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ABSTRACT

This formal monthly report covers the studies related to the use of $^{238}\text{PuO}_2$ in radioisotopic power systems carried out for the Space and Terrestrial Systems Division of the U. S. Department of Energy by the Los Alamos National Laboratory.

Most of the studies discussed here are of a continuing nature. Results and conclusions described may change as the work continues. Published reference to the results cited in this report should not be made without the explicit permission of the person in charge of the work.

SPACE NUCLEAR SAFETY AND FUELS PROGRAM

MARCH 1981

Compiled by
S. E. Bronisz

I. GENERAL-PURPOSE HEAT SOURCE

A. Impact Tests (R. W. Zocher)

The schedule for the first Design Iteration test impact has been changed. Savannah River Plant was to have supplied the fueled clads for this test, but their inability to make acceptable closure welds has made this impossible. Instead, Savannah River will supply typical production pellets and Los Alamos will weld them into the iridium capsules. The revised schedule for the first test is shown in Fig. 1.

The impact shells that will be used in the Design Iteration test have 0.5-mm pyrolytic graphite coatings applied to their corners. Figure 2 shows the impact shells after outgassing for 36 h at 1500°C in vacuum. The corner coatings survived the heat treatment intact. The hole seen in the right-hand impact shell is for the thermocouple that will be used in the impact test.

B. Thermal Response (C. Frantz)

As part of a program to determine how fast the General-Purpose Heat Source (GPHS) module will heat up when it is placed in the Los Alamos impact gun, a module containing two $^{238}\text{PuO}_2$ fueled clads and two UO_2 simulant fueled clads (the configuration for the future safety tests) was loaded into the gun breach. The module was in the "flat" orientation contained in an impact can 130-mm diam by 76 mm high, made from 0.18-mm-thick stainless steel. The impact can was separated from the top of the projectile by three layers, a 0.025-mm nickel foil, a 6-mm layer of graphite felt, and another 0.025-mm nickel foil. Two chromel-alumel thermocouples were used, one entering through the back of the module and in close proximity to one of the fueled clads and one on the surface of the impact can.

The gun was evacuated to $<10^{-4}$ torr and backfilled with welding-grade helium to 20.7 kPa. After about 20 h the indicated temperatures were: iridium clad 304°C, impact can 173°C. Then the gun was evacuated. Temperatures vs time are given in Table I and Figs. 3 and 4 for the evacuated condition. After about 29 h in vacuum the iridium temperature was 783°C. When the gun was again evacuated to $<10^{-4}$ torr and backfilled with nitrogen, the iridium temperature was 545°C after about 20 h. Three layers of 0.025-mm nickel foil were added to the exposed surfaces of the impact can. After a time of >24 h, an iridium temperature of 875°C was obtained. This indicates that the impact temperature of 930°C may be obtainable by additional insulation on the impact can.

C. Iridium Impact Behavior (F. W. Schonfeld)

A review of all available General-Purpose Heat Source (GPHS) impact data and much of the Multi-hundred Watt (MHW) impact data has shown that both brittle and ductile behaviors can be displayed within the same iridium-alloy capsule or sphere; in several instances, brittle and ductile fractures were found in the same piece part, separated by only a few millimeters. An example of this brittle-ductile pairing is shown in Figs. 5 and 6.

