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Compiled by
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SPACE NUCLEAR SAFETY AND FUELS PROGRAM

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ABSTRACT

This formal monthly report covers the studies related to the use of $^{238}\text{PuO}_2$ in radioisotope power systems carried out for the Office of Coordination and Special Projects 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.

I. GENERAL-PURPOSE HEAT SOURCE (R. Zocher and C. Frantz)

A. Design Iteration Test

A revised test plan (CMB-5-C-81-22R) was written for the first Design Iteration Test (DIT-1) impact. Copies of the revised DIT-1 impact plan were distributed to attendees at the Eleventh Heat Source Status Review meeting in Miamisburg, Ohio, on April 22, 1981.

Extensive changes to the original test configuration were incorporated in the revised DIT-1 impact plan. Thermal calculations on the modified heat-source module by General Electric Company (GE) personnel now place the clad impact temperature for orbital decay at $930^\circ \pm 5^\circ\text{C}$. A corresponding operational temperature of 1306°C was calculated for the same module design modifications.

The impact plan now has, as the primary option, four fueled clads in the General-Purpose Heat Source (GPHS) module. Test procedures, hardware, and handling fixtures must be in place before the full-module test can be run. Work has been progressing on evaluation tests, handling fixtures, and procedural changes in support of the full-module impact outlined in the revised

DIT-1 impact plan. The revised DIT-1 schedule is given in Fig. 1.

A thermal test in the impact gun, using two fueled clads and two UO_2 -containing clads, was reported in the March Report. That test established the clad steady-state temperatures in helium and vacuum with a test module containing two FC's. However, the primary interest was the temperature-rise rate for the clad, since approximately the same rate will prevail for the clad in the DIT-1 test module. Test precision depends on firing the gun when the desired clad temperature has been reached. The concern was that the temperature rise would be so fast that this could not be accomplished, but the $20^\circ\text{C}/\text{min}$ rate is entirely satisfactory in this regard.

B. Module Handling Procedures

The DIT-1 module, which will contain four fueled capsules, will be hotter under any given condition than the two-capsule test assemblies handled up to now. The impact gun must be loaded in air; therefore, the graphite components of the module must be maintained below the temperature at which they oxidize.

Fig. 1. GPHS project schedule, DIT-1 (revised April 30, 1981).

To accomplish transfer of the module from its assembly station in the CMR Building to the impact facility at Ten Site, the transfer container shown in Fig. 2 was designed and built. The bottom of the container is the impact sabot. Tests with 250-W electric heaters showed that the finned cover would reach 78°C without forced cooling and 45°C with it.

C. Test Impact

The simulated GPHS module, made of a bulk graphite block and four copper pellets is shown in Fig. 3. It was impacted at 58 mps and 23°C to demonstrate the handling procedures and to determine if the module, which is a close fit in the catch tube, would hit flat.

The impact was flat. Dents in the copper pellets prevented highly accurate measurements of the post-impact diameters, but apparently the centrally located diametral strains ranged from 0.023-0.024.

Using procedures that will be used for DIT-1 took 146 s from the time the seal on the transfer container was broken until the gun was sealed. Thus, it appears that we

Fig. 3. The DIT-1 support test used a simulated module made of bulk graphite and copper.

will be able to carry out an impact test using a full module without endangering the integrity of the graphite.

D. Welding (R. Kent)

Los Alamos is continuing to support the production welding effort at Savannah River Plant. In April two development iridium capsules were welded over a $^{238}\text{PuO}_2$ GPHS pellet. After welding, the capsules were cut open and sent for metallographic examination. The same Los Alamos fuel pellet, GP58, was used for both welds. It remained free of surface cracks throughout the procedures.

E. In-Module Fuel Reduction (D. Peterson)

Mound Facility has proposed that GPHS modules be exposed to controlled reducing atmospheres before being put in the converter to suppress the formation of CO in the converter. The thermodynamics of this treatment were analyzed with the assumptions that the fuel would be at 1200°C, the graphite impact shell would be at 1000°C, and the reducing gas would contain 0.04 atm of CO. Equilibration of the CO with the graphite establishes the CO_2/CO ratio according to the reaction,

Fig. 2. An aluminum-alloy transfer container featuring forced-air cooling designed and built so the full GPHS modules can be stored and transferred.

