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ISOTOPIC POWER FUELS MONTHLY STATUS REPORT FOR JULY 1973

Eugene Lamb

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ISOTOPES DEVELOPMENT CENTER

ISOTOPIC POWER FUELS MONTHLY STATUS REPORT FOR JULY 1973

Eugene Lamb

Isotopes Division

AUGUST 1973

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ISOTOPIC POWER FUELS MONTHLY STATUS REPORT FOR JULY 1973

Eugene Lamb

CURIUM-244 FUEL DEVELOPMENT

(Division of Space Nuclear Systems Program LR 30 01 03 3)

$^{244}Cm_2O_3$ Compatibility Program

Compatibility Couple Tests
(T. A. Butler and J. R. DiStefano)

The status of the 900, 1100, and 1400°C compatibility test matrix on July 31, 1973, is summarized in Table 1. Arrangements were made with HRLEL personnel to begin examination of the couples contained in Sets 3, 4, 5, and 6 which are scheduled for completion of the heat soak early in August.

Cm_2O_3 Compatibility With Ceramic Materials *(P. Angelini)*

The following experiments have been completed:

Experiment	Material	Temperature (°C)	Time (hr)
E	ZrO_2	1500	24
F	ZrO_2	1300	100

The specimens have been mounted and are being polished. No optical microscopy has yet been performed.

The optical microscopy is almost complete on the ThO_2 compatibility couples. The first optical results show that very little reaction occurred at 1300°C. Some reaction was seen at 1500°C to an extent of ∿20 mils into the Cm_2O_3. These specimens are being prepared for examination with the electron-beam microprobe.

The Al_2O_3 compatibility capsules and inserts have been received.

Reaction of Cm_2O_3 With Pt–26% Rh–8% W Alloy *(E. E. Ketchen)*

The alloy Pt–26% Rh–8% W (53.33 atom % Pt, 39.82 atom % Rh, 6.86 atom % W) is being considered as a capsule material to contain $^{244}Cm_2O_3$ heat source material. Will the Cm_2O_3 product react with this alloy?

The purification of the curium produced includes plutonium removal. However, the curium product probably would be held at least a year after the purification step before it is fabricated as a heat source. During this

Table 1. Status of Compatibility Tests

Set[a]	Start of Test	Thermal Cycles	Cumulative Hours	Projected End of Test
1	5-3-73	None	2136	11-28-73
2	5-3-73	None	2136	11-28-73
3	4-16-73	3	2416	8-4-73
4	4-16-73	3	2416	8-4-73
5	4-17-73	2	2294	8-9-73
6	4-17-73	2	2294	8-9-73
7	5-3-73	2	2023	12-3-73
8	5-3-73	2	2023	12-3-73
9	5-3-73	2	2016	12-3-73
10	5-3-73	2	2016	12-3-73

[a]Set identifications:

1. 900°C; 5000 hr; helium atmosphere; Ir, C, Pt, Hf—1% Pt—0.5% Pd, Hastelloy C-276, Haynes 25, Haynes 188, ThO_2, Pt—20% Rh, and Pt—26% Rh—8% W.
2. 900°C; 5000 hr; graphite, helium atmosphere; Ir, Pt, Hf—1% Pt—0.5% Pd, Hastelloy C-276, Haynes 25, Haynes 188, ThO_2, Pt—20% Rh, and Pt—26% Rh—8% W.
3. 1100°C; 2500 hr; graphite, helium atmosphere; Ir, Pt, Pt_3Ir, and Pt—26% Rh—8% W—0.5% Ti.
4. 1100°C; 2500 hr; dynamic vacuum; Ir, Pt, Pt_3Ir, C, and Pt—26% Rh—8% W—0.5% Ti.
5. 1100°C; 2500 hr; helium atmosphere; Ir, Pt, Pt_3Ir, C, and Pt—26% Rh—8% W—0.5% Ti.
6. 1100°C; 2500 hr; graphite, dynamic vacuum; Ir, Pt, Pt_3Ir, and Pt—26% Rh—8% W—0.5% Ti.
7. 1400°C; 5000 hr; graphite, helium atomsphere; Ir, C, Mo, Mo—46% Re, Ta, T-111, W, and W—26% Re.
8. 1400°C; 5000 hr; dynamic vacuum; Ir, Mo, Mo—46% Re, Ta, T-111, W, and W—26% Re.
9. 1400°C; 5000 hr; helium atmosphere; Ir, Mo, Mo—46% Re, Ta, T-111, W, and W—26% Re.
10. 1400°C; 5000 hr; graphite, dynamic vacuum; Ir, C, Mo, Mo—46% Re, Ta, T-111, W, and W—26% Re.

period of time about 5% of the ^{244}Cm would have decayed to ^{240}Pu. The procedure used to reduce the CmO_2 to Cm_2O_3 before the fabrication step would not convert the PuO_2 to Pu_2O_3. Hence the plutonium is present as PuO_2 in the Cm_2O_3 product. For purposes of this thermodynamic study, it was assumed that the Cm_2O_3 contained 5 mole % PuO_2.

The thermodynamic data needed to calculate the free energy of mixing of 95 mole % Cm_2O_3 with 5 mole % PuO_2 are not available. Since this partial molar free energy of mixing is probably small, it was neglected in the following calculations.