This finding does not support the intuitive postulate that "good" and "bad" impact performances might be related to lot number, production run, or individual part. Rather, it suggests that the inherent deformation characteristics of the alloy are such that behavior is strongly influenced by stress state as well as strain rate and temperature. Since the complex interactions between fuel capsule and graphite produce large point-to-point variations in stress state and magnitude, impact response must be examined on a local rather than an overall basis. The noted similarities in the deformation and fracture behaviors of impacted GPHS capsules and MHW shells further suggest that the effects of test temperature and impact velocity may be of lesser importance than has been assumed. MHW impacts have usually been carried out at higher temperatures ($\sim 1450^{\circ}\text{C}$) and higher velocities ($\sim 80\text{ m/s}$) than have GPHS impacts ($\sim 800\text{--}1000^{\circ}\text{C}$, $\sim 60\text{ m/s}$), so it is possible that the similar responses indicate a temperature-compensated strain-rate sensitivity. However, that possibility does not mask the consistently differing responses to simple vs complex stressing. The few examples of high ductility found in GPHS and MHW tests, identified by large changes in length or in thickness, were the result of the "push-through" phenomenon. "Push-through" (or perhaps "push-deformation" is a better term) first produces a shear stress that is quickly converted into bending (by sliding of the iridium since it is imperfectly constrained) and a tensile component that operates between the two opposing bends. It is this largely uniaxial tensile loading that produces the significant thinning that has been observed. If the original shear stress is maintained by circumstances that impose relatively rigid restraints on the iridium, then complete "push-through" can take place, as illustrated in Fig. 7, while still exhibiting significant ductility. This view of events is in accord with the observed behavior of iridium-alloy tensile-impact specimens.

Why then is the ability of the alloy to deform plastically replaced by brittle fracture under other circumstances? The work of Hecker et al.¹ and of Rohr et al.² provide a rational and reasonable basis for understanding by making the following points:

1. Cleavage in iridium single crystals on (100) and (111) planes is intrinsic.
2. Brittle intergranular fracture in polycrystalline iridium is also intrinsic and not impurity related.
3. Grain boundaries contain many ledges, and a major fraction of these ledges are aligned with the $\{111\}$ planes in one or both of the adjacent grains, which facilitates crack nucleation and propagation along these $\{111\}$ planes and leads to grain boundary separation.

With these points the propensity of the iridium-alloy to deform plastically at one point while fracturing in a brittle manner at another is understandable. The presence of large numbers of grain-boundary ledges converts many of the grain boundaries into incipient cracks, and some of these incipient cracks must intersect the surface. Compression stresses do not open cracks, and the relatively good ductility exhibited by the alloy in uniaxial tension suggests that under favorable conditions surface cracks can be blunted by a plastic flow. However, when flow is constrained by biaxial or triaxial stress patterns the yield stress is increased and plastic flow is restricted to the material at the roots of the cracks. Since the cracks produced by grain-boundary ledges must be very sharp indeed, they can further increase elastic and plastic constraints to the point of catastrophic failure.

Many sources of biaxial loadings in impact tests of both MHW and GPHS assemblies must exist. The symmetry of the MHW case produces a simple situation for visualization, but the basic interactions are quite similar. In a MHW test the most obvious source of biaxial stresses is simple geometry; any distortion of a sphere reduces its ratio of volume to surface, thus any significant departure from sphericity of the essentially incompressible fuel ball must produce an internal "pressure." Another source of biaxial stressing is the actual flattening of the iridium and the fuel at the impact face. The mechanical action is not unlike reversing the draw-forming of the original hemisphere from flat sheet. This must produce circumferential tensile stresses that move outward as the flattened area increases. Radial forces and bending stresses must also be present on the face, but a more important source of biaxial stretching and fracture springs from the different characteristics of the materials involved. On impact, an increasing circular area of iridium is held firmly between the graphite and the fuel while both are expanding radially. The fuel and graphite are not ductile materials, both shatter into many fragments and so acquire a pseudo-viscous flow capability that permits movement. The movement outward from the center of the impact face takes place under the compressive force of the impact so that the frictional drag between iridium and the other components must be large and must generate biaxial stresses of significant magnitude. The mechanics of impact and the geometry of the system require that the materials of the outer shell (the graphite) move farther than the material on the inside (fuel), thus, it seems likely that the graphite begins to flow before the fuel does, and continues to flow after the fuel has approximated its final shape, imposing an additional stress gradient upon the iridium that contributes to the brittle intergranular fracture ("fingerprint") illustrated in Fig. 6.