Using the free energy of reaction, ΔG° , of -12284 cal at 1000°C , calculated from Kubaschewski and Alcock,¹ an equilibrium constant of $128.45 \text{ atm}^{-1/2}$ for reaction (1) was calculated from the relationship,

$$\Delta G^\circ = -RT \log K \quad (2)$$

At a total ($\text{CO} + \text{CO}_2$) pressure of 0.04 atm , the $P_{\text{CO}_2}/P_{\text{CO}}$ ratio calculated was 3.1128×10^{-4} . Equilibration of the CO_2/CO with the fuel oxygen at 1200°C is given by the reaction,

With $\Delta G^\circ = -36935$ cal and $K = 3.017 \times 10^5 \text{ atm}^{1/2}$, using the calculated $P_{\text{CO}_2}/P_{\text{CO}}$ ratio and the equilibrium constant for reaction (3) yields an oxygen pressure of $1.06 \times 10^{-18} \text{ atm}$. This oxygen level corresponds to a fuel composition of $\text{PuO}_{1.90}$ based on Atlas and Schleman² or $\text{PuO}_{1.88}$ based on Markin and Rand.³ Attainment of this composition would be achieved only in a long-term flow experiment or with numerous evacuations and refills with CO at 0.04 atm .

Two CO molecules are formed for every CO_2 molecule produced by the fuel reduction. Therefore, as reduction proceeds in a closed container, CO pressure increases, resulting in a higher oxygen potential in the gas and progressively less fuel reduction.

Raising the graphite temperature to 1100°C , while maintaining the fuel at 1200°C and the CO pressure at 0.04 atm , results in reduction to $\text{PuO}_{1.82}\text{-PuO}_{1.80}$, whereas a graphite temperature of 1200°C leads to a composition of $\text{PuO}_{1.75}\text{-PuO}_{1.80}$.

These theoretical conclusions are supported experimentally by the measurements of Kent and Zoher.⁴ They found, at a fuel temperature of 1227°C with a graphite temperature of 1027°C and a CO pressure of 0.08 atm , that the fuel was reduced to $\text{PuO}_{1.85}$, which correlates well with the composition of $\text{PuO}_{1.88}$ calculated from the data of Markin and Rand.³ Use of a higher CO pressure of 0.8 atm , with the same temperature distribution, produced less reduction with the fuel attaining $\text{PuO}_{1.95}$, as predicted.

Additional support for these thermodynamic results can be obtained from measurements by Peterson and Pavone⁵ who determined the rate of CO production from a Multi-Hundred Watt (MHW) fuel assembly. The wide

range of temperatures and CO pressures used in this experiment encompassed the conditions proposed by Mound. The fuel was reduced to approximately $\text{PuO}_{1.98}$, which is consistent with the CO pressures employed in most of the experiment. The production of CO from MHW units has been analyzed thermodynamically by Peterson and Mulford,⁶ who showed that the CO production was consistent with the thermochemical data for the PuO_{2-x} and CO/CO_2 systems.

In summary, the following conclusions can be drawn from this analysis:

1. Assuming $P_{\text{CO}} = 0.04 \text{ atm}$, $T_{\text{graph.}} = 1000^\circ\text{C}$, and $T_{\text{fuel}} = 1200^\circ\text{C}$, the fuel cannot be reduced below $\text{PuO}_{1.89}$, even in an extended flow experiment.
2. Given higher graphite impact shell (GIS) or fuel temperatures, further reduction would be achieved.
3. As the reduction proceeds, CO is produced thus limiting the reduction in a closed volume.
4. Use of a CO pressure higher than 0.04 atm or leaks of air into the treatment chamber could reoxidize the fuel following an initial reduction step. This effect would lead to lattice re-expansion and possible breakup of the pellet.
5. Evacuation of the treatment chamber reduces the CO pressure while increasing the GIS and fuel temperatures. Therefore, during the pumping phase, reduction of the fuel would be accelerated. Purging with argon should be considered to avoid this high-temperature excursion.
6. The free energy of reaction (1) becomes more positive as the temperature is lowered. Hence, the formation of CO_2 from CO may deposit carbon in the chamber's cooler regions, the net effect of which will be thinning the GIS during CO heat treatment. Keeping the external walls of the chamber as close to the GIS temperature as possible will reduce this effect.

II. LIGHT-WEIGHT RADIOISOTOPE HEAT SOURCE (R. Tate, R. Kent)

The loading plan for the remaining capsules was completed. The capsules will be loaded and welded next month.

Calorimetric measurements have been completed for all 58 of the LWRHU pellets encapsulated in December, 1980. These data, together with other nondestructive test data, are listed in Tables I through III.