The partial molar free energies of mixing of Pt, W, and Rh were calculated using the following equation.[1]

$$\Delta\overline{G}_k = -RT \ln x_k + V_k[\sum_i(\delta_k - \delta_i)\phi_i]^2 \qquad (1)$$

where

$\Delta\overline{G}_k$ = Gibbs partial molar free energy of the k'th component

R = 1.9872 cal

T = absolute temperature

x_k = mole fraction of the k'th component

V_k = volume of the k'th component

δ_k = solubility parameter of the k'th component

δ_i = solubility parameter of the i'th component

ϕ_i = volume fraction of the i'th component

The solubility parameters were taken from data by Hildebrand and Scott.[1] In the derivation of Eq. 1, it was assumed that there was no compound formation of the metals with each other. The partial molar free energies are shown in Table 2 at 600-1000°C.

Table 2. Partial Molar Free Energies of Mixing of Components in Pt—26% Rh—8% W

Temperature (°C)	$-\Delta\overline{G}$ (cal/mole)		
	Pt	W	Rh
600	979	4407	1563
800	1229	5471	1930
1000	1481	6535	2294

[1]J. H. Hildebrand and R. L. Scott, *The Solubility of Non-electrolytes*, Reinhold Publishing Corporation, New York, 1950, pp. 198-209 and 320-347.

The free energies of formation of Pt_3O_4, WO_2, Rh_2O, Cm_2O_3, PuO_2, and Pu_2O_3 were calculated from a data compilation by Alvin Glassner[2] and a review article by F. L. Oetting[3] and are shown in Table 3. Since the free energies of Pt_3O_4 are positive, this reaction will not proceed in the presence of oxygen at 600-1000°C. However, the free energies of tungsten and rhodium with oxygen over the 600-1000°C temperature range are negative, and these reactions could proceed.

Table 3. Gibbs Free Energy of Formation of Pt_3O_4, WO_2, Rh_2O, Cm_2O_3, PuO_2, and Pu_2O_3

Temperature (°C)	ΔG_f (cal/mole)					
	Pt_3O_4	WO_2	Rh_2O	Cm_2O_3[a]	PuO_2	Pu_2O_3
600	10,300	-101,160	-11,350	-352,120	-215,550	-352,120
800	26,400	-93,850	-9,090	-339,320	-207,050	-339,320
1000	42,080	-86,780	-6,940	-326,900	-198,700	-326,900

[a]Pu_2O_3 data used as stand-in for Cm_2O_3.

The free energy data for Pu_2O_3 were used as a stand-in for Cm_2O_3 over the 600-1000°C temperature range. At higher temperature, where the free energies of Cm_2O_3 could be calculated, the Pu_2O_3 values were in good agreement with Cm_2O_3. Hence the use of the free energy values of Pu_2O_3 as a substitute for Cm_2O_3 may be a reasonable approximation.

The free energies from Tables 2 and 3 were used to calculate the free energies for Eqs. 2-5. The free energies for Eqs. 2-5 are shown in Table 4.

$$3W(\text{in Pt—26\% Rh—8\% W alloy})(s) + 2Cm_2O_3(s) \rightarrow 4Cm(s) + 3WO_2(s) \quad (2)$$

$$6Rh(\text{in Pt—26\% Rh—8\% W alloy})(s) + Cm_2O_3(s) \rightarrow 3Rh_2O(s) + 2Cm(s) \quad (3)$$

$$W(\text{in Pt—26\% Rh—8\% W alloy})(s) + 4PuO_2(s) \rightarrow WO_2(s) + 2Pu_2O_3(s) \quad (4)$$

$$2Rh(\text{in Pt—26\% Rh—8\% W alloy})(s) + 2PuO_2(s) \rightarrow Rh_2O(s) + Pu_2O_3(s) \quad (5)$$

Table 4. Gibbs Free Energy of Reaction for Eqs. 2-5

Temperature (°C)	ΔG_r (cal/reaction)			
	Eq. 2	Eq. 3	Eq. 4	Eq. 5
600	413,980	327,450	61,190	70,750
800	413,500	323,630	61,160	69,540
1000	413,110	319,840	60,700	68,120

[2]Alvin Glassner, *The Thermochemical Properties of the Oxides, Fluorides, and Chlorides to 2500°K*, ANL-5750, Argonne National Laboratory (1957).
[3]F. L. Oetting, "The Chemical Thermodynamic Properties of Plutonium Compounds," *Chem. Rev.* 67, 261-97 (June 1967).

Since the free energies are positive for the reactions to form WO_2 and Rh_2O from the alloy and Cm_2O_3 or PuO_2, this reaction should not occur at 600-1000°C.

In conclusion it appears from thermodynamic considerations that the Pt—26% Rh—8% W alloy will not react with Cm_2O_3 fuel containing 5% PuO_2 at 600-1000°C. The calculations are based on the assumption that there is no compound formation between the metals in the alloy.

Reaction of Cm_2O_3 With T-111 Alloy *(E. E. Ketchen)*

The alloy T-111 (90.09 atom % Ta, 7.88 atom % W, and 2.03 atom % Hf) is being considered as a capsule material to contain $^{244}Cm_2O_3$ heat source material. Will the Cm_2O_3 product react with this alloy?