Since the GPHS capsule is itself a reasonable approximation to a sphere, the observed differences in behavior must result from the differing surroundings. The GPHS graphite is much stronger than the filament-wound MHW graphite, so that the relative magnitudes of specific interactions may be quite different, but the basic causes of iridium failures remain unchanged.

D. Carbon-Monoxide Treatment (D. Peterson)

A thermodynamic analysis of the treatment of GPHS modules with carbon-monoxide was performed this month. Given a flowing gas with a total ($\text{CO} + \text{CO}_2$) pressure of 0.1 atm and assuming a 100°C gradient between the fuel and the graphite, a minimum fuel temperature of 1250°C would be required to attain a composition of $\text{PuO}_{1.99}$. At higher CO pressures in a static system or with lower fuel/graphite gradients, higher fuel temperatures would be required to effect the same reduction.

E. Reentry (J. Starzynski)

The first draft of a report describing the simulation of the reentry of IRG-62 was completed this month. Further analyses of the helium released during the pre-reentry heat treatment were performed using a two-particle model. The calculated curve for the two-particle case fits the experimental data better than the single-particle model.

II. SYSTEMS SUPPORT (D. Pavone)

A. Postmortem Examination of MHFT-74

The majority of the results of post-impact examination of this MHW production surveillance sample were reported in the February report.

Metallographic examination of cross sections of both vent assemblies revealed heavy depositions of nonmetallic materials in the pores of the iridium filter element, as illustrated in Fig. 8. In addition to a major phase and two minor

phases, a two-phase constituent appearing as a eutectic structure indicative of solidification from the liquid was present. This is the first occurrence for such a constituent in the series of Galileo samples examined. Electron microprobe analysis of the nonmetallic phases indicated that the compositions of the various phases were those listed in Table II. The appearance of phosphorus in the deposit suggests that phosphorus will also be detected in the Auger analysis of the iridium grain boundaries and correlate with the anomalous large grain size observed in the iridium containment shell.

A condensed summary of results of impact tests in the Galileo test series is presented in Table III. This tabulation will be up-dated as further tests are completed.

B. Iridium Grain Growth Experiments

Several samples of DOP-26 iridium from a sheet identified as L-267-4 were obtained from R. Heestand of Oak Ridge National Laboratory for additional experiments on the kinetics of grain growth of P-doped iridium.

A sample P-doped and annealed at 1330°C for 500 h showed a grain size of 14.7 grains/thickness. This result correlates well with previously obtained data.

C. Impurity Effects in Ir-0.3% W

The results of Auger analyses of the grain boundary chemistries of the iridium containments of two MHW and one GPHS assembly are shown in Table IV and summarized in Table V.

The results do not indicate any noteworthy impurities in the iridium of MHF-428. This assembly was treated for 37 months as a test assembly at 800°C. The iridium temperature was, therefore, considerably lower than that which exists at the operating temperatures of other MHW samples we have examined.

One hemishell of MHFT-68 showed small quantities of phosphorus, while the other hemishell had small quantities of sulfur. The reason for the inconsistent distribution of these elements is not clear. No failures were observed in the impact test, indicating that these impurities did not degrade the ability of the iridium to withstand the imposed deformation at high temperatures.

The iridium of IRG-61 did show a marked depletion of thorium at the inside edge. Severe iridium transport was also observed in this test capsule. Both effects, thorium depletion and iridium transport, have been related to the presence of oxygen in the test environment and, therefore, appear to be consistent observations.

