axial cross section, (b) transverse cross section (50X).

Fig. 14. Typical microstructures of the shield cup of HF-9: (a) axial cross section, (b) transverse cross section (50X).

TABLE II. Grain Sizes of the Iridium Cups in the Second Early Compatibility Test

Capsule	Grain Size			
	Intercept (μm)		Grains/640- μm Thickness	
	Transverse	Axial	Transverse	Axial
HF-8				
Vent Cup (LR 236)	56.0	59.2	11.4	10.8
Shield Cup (LR 234)	53.2	58.8	12.0	10.9
HF-9				
Vent Cup (LR 237)	53.9	49.3	11.9	13.0
Shield Cup (LR 234)	52.4	55.2	12.2	11.6

Fig. 15. Microstructures of the iridium clads adjacent to the vent holes: (a) HF-8 (b) HF-9 (50X).

The comparatively fine grains adjacent to the vent holes in HF-8 and HF-9 (Fig. 15) are in sharp contrast to those observed in the HF-2 and HF-5 vent cups of the first test. As previously reported^{2,3} those clads both had large quantities of iridium-silicon intermetallic reaction products on the exterior surface adjacent to the vent hole

and grain sizes of nearly one grain across the thickness. This observation strongly suggests that grain growth is accelerated by fuel impurities that vaporize through the vent, react with graphite, and then react with the iridium boundaries. The suspected species are phosphorus, sulfur, and silicon.

II. SYSTEM COMPATIBILITY (D. Peterson)

We have obtained a copy of the SOLGASMIX computer program, which we will install along with a suitable data base of thermodynamic functions. This program will be used to model the compatibility and transport of normal components and impurities in radioisotopic thermoelectric generators. Its first use will be to try to understand the results observed in the GPHS early compatibility tests.

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