The partial molar free energies of mixing for Ta, W, and Hf were calculated using the method described in the previous section, "Reaction of Cm_2O_3 With Pt—26% Rh—8% W Alloy." The results are shown in Table 5 from 600-1000°C. It should be noted again that this method of calculation assumes no compound formation of the metals with each other.

Table 5. Partial Molar Free Energies of Mixing of Components in T-111

Temperature (°C)	$-\Delta\overline{G}$ (cal/mole)		
	Ta	W	Hf
600	181	3935	3836
800	222	4850	4752
1000	264	5766	5667

The free energies of formation of Ta_2O_5, WO_2, HfO_2, Cm_2O_3, PuO_2, and Pu_2O_3 were calculated from a data compilation by Glassner[4] and a review article by Oetting.[5] The results are shown in Table 6. Since the free energies of formation of Ta_2O_5, WO_2, and HfO_2 are negative over the 600-1000°C temperature range, the metals will react with oxygen.

Table 6. Gibbs Free Energy of Formation of Ta_2O_5, WO_2, HfO_2, Cm_2O_3, PuO_2, and Pu_2O_3

Temperature (°C)	$-\Delta G_f$ (cal/mole)					
	Ta_2O_5	WO_2	HfO_2	Cm_2O_3[a]	PuO_2	Pu_2O_3
600	395,690	101,160	225,450	352,120	215,550	352,120
800	374,820	93,850	216,450	339,320	207,050	339,320
1000	354,180	86,780	207,770	326,900	198,700	326,900

[a]Pu_2O_3 data used as stand-in for Cm_2O_3.

[4]Glassner, Thermochemical Properties.
[5]Oetting, Chemical Thermodynamic Properties.

The free energies from Tables 5 and 6 were used to calculate the free energy for Eqs. 6-11. Results are shown in Table 7.

$$6Ta(\text{in T-111 alloy})(s) + 5Cm_2O_3(s) \rightarrow 3Ta_2O_5(s) + 10Cm(s) \quad (6)$$

$$3W(\text{in T-111 alloy})(s) + 2Cm_2O_3(s) \rightarrow 3WO_2(s) + 4Cm(s) \quad (7)$$

$$3Hf(\text{in T-111 alloy})(s) + 2Cm_2O_3(s) \rightarrow 3HfO_2(s) + 4Cm(s) \quad (8)$$

$$2Ta(\text{in T-111 alloy})(s) + 10PuO_2(s) \rightarrow Ta_2O_5(s) + 5Pu_2O_3(s) \quad (9)$$

$$W(\text{in T-111 alloy})(s) + 4PuO_2(s) \rightarrow WO_2(s) + 2Pu_2O_3(s) \quad (10)$$

$$Hf(\text{in T-111 alloy})(s) + 4PuO_2(s) \rightarrow HfO_2(s) + 2Pu_2O_3(s) \quad (11)$$

Table 7. Gibbs Free Energy of Reaction for Eqs. 6-11

Temperature (°C)	ΔG_r (cal/reaction)					
	Eq. 6	Eq. 7	Eq. 8	Eq. 9	Eq. 10	Eq. 11
600	574,610	412,570	39,400	-480	60,720	-63,670
800	573,470	411,640	43,550	-530	60,540	-62,160
1000	573,500	410,760	47,490	-1,300	59,930	-61,160

Since the free energies for Eqs. 6-8 are positive, the T-111 alloy will not be expected to react with pure Cm_2O_3. However, since the curium product will contain PuO_2 probably in excess of 4%, the reaction of PuO_2 with T-111 was considered. Since the free energies for Eqs. 9 and 11 are negative, the PuO_2 in the curium product would be expected to react with the tantalum and hafnium of the T-111 alloy.

In conclusion, it may be stated that a reaction between the tantalum and hafnium of the T-111 alloy and the PuO_2 would be expected. Experimental verification of the calculations is needed.

Reaction of Cm_2O_3 With Hafnalloy 20-20 *(E. E. Ketchen)*

The Hafnalloy 20-20 (95.78 atom % Hf, 3.71 atom % Sn, 0.33 atom % Pd, and 0.18 atom % Pt) is one of the materials under consideration as a capsule material to contain $^{244}Cm_2O_3$ heat source. Will the Cm_2O_3 source material react with this alloy at 600-1000°C?

The partial molar free energies of mixing for Hf, Sn, Pd, and Pt were calculated using the method reported in the Section, "Reaction of Cm_2O_3 With Pt–26% Rh–8% W Alloy." The results are shown in Table 8 from 600-1000°C. The method of calculation of the partial molar free energies assumes that no compound formation is present in the alloy mixture.

Table 8. Partial Molar Free Energies of Mixing of Components in Hafnalloy 20-20

Temperature (°C)	$-\Delta\overline{G}$ (cal/mole)			
	Hf	Sn	Pd	Pt
600	18	4495	9,997	11,109
800	35	5816	12,118	13,331
1000	52	7110	14,542	16,109

The free energies of formation of HfO_2, SnO_2, PdO, Pt_3O_4, Cm_2O_3, PuO_2, and Pu_2O_3 were calculated from a data compilation by Glassner[6] and a review article by Oetting.[7] The results are shown in Table 9. Since the free energies of formation of HfO_2 and SnO_2 are negative, they should react with oxygen over the 600-1000°C temperature range.