III. LIGHT-WEIGHT RADIOISOTOPE HEAT UNIT (R. E. Tate)

A verbal agreement was reached with J. Terhune of NASA/Ames and M. Harris of General Electric Reentry Systems on a procedure to install a simulant fueled Light-Weight Radioisotopic Heater Unit (LWRHU) on an adapter plate for a high-G load test. The LWRHU will be held in a bracket designed for the Galileo Probe. Blind fasteners are used to attach the clamps and the nuts on these will be torqued to 15-18 in./lb. Torque will be applied with an 11/32-in. box wrench and a small spring balance, while the fastener is held from rotating with a 1/16-in. Allen key inserted in the hole in the tip of the fastener. Epoxy resin will be applied to the centrifuge adapter plate to support the convex mounting surface of the bracket. Grease will be applied to convex surface prior to assembly so the hardened epoxy will support the bracket, but not bond it to the adapter, which will permit eventual removal of it.

A pilot experiment has established that the UCAR C-34 cement for the LWRHU aeroshells meets the NASA/Ames non-volatile requirement without question. On the two samples tested the total mass loss was significantly less than the acceptable

1%; the mass loss was undetectable, less than 0.2% and may be zero. One hundred-milligram ribbons of freshly prepared cement were placed on squares of aluminum foil 50 mm on a side and the cement was cured at 130°C for 16 h in accordance with the instructions of Union Carbide. The samples of cement lost about 25% of their mass during the cure cycle. The cement/foil units were carefully weighed on an analytical balance reading to 0.1 mg. The foils were then folded and the units were placed in tubes on a high-vacuum manifold. The manifold was pumped to 2×10^{-6} torr and the two tubes were heated by electric furnaces to 125°C for 24 h. The cement/foil units were reweighed on the same balance after this vacuum heat treatment and were found to weigh exactly the same as before the treatment.

IV. ERRATA

The February monthly progress report, LA-8865-PR, was inadvertently printed with the wrong date. The correct date should read February 1981.

In Fig. 5 of the February monthly report, the magnification of (b) should be 85X and that of (c) should be 700X. In Fig. 13 of the same report (a) is the stem crack and (b) is the base crack.

REFERENCES

1. S. S. Hecker, D. L. Rohr, D. F. Stein, "Brittle Fracture in Iridium," Met-Trans. A, V9A, April 1978, pp. 481-487.
2. D. L. Rohr, L. E. Murr, and S. S. Hecker, "Brittle Fracture in Polycrystalline Ir-0.3 Pct W," Met. Trans. A, V10K, April 1979, pp. 299-405.

Fig. 1.

The current schedule for the first Design Iteration test.

Fig. 2.

The impact shells have corner coatings of pyrolytic graphite.

Fig. 3.

The iridium clad temperature in a GPHS module containing two fueled clads.

Fig. 4.
The first 1.45 ks of the curve in Fig. 3.

Fig. 5.
Metallographic cross section through a fuel fragment punch displacement on the impact face of the shell of MHFT-61.

Fig. 6.
Metallographic cross section through the fingerprint-type cracks on the impact face of the shell of MHFT-61.

(a)

(b)

Fig. 7.

A shear fracture in MHFT-62; (a) 7X, and (b) 50X.

Fig. 8.

Metallographic cross section of the filter element of MHFT-74 illustrating multi-phase nonmetallic deposits. 250X.

TABLE I
IRIDIUM CLAD TEMPERATURE VS TIME IN A GPHS MODULE
CONTAINING TWO FUELED CLADS

Time (ks)	Temp. Ir (°C)	Temp. Can (°C)	Pressure (mm Hg)	Time (ks)	Temp. Ir (°C)	Temp. Can (°C)	Pressure (mm Hg)
0	304	173	580	1.26	611		
.01	304			1.32	616		
.02	304			1.38	620		
.04	303			1.44	624		
.06	302			1.50	629		
.12	301			1.56	632		
.18	306			1.62	636		
.21	309		.01	1.74	643		
.24	315			1.92	653		
.30	330			2.22	667		
.36	353			2.52	681	153	
.42	379			2.82	693		
.48	408			3.12	702		
.54	437			3.42	711		
.60	465			3.72	716		
.66	492			4.02	725		
.72	515			4.32	731		
.78	536			4.62	736	208	<1 E-4
.84	553			5.22	744		
.90	566			5.82	750		
.96	578			7.02	758		
1.02	587			10.26	769	243	
1.08	594			13.14	772		
1.14	601			14.40	774		
1.20	607			15.06	774	247	
				102.60	783		

TABLE II
CONSTITUENTS OF NONMETALLIC VENT DEPOSITS IN MHFT-74

Major (medium gray) - Ca, P, Ba, Pu, Fe, Mn, Si, and O.