TABLE I. NDT for Encapsulated RHU Pellets-1

Fuel Pellet		Capsule Body No.	α Swipe (Count/min) ^a	Helium Leak Rate (cm ³ /s)	Neutron Emission Rate (n/s-g ²³⁸ Pu)	Calorimetry (Watts)	Comments
No.	Weight (g)						
RU3- 8	2.665	021	0	< 9x10 ⁻¹⁰	5 215	1.116	
RU3- 9	2.663	022	0	< 9x10 ⁻¹⁰	5 889	1.113	
RU3-10	2.665	024	0	< 9x10 ⁻¹⁰	5 612	1.116	
RU3-11	2.664	029	0	< 9x10 ⁻¹⁰	5 803	1.111	
RU3-12	2.653	031	0	< 9x10 ⁻¹⁰	5 613	1.118	
RU3-13	2.663	032	0	< 9x10 ⁻¹⁰	5 460	1.117	
RU3-14	2.662	033	0	< 6x10 ⁻¹⁰	5 539	1.114	
RU3-15	2.662	042	0	< 6x10 ⁻¹⁰	5 547	1.116	
RU3-16	2.661	045	0	< 6x10 ⁻¹⁰	5 293	1.117	
RU4- 3	2.656	048	0	< 6x10 ⁻¹⁰	4 775	1.111	
RU4- 4	2.670	049	0	< 6x10 ⁻¹⁰	4 843	1.118	
RU4- 5	2.663	051	0	< 6x10 ⁻¹⁰	4 785	1.115	
RU4- 6	2.666	054	0	< 6x10 ⁻¹⁰	4 923	1.118	
RU4- 7	2.666	055	0	< 6x10 ⁻¹⁰	4 991	1.117	
RU4- 8	2.668	056	0	< 6x10 ⁻¹⁰	4 946	1.117	
RU4- 9	2.663	059	0	< 6x10 ⁻¹⁰	4 636	1.116	
RU4-10	2.667	060	0	< 6x10 ⁻¹⁰	4 616	1.118	
RU4-11	2.666	061	0	< 6x10 ⁻¹⁰	4 738	1.116	
RU4-12	2.666	062	0	< 6x10 ⁻¹⁰	4 725	1.116	
RU4-13	2.671	063	0	< 6x10 ⁻¹⁰	4 691	1.118	
RU4-14	2.668	065	0	< 9x10 ⁻¹⁰	4 763	1.117	
RU4-15	2.669	066	0	< 9x10 ⁻¹⁰	4 785	1.118	
RU4-16	2.666	067	0	< 9x10 ⁻¹⁰	4 775	1.116	

^aThe specifications are: α Swipe, <220 counts/min
Helium Leak Rate, <1x10⁻⁶ cm³/s
Neutron Emission Rate, <6000 n/s-g ²³⁸Pu
Calorimetry, 1.10 $\pm$ 0.03 watts

TABLE II. NDT for Encapsulated RHU Pellets-2

Fuel Pellet		Capsule Body No.	α Swipe (Count/min) ^a	Helium Leak Rate (cm ³ /s)	Neutron Emission Rate (n/s-g ²³⁸ Pu)	Calorimetry (Watts)	Comments
No.	Weight (g)						
RU5- 3	2.666	068	0	< 9x10 ⁻¹⁰	4 626	1.115	
RU5- 4	2.669	069	0	< 9x10 ⁻¹⁰	4 632	1.112	
RU5- 5	2.670	070	0	< 9x10 ⁻¹⁰	4 674	1.119	
RU5- 6	2.666	071	0	< 9x10 ⁻¹⁰	4 723	1.119	
RU5- 7	2.669	073	0	< 9x10 ⁻¹⁰	4 739	1.116	
RU5- 8	2.668	075	0	< 9x10 ⁻¹⁰	4 674	1.117	
RU5- 9	2.668	076	0	< 9x10 ⁻¹⁰	4 292	1.119	
RU5-10	2.668	078	0	< 9x10 ⁻¹⁰	4 259	1.117	
RU5-11	2.671	079	0	< 9x10 ⁻¹⁰	4 377	1.118	
RU5-12	2.668	084	0	< 3x10 ⁻¹⁰	4 398	1.117	
RU5-13	2.668	089	0	< 3x10 ⁻¹⁰	4 350	1.117	
RU5-14	2.666	090	0	< 3x10 ⁻¹⁰	4 449	1.117	
RU5-15	2.668	091	0	< 3x10 ⁻¹⁰	4 492	1.118	
RU5-16	2.658	093	0	< 3x10 ⁻¹⁰	4 458	1.113	

^aThe specifications are: α Swipe, <220 counts/min
Helium Leak Rate, <1x10⁻⁶ cm³/s
Neutron Emission Rate, <6000 n/s-g ²³⁸Pu
Calorimetry, 1.10 $\pm$ 0.03 watts