Table 9. Gibbs Free Energy of Formation of HfO_2, SnO_2, PdO, Pt_3O_4, Cm_2O_3, PuO_2, and Pu_2O_3

Temperature (°C)	ΔG_f (cal/mole)						
	HfO_2	SnO_2	PdO	Pt_3O_4	Cm_2O_3[a]	PuO_2	Pu_2O_3
600	-225,450	-95,730	-52	10,300	-352,120	-215,550	-352,120
800	-216,450	-85,830	4,520	26,400	-339,320	-207,090	-339,320
1000	-207,770	-76,080	8,780	42,080	-326,900	-198,700	-326,900

[a]Pu_2O_3 data used as stand-in for Cm_2O_3.

The free energies from Tables 8 and 9 were used to calculate the free energies for Eqs. 12-17. Results are shown in Table 10.

$$3Hf(\text{in Hafnalloy 20-20})(s) + 2Cm_2O_3(s) \rightarrow 3HfO_2(s) + 4Cm(s) \quad (12)$$

$$Hf(\text{in Hafnalloy 20-20})(s) + 4PuO_2(s) \rightarrow HfO_2(s) + 2Pu_2O_3(s) \quad (13)$$

$$3Sn(\text{in Hafnalloy 20-20})(s) + 2Cm_2O_3(s) \rightarrow 3SnO_2(s) + 4Cm(s) \quad (14)$$

$$Sn(\text{in Hafnalloy 20-20})(s) + 4PuO_2(s) \rightarrow SnO_2(s) + 2Pu_2O_3(s) \quad (15)$$

$$3Pd(\text{in Hafnalloy 20-20})(s) + Cm_2O_3(s) \rightarrow 3PdO(s) + 2Cm(s) \quad (16)$$

$$Pd(\text{in Hafnalloy 20-20})(s) + 2PuO_2(s) \rightarrow PdO(s) + Pu_2O_3(s) \quad (17)$$

[6]Glassner, Thermochemical Properties.
[7]Oetting, Chemical Thermodynamic Properties.

Table 10. Gibbs Free Energy of Reaction for Eqs. 12-17

Temperature (°C)	ΔG_r (cal/reaction)					
	Eq. 12	Eq. 13	Eq. 14	Eq. 15	Eq. 16	Eq. 17
600	27,940	-67,490	430,540	66,710	382,000	88,920
800	29,400	-66,880	438,600	69,530	389,230	91,410
1000	30,650	-66,780	446,890	71,970	396,870	93,790

Since the free energies for Eq. 13 are negative, the hafnium in Hafnalloy 20-20 will be expected to react with PuO_2 in the 600-1000°C temperature range. Experimental verification of the calculations is needed.

$^{244}Cm_2O_3$ Property Characterization

Helium Release *(P. Angelini)*

The final vacuum checks have been completed on the furnaces and headers. The three molybdenum sample tubes have been outgassed to 1900°C.

Vapor Pressure *(P. Angelini)*

The high-temperature vacuum furnace has been tested at temperatures up to 1900°C. The new beam chopper has been received and is being installed in the mass spectrometer furnace.

X-Ray Diffraction *(P. Angelini)*

The written procedure and description of the x-ray diffraction glove box system have been sent to the Safety Committee. The system has been approved for radioactive operation.

A new entry glove box to the process glove box has been fabricated and will be installed next month.

The final alignment has been performed with the furnace hot stage in place. Silicon powder specimens were used in this procedure.

The equipment and supplies needed for the first planned experiment have been gathered and placed in the process glove box. The first pellet used in solubility experiments is scheduled to be transferred.

Emissivity *(P. Angelini)*

The testing of the spectroradiometer has begun. Minor vacuum leaks have been located and are being removed.

Heat Capacity *(P. Angelini)*

The experimental equipment is ready for operation upon receipt of a pellet prepared from $^{244}Cm_2O_3$ freshly separated from $^{244}PuO_2$.

Dimensional Stability *(T. A. Butler and C. L. Ottinger)*

It has been decided to begin these experiments using $^{244}Cm_2O_3$ recently purified in the ORNL TRU facility. The material will be available next month.

Rate of Solution of Cm_2O_3 in Sea Water *(J. C. Posey)*

A test of the rate of loss of ^{244}Cm from the surface of a pellet of Cm_2O_3 in sea water is in progress. In this test a steady stream of air-saturated sea water passes over a hot-pressed $^{244}Cm_2O_3$ pellet. The stream of water is sampled periodically and analyzed for ^{244}Cm by an alpha counting procedure. The rate of solution is calculated from the concentration, the flow rate, and the surface area of the pellet. (The experimental procedure is essentially the same as that used in the distilled water experiment.[8])

The test has continued for more than 800 hr without visible disintegration of the pellet. Chemical reaction without an obvious physical change, however, is possible. The water flow was interrupted twice for periods of several hours because of equipment failure. The water flow interruption had no observable effect on the pellet.