Minor (light gray) - Ca, P, Ba, Fe, Mn, Si, Ti, Cr, and O.

Minor (dark gray) - Ca, Ba, Pu, Fe, Ti, Si, and O.

Eutectic - Pu (strong), Ca, P, Ba, Fe, Si, and O.

TABLE III

IMPACT TEST SUMMARY OF GALILEO MHW FUEL-SPHERE ASSEMBLIES

Assembly Id.	Aging-Time Hours	Impact Test Conditions	Post Impact Dimensions			Failure Type				Iridium	
			Height mm	Diam. mm	% Strain Height/Diam. ^a	Hoop	Fingerprint ^b	Fuel Push-thru	Other	Type	Identification
MHFT-61	8834	1210°C-GIS Reentry T/Impact T Vacuum Velocity	31.09	43.46	-23.5/+ 6.9	No failures				DOP-26	5-1/5-4
MHFT-62	720	1550°C/1440°C 85 m/s	30.38	45.34	-25.6/+12.4	---	---	X	Weld bead Back side	HDR	467-4/468-1
MHFT-64	720	1475°C/1300°C 85 m/s	29.03	45.11	-28.7/+10.8		(X)	---	---	DOP-26	2-1/1-2
MHFT-65	4320	1550°C/1440°C 85 m/s	28.55	43.84	-28.7/+ 8.0		(X)	X	IF crack ex- tends hoop area	DOP-26	1-1/1-4
MHFT-66	720	1550°C/1440°C 85 m/s	30.68	43.64	-23.8/+ 6.9	---	X	---	---	HD	462-1/462-4
MHFT-67	100	1500°C/1430°C 74 m/s	29.64	43.66	-27.1/+ 7.4			No failures		DOP-26	J7-3/J9-2
MHFT-68	110	1792°C/1579°C 82 m/s	30.07	43.66	-27.6/+ 7.4			No failures		DOP-26	J4-4/J5-2
MHFT-69	100	-----/1037°C 82 m/s	30.99	43.71	-23.8/+ 7.6	---	(X)	---	Weld bead crack-IF	DOP-26	J6-1/J9-4
MHFT-70	720	1500°C/1430°C 32 m/s	29.16	43.26	-28.3/+ 6.4	---	X	X	---	DOP-26	J4-2/J4-3
MHFT-72	720	1500°C/1430°C 82 m/s	30.43	43.76	-25.1/+ 7.6			No failures		DOP-26	J22-4/J23-3
MHFT-74	720	1500°C/1430°C 31 m/s	29.74	43.54	-26.8/+ 7.1			No failures		DOP-26	JR50-1/JR51-2

^aBased upon nominal 40.64-mm diam except MHFT-62 thru -66 based upon MF gaging.

^bParentheses indicate penetration confirmed metallographically.