TABLE III. NDT for Encapsulated RHU Pellets-3

Fuel Pellet No.	Weight (g)	Capsule Body No.	α Swipe (Count/min) ^a	Helium Leak Rate (cm ³ /s)	Neutron Emission Rate (n/s-g ²³⁸ Pu)	Calorimetry (Watts)	Comments
RU7- 2	2.660	094	0	< 3x10 ⁻¹⁰	4 252	1.116	
RU7- 3	2.665	095	0	< 3x10 ⁻¹⁰	4 240	1.115	
RU7- 4	2.664	096	0	< 3x10 ⁻¹⁰	4 532	1.115	
RU7- 6	2.665	097	0	< 3x10 ⁻¹⁰	4 527	1.114	
RU7- 7	2.664	098	0	< 3x10 ⁻¹⁰	4 492	1.116	
RU7- 8	2.663	099	0	< 3x10 ⁻¹⁰	4 468	1.114	
RU7- 9	2.663	100	0	< 3x10 ⁻¹⁰	4 329	1.116	
RU7-10	2.662	101	0	< 3x10 ⁻¹⁰	4 343	1.115	
RU7-11	2.662	102	0	< 1x10 ⁻¹⁰	4 411	1.114	
RU7-12	2.662	103	0	< 1x10 ⁻¹⁰	4 498	1.116	
RU7-13	2.661	105	0	< 1x10 ⁻¹⁰	4 495	1.113	
RU7-14	2.657	107	0	< 1x10 ⁻¹⁰	4 395	1.116	
RU7-15	2.661	108	0	< 1x10 ⁻¹⁰	4 504	1.119	
RU7-16	2.659	109	0	< 1x10 ⁻¹⁰	4 508	1.120	
RU10-1	2.661	110	0	< 1x10 ⁻¹⁰	4 722	1.121	
RU10-2	2.660	112	0	< 1x10 ⁻¹⁰	4 686	1.120	
RU10-3	2.660	114	0	< 1x10 ⁻¹⁰	4 917	1.120	
RU10-4	2.660	115	0	< 1x10 ⁻¹⁰	5 042	1.120	
RU10-5	2.663	116	0	< 1x10 ⁻¹⁰	4 939	1.119	
RU10-6	2.663	117	0	< 1x10 ⁻¹⁰	4 918	1.120	
RU10-7	2.662	118	0	< 1x10 ⁻¹⁰	4 981	1.118	

^aThe specifications are: α Swipe, <220 counts/min
Helium Leak Rate, <1x10⁻⁶ cm³/s
Neutron Emission Rate, <6000 n/s-g ²³⁸Pu
Calorimetry, 1.10 $\pm$ 0.03 watts

III. FUEL

A. Characterization

Spectrochemical data for four lots of SRP feed powder are listed in Table IV. Neutron emission rate data for

seven lots of as-received feed are listed in Table V. Particle size data for seven lots of as-received feed, together with data for five lots ball-milled 44 h, are listed in Table VI. Isotopic data for two lots of feed enriched at 80 at.% ^{238}Pu and for one enriched at 83.5 at.% ^{238}Pu are listed in Table VII.

TABLE IV. Spectrochemical Data for Feed Lots
(ppm by Weight)

Species	Sensitivity Limit	Lot 46	Lot ^a 47	Lot 53	Lot 56
Al	5	80	15	90	20
B	1	<	<	<	<
Ba	2	<	40	15	<
Bi	1	<	<	<	<
Ca	3	250	50	15	35
Cd	10	<	<	<	<
Co	2	3	<	<	<
Cr	5	65	120	120	80
Cu	1	15	<	3	<
Fe	5	350	170	270	190
Mn	1	15	10	4	4
Ni	5	45	25	40	70
Pb	5	10	<	<	<
Si	5	110	180	85	100
Sn	5	10	<	<	<
Zn	5	<	15	20	<

^aLot 47 was analyzed after the material was processed at Los Alamos. The other data are for as-received lots.

TABLE V. Neutron Emission Rate Data for As-Received Feed Lots

Los Alamos Lot	SRP No.		n/s-g ²³⁸ Pu
	Lot	Run	
45	411	802773	17 987
46	418	805808	18 198
47	421	806815	18 743
56	408	712754	17 021
57	475	006118	16 516
58	409	801759	17 869
60	475	006119	17 190
			17 646
			± 769

TABLE VI. Particle-Size Data for Feed Lots

Los Alamos Lot	Mass Median Diameter (μm)	
	As-Received	Ball-Milled
		44 h
45	2.96	0.56
46	3.45	0.79
47	2.63	0.65
56	3.30	0.79
57	3.72	0.94
58	3.65	-
60	3.45	-
3.31		0.75
±0.39		±0.15

**TABLE VII. Isotopic Data for Feed Lots
(Weight Percent)**

Los Alamos Lot	Lab	Plutonium Isotope				
		238	239	240	241	242
58	SR	80.61	15.98	2.61	0.64	0.16
	Los Alamos	80.55	16.03	2.62	0.63	0.16
60	SR	83.15	14.22	2.05	0.43	0.15
	Los Alamos	83.21	14.15	2.06	0.43	0.15
61	SR	80.56	16.17	2.52	0.58	0.17
	Los Alamos	80.53	16.19	2.52	0.58	0.18

^aSR values corrected for decay to date of Los Alamos analyses.

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