The rate data are shown in Fig. 1. The first rate, 647 $\mu g/cm^2 \cdot hr$ at 0.12 hr, was much higher than the subsequent rates and is far off scale on Fig. 1. It indicates loose particulate material on the surface of the pellets. The same phenomon was observed in the test using air-saturated distilled water.

The rates calculated from the concentrations of filtered samples average lower than those calculated from the concentrations of samples that were not filtered. This indicates the presence of particulate material and suggests that material leaves the surface by a mechanism other than simple solution.

After about 360 hr, a gelatinous rust-red material was observed in the system. This material forms in the line before the water contacts the Cm_2O_3. It is believed to be largely ferric hydroxide caused by the corrosion of stainless steel lines by the sea water.

The scatter of the data increased at about the same time the corrosion material appeared. This scatter may be caused by the adsorption of curium by particles of ferric hydroxide. Since the curium concentrations are in the order of only 0.01 ppm, very little adsorption is required to

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[8]E. Lamb, *Isotopic Power Fuels Monthly Status Report for May 1973*, ORNL-TM-4282, Oak Ridge National Laboratory, pp. 6-9.

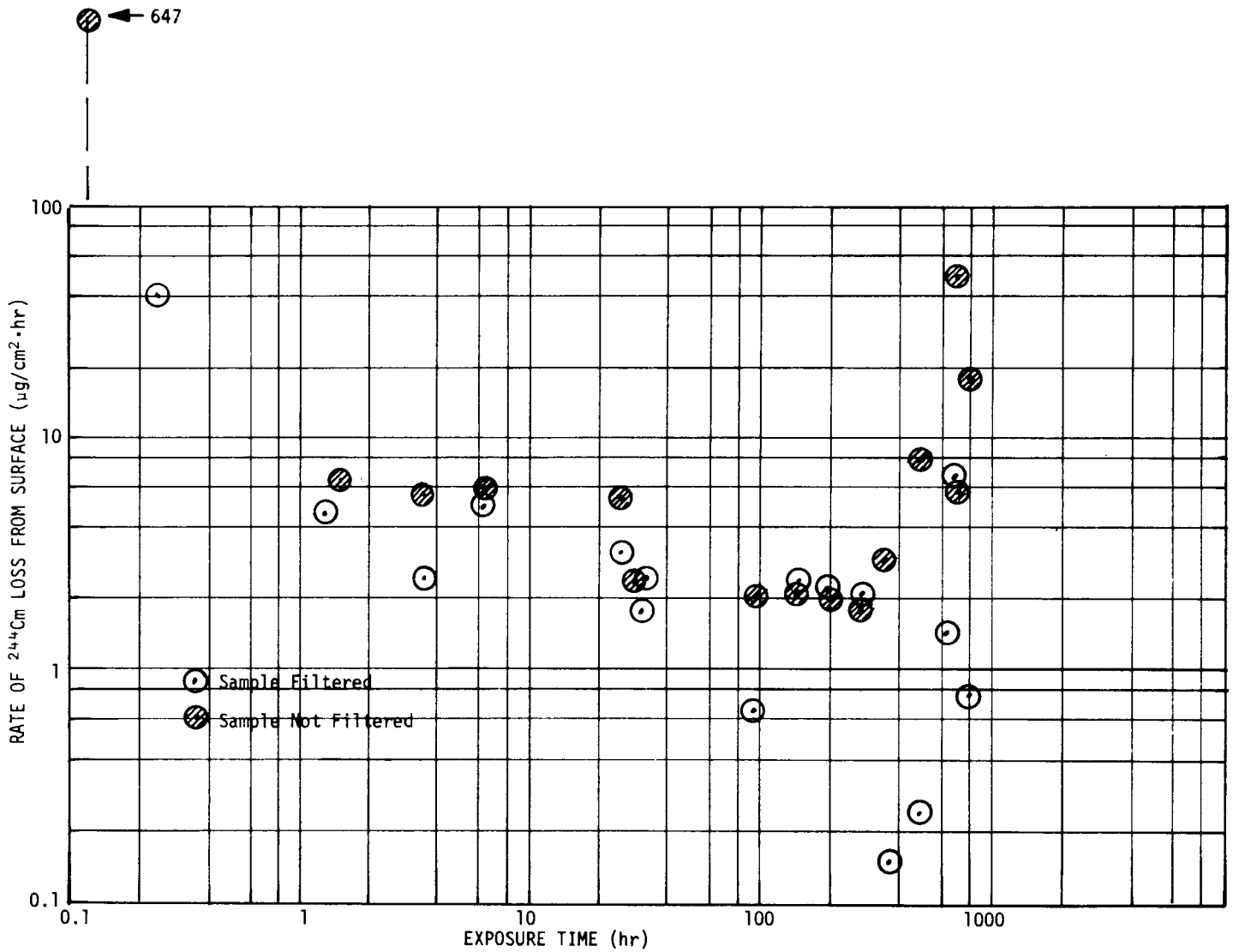


Fig. 1. Rate of Loss of ^{244}Cm from Surface of Pellet in Sea Water.

have a large relative effect. The sample would give high or low results depending on the presence or absence of adsorbing particles.

The test will be continued until the pellet disintegrates according to current plans.