TABLE IV
AUGER ANALYSIS OF MHW AND GPHS IRIIDIUM

Specimen	Treatment	Location	T_{65}/Ir_{229}	P_{120}/Ir_{229}	C_{270}/Ir_{229}	C_{510}/Ir_{229}	S_{102}/Ir_{229}	S_{150}/Ir_{229}	T_{65}/Ir_{229}
MHW-42B-A1	37 mos. SIPS Assembly @ 800°C	Edge						0.17	
		Inside	0.81	- - -	0.30	0.39	- - -	P_{180} or C_{180}	0.10
		Center	0.90	- - -	- - -	- - -	- - -	- - -	0.10
		Edge Outside	0.96	0.15	0.30	0.51	- - -	- - -	0.11
MHW-42B-A2	37 mos. SIPS Assembly @ 800°C	Edge							
		Inside	0.76	- - -	0.87	0.68	- - -	- - -	0.08
		Center	0.66	- - -	0.39	0.63	- - -	- - -	0.07
		Edge Outside	0.49	- - -	0.87	0.55	- - -	- - -	0.06
MHW-42B-B1	37 mos. SIPS Assembly @ 800°C	Edge							
		Inside	1.16	- - -	0.15	- - -	- - -	- - -	0.13
		Center	0.96	- - -	- - -	- - -	- - -	- - -	0.10
		Edge Outside	0.69	- - -	- - -	- - -	?	- - -	0.07
MHW-42B-B2	37 mos. SIPS Assembly @ 800°C	Edge							
		Inside	1.10	- - -	0.15	0.41	- - -	- - -	0.13
		Center	1.05	- - -	- - -	- - -	- - -	- - -	0.12
		Edge Outside	0.74	- - -	0.20	0.26	Trace	- - -	0.09
MHW-68-A1	110 hours @ 1210°C-GIS	Edge							
		Inside	0.62	- - -	1.35	0.69	- - -	0.17	0.09
		Center	0.72	- - -	1.74	1.10	- - -	?	0.12
		Edge Outside	0.84	- - -	1.29	0.91	- - -	0.14	0.18
MHW-68-A2	110 hours @ 1210°C-GIS	Edge							
		Inside	0.67	- - -	0.91	0.93	- - -	0.17	0.09
		Center	0.74	- - -	0.76	0.64	- - -	0.16	0.03
		Edge Outside	0.84	- - -	1.47	1.23	- - -	0.11	0.11
MHW-68-B1	110 hours @ 1210°C-GIS	Edge							
		Inside	1.02	- - -	0.26	0.43	- - -	P_{180} or C_{180}	0.11
		Center	1.06	0.14	0.35	0.59	- - -	- - -	0.12
		Edge Outside	0.82	0.19	0.74	0.87	- - -	- - -	0.11
MHW-68-B2	110 hours @ 1210°C-GIS	Edge							
		Inside	0.78	0.07	0.25	0.29	- - -	- - -	0.09
		Center	0.94	0.11	0.18	0.19	- - -	- - -	0.12
		Edge Outside	0.83	0.17	0.35	0.35	- - -	- - -	0.10
ING-61-A	4613 hours @ 1300°C-Ir	Edge							
		Inside	0.21	- - -	0.61	0.49	- - -	0.05	0.02
		Center	0.71	Trace	0.83	0.70	- - -	- - -	0.08
		Edge Outside	0.71	- - -	1.16	0.70	- - -	0.07	0.07
ING-61-B	4613 hours @ 1300°C-Ir	Edge							
		Inside	0.10	- - -	0.27	0.24	- - -	- - -	0.01
		Center	0.95	- - -	0.20	0.19	- - -	- - -	0.11
		Edge Outside	0.98	- - -	0.49	0.40	- - -	- - -	0.11

TABLE V
SUMMARY OF AES RESULTS FOR MHW AND GPHS IRIIDIUM

<u>Specimen</u>	<u>Location</u>	<u>Th₆₅/Ir₂₂₉</u>	<u>P₁₂₀/Ir₂₂₉</u>	<u>S₁₅₀/Ir₂₂₉</u>
MHF-428	Inside	0.96	0	0
	Center	0.89	0	0
	Outside	0.72	0.15	0
	Average	0.86	0.05	0
MHFT-68	Inside	0.77	0.02	0.09
	Center	0.74	0.06	0.04
	Outside	0.83	0.09	0.06
	Average	0.78	0.06	0.06
IRG-61	Inside	0.16	0	0.03
	Center	0.83	0	0
	Outside	0.85	0	0.04
	Average	0.61	0	0.04