Impact Testing of Heat Source Materials *(D. W. Ramey)*

Since the impact testing of curium fuel has been postponed for a period of 1 year, provisions are being made to store all equipment for the project. In addition to preparing various pieces of equipment for storage, a safety analysis was written for the operation of the impact gun in Room 12, Building 3026-C. This analysis will be subject to the review and approval of the Radiation Safety Review Committee.

$^{244}Cm_2O_3$ Radiation Measurements *(K. W. Haff)*

The radiation measurements of the 61.3 W_t $^{244}Cm_2O_3$ source were completed, and the data are presently in the computer being analyzed. The analyzed data are expected to be completed in early August.

Assay of SRL-31 Curium-244 Oxide *(T. A. Butler)*

Curium-244 oxide batch SRL-31 was assayed to provide additional $^{244}Cm_2O_3$ for characterization experiments. Analyses were completed with the exception of the determination of specific power pending a recalibration of the calorimeter.

The isotopic abundances of the two principal elements, curium and plutonium, are given in Table 11.

Table 11. Isotopic Abundances of Curium and Plutonium in SRL-31 Oxide

Curium		Plutonium	
Mass No.	Isotopic Abundance (atom %)	Mass No.	Isotopic Abundance (atom %)
244	94.26	238	0.091
245	0.880	239	0.028
246	4.74	240	99.86
247	0.077	241	0.002
248	0.042	242	0.022
		244	≤ 0.004[a]

[a]Possible interference from ^{244}Cm.

The curium and plutonium content of SRL-31 was determined by radiochemical analysis for ^{244}Cm and ^{240}Pu, and the total element weight was calculated by use of the isotopic abundances given in Table 11. The results of the assay are shown in Table 12.

Table 12. Curium and Plutonium Content in SRL-17 Oxide

Sample No.	Weight (mg)	Curium (mg/g)	Plutonium (mg/g)	Cm_2O_3 (mg/g)	PuO_2 (mg/g)
Cm-31-4C	19.3	753	78	827	88
Cm-31-5C	20.0	807	70	888	79
Cm-31-6C	13.9	839	73	922	83
Average		800	74	879	84
Adjusted average[a]					

[a]Calorimetric value of specific power (W ^{244}Cm per g) not available.

The radioactive impurities were determined by standard germanium crystal gamma spectrometry and are listed in Table 13.

Table 13. Radioactive Impurities in SRL-31 Oxide

Nuclide	Curies/g
Ruthenium-103	$\leq 9 \times 10^{-6}$
Ruthenium-106	$\leq 3 \times 10^{-5}$
Cesium-134	$\leq 3 \times 10^{-6}$
Cesium-137	$\leq 3 \times 10^{-6}$
Cerium-144	$\leq 4 \times 10^{-4}$
Praseodymium-144	$\leq 4 \times 10^{-4}$
Zirconium-95	$\leq 1 \times 10^{-5}$
Niobium-95	$\leq 4 \times 10^{-6}$
Europium-154	$\leq 1 \times 10^{-5}$
Americium-243	9×10^{-3}

The non-radioactive impurities were determined by spark source mass spectrometry and are listed in Table 14. The weight of these impurities in the form of oxides is ∿15 mg per gram of sample. The principal individual impurities are 10.9 mg of AmO_2 and 1.8 mg of Fe_2O_3.

Table 14. Impurities in Batch SRL-31 Curium Oxide

Element	Wt (ppm of Cm)	Element	Wt (ppm of Cm)
Aluminum	20	Molybdenum	200
Arsenic	<10	Sodium	200
Boron	<10	Nickel	100
Barium	<10	Lead	100
Calcium	20	Platinum	100
Cobalt	<10	Silicon	100
Chromium	100	Tantalum	150
Copper	30	Titanium	<100
Iron	1,500	Zinc	<20
Potassium	10	Zirconium	<500
Magnesium	<10	Americium	12,000
Manganese	30	Rare Earths	300

In summary, the composition of SRL-31 oxide is:

Material	mg/g	Percent
Cm_2O_3	879	87.9
PuO_2	84	8.4
Impurities	∿15	1.5
Unknown	22[a]	2.2[a]

[a]Adjustment of the curium and plutonium assays when calorimetric results are available may change this value.

Curium Oxide Purification *(T. A. Butler and C. L. Ottinger)*

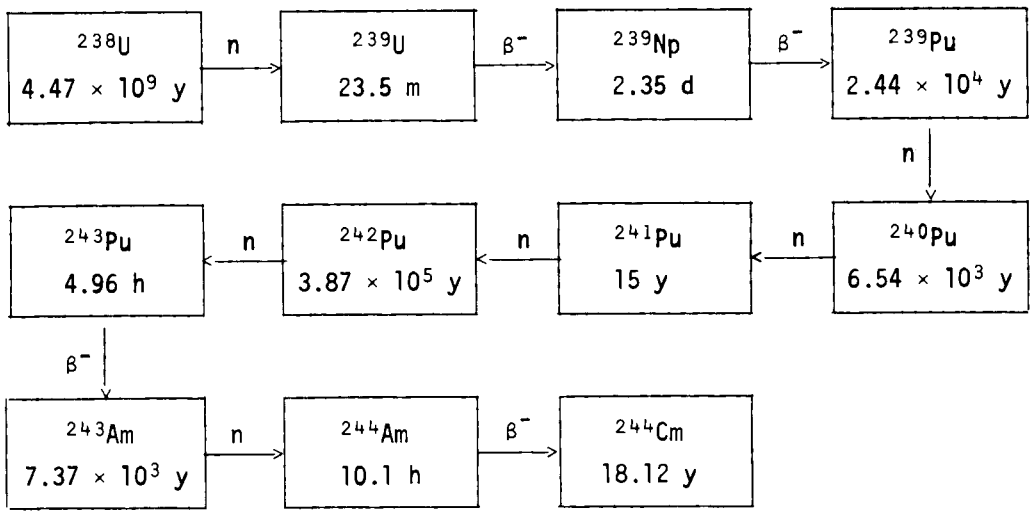
An additional fraction of ^{244}Cm residues containing about 50 g of ^{244}Cm resulting from past experimental work was transferred to the TRU facility for reprocessing. The total estimated ^{244}Cm content in the two fractions was ∿100 g. This material is being processed at TRU to remove the plutonium; then the purified ^{244}Cm fraction will be returned to the CSFF for experimental work. Processing of the first fraction is essentially complete. The starting material contained 46.5 g of ^{244}Cm, and the yield of purified material was 40.4 g; the remaining material will be retained in the TRU inventory.

Curium-244 Yield Calculations *(T. A. Butler)*

The future source of ^{244}Cm in quantity is expected to derive from the reprocessing of power reactor fuels. A study was begun to examine some of the factors which influence the amount of ^{244}Cm produced in the reactor fuels and to estimate future quantities available from this source for isotopic power devices. In this study, the considerable assistance of

C. W. Kee, Engineering Coordination and Analysis Section, ORNL Chemical Technology Division, and of F. T. Binford, Development Department, ORNL Operations Division, is gratefully acknowledged. Computer calculations and interpretation of the data are supplied by C. W. Kee.

The more important nuclear reactions which lead to the formation of ^{244}Cm in uranium-fueled reactors are as follows:



In a previous report[9] the yield of ^{244}Cm from reference PWR reactors was given as a function of burnup (MWD/MT) for both enriched ^{235}U and plutonium recycle fueled reactors for specified operating conditions. A large increase in ^{244}Cm yield from plutonium recycle fuel is evident, but significant quantities of these fuels are not expected to be available for a decade or so. Therefore, the present study will be concerned with PWR and BWR reactors fueled with uranium containing low enrichments of ^{235}U.

The production rate of ^{244}Cm in enriched uranium fuels is dependent on the total fluence (neutron flux × time) rather than on burnup alone. The neutron flux required to maintain constant reactor power levels is greater for less enriched fuels while the amount of ^{238}U present is about the same for low-enriched fuels. The rate of neutron captures leading eventually to ^{244}Cm is therefore increased. To demonstrate this effect, ORIGEN[10] calculations were made for PWR reference reactors operating at a power of 30 MW/MT and fueled with 1.5, 2.0, 2.5, 2.8, and 3.3% ^{235}U enrichments.

In these calculations, it was assumed that the neutron energy spectrum is the same in all cases. This is approximately true since most of the fissions are from ^{235}U. Small differences occur because of the buildup

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[9] E. Lamb, ORNL-TM-4282, p. 10

[10] M. J. Bell, *ORIGEN — The ORNL Isotope Generation and Depletion Code*, ORNL-4628, Oak Ridge National Laboratory (May 1973).

of a varying amount of plutonium during the life of the reactor fuel; however, this should not introduce large errors in the calculated yields of ^{244}Cm. The results of the calculations are given in Tables 15 and 16 and Fig. 2. Figure 2 is a graphic display of values given in Table 16.

Table 15. Average Neutron Flux as a Function of Fuel Enrichment

Reactor Power: 30 MW/MT
Fuel Burnup: 33 GWD/MT

^{235}U Enrichment (%)	Average Neutron Flux ($n/cm^2 \cdot sec$)
1.5	4.25×10^{13}
2.0	3.83×10^{13}
2.5	3.45×10^{13}
2.8	3.24×10^{13}
3.3	2.92×10^{13}

Table 16. Influence of ^{235}U Enrichment on ^{244}Cm Yield in Power Reactor Fuel

Burnup [GWD/MT(U)]	^{244}Cm Yield [g/MT(U)]				
	^{235}U Enrichment (%)				
	1.5	2.0	2.5	2.8	3.3
3.3	0.004	0.001	0.0003	0.0002	0.00008
6.6	0.11	0.038	0.015	0.009	0.004
9.9	0.75	0.31	0.14	0.09	0.04
13.2	2.9	1.3	0.63	0.42	0.22
16.5	7.8	3.9	2.0	1.4	0.74
19.8	17.3	9.3	5.1	3.6	2.0
23.1	32.9	19.1	11.1	8.0	4.7
26.4	56.9	35.0	21.2	15.7	9.5
29.7	89.8	58.1	37.1	28.1	17.6
33.0	133.0	89.8	59.8	46.4	30.3

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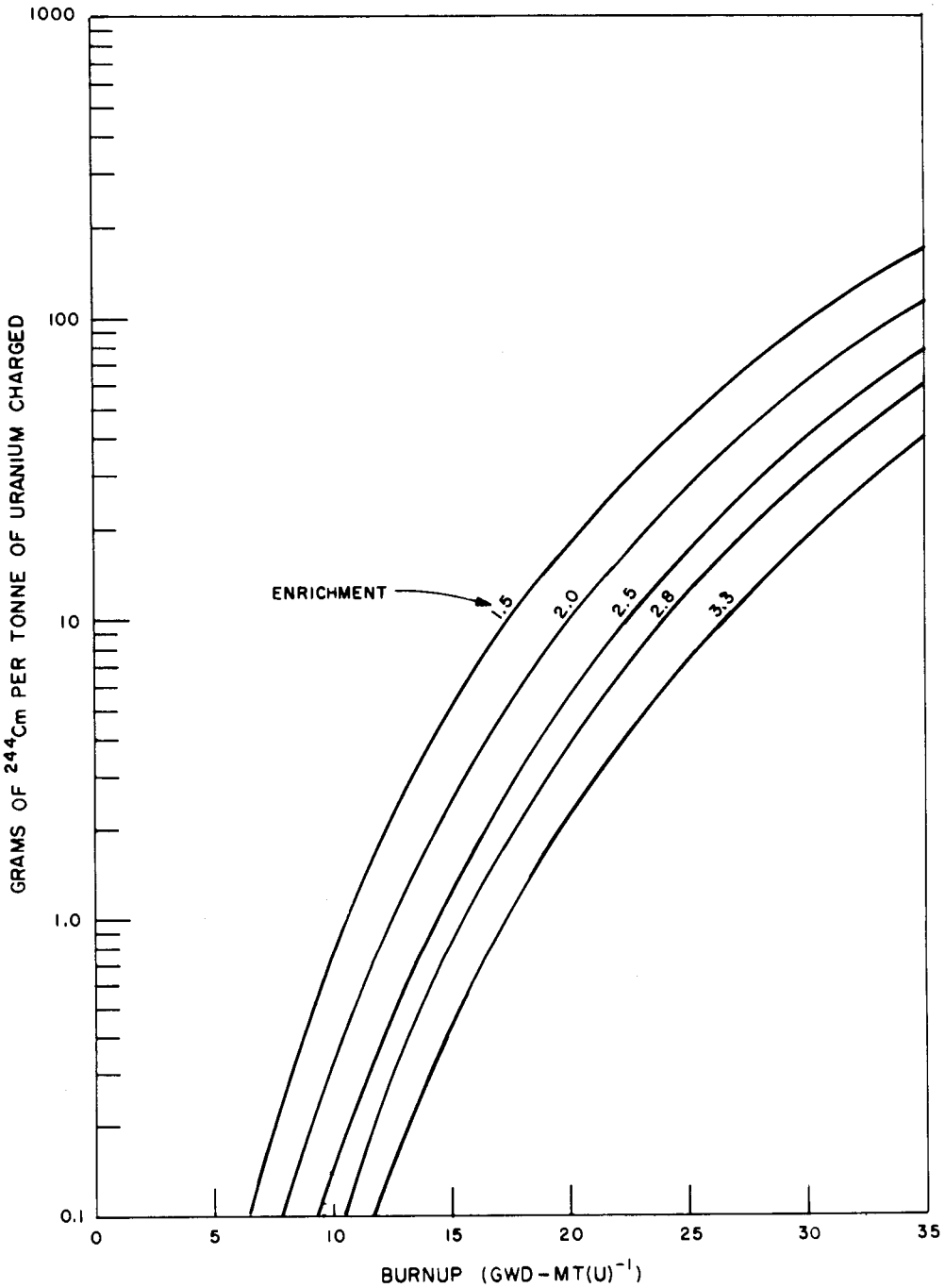

Fig. 2. Influence of Enrichment on Curium Production.

Curium Source Fabrication Facility Operation

(C. L. Ottinger)

Some equipment repairs were made and a considerable amount of solid waste disposal was done during July. Experiments with the purified ^{244}Cm should begin early in August.

Transfers of two batches of residues to TRU facility for processing and removal of ^{240}Pu was completed.

APPENDIX

Curium-244 Inventory

	^{244}Cm Status (g)		
	7-1-73	Change	8-1-73
Receipts from SRL			
Allocated to LR Heat Source Program	1,902		1,902
Allocated to NT Target Program	302		302
Total Receipts from SRL	2,204		2,204
Withdrawals from LR Program			
Analytical samples	2		2
Shipped to Lovelace Foundation	9		9
Shipped to SRL	2		2
Discarded to ORNL waste	6		6
Unaccounted for	80		80
Total Withdrawals from LR Program	99		99
Balance Remaining in LR Program			
Stored residues for recovery	322		322[a]
In-use material	146		146
Stored raw material	1,335		1,335
Total Balance in LR Program	1,803		1,803
Withdrawals from NT Program			
Transferred to TRU	146		146
Shipped to other sites	44	1	45
Transferred to Target Group	5		5
Total Withdrawals from NT Program	195	1	196
Balance Remaining in NT Program	107	-1	106

[a]Approximately 100 g was transferred to TRU for purification. The purified material will be returned to Isotopes Division, and the difference transferred to TRU inventory during August.

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