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ORNL ISOTOPIC POWER FUELS  
QUARTERLY REPORT  
FOR PERIOD ENDING JUNE 30, 1973

Eugene Lamb

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ISOTOPES DEVELOPMENT CENTER

ORNL ISOTOPIC POWER FUELS QUARTERLY REPORT  
FOR PERIOD ENDING JUNE 30, 1973

Eugene Lamb  
Isotopes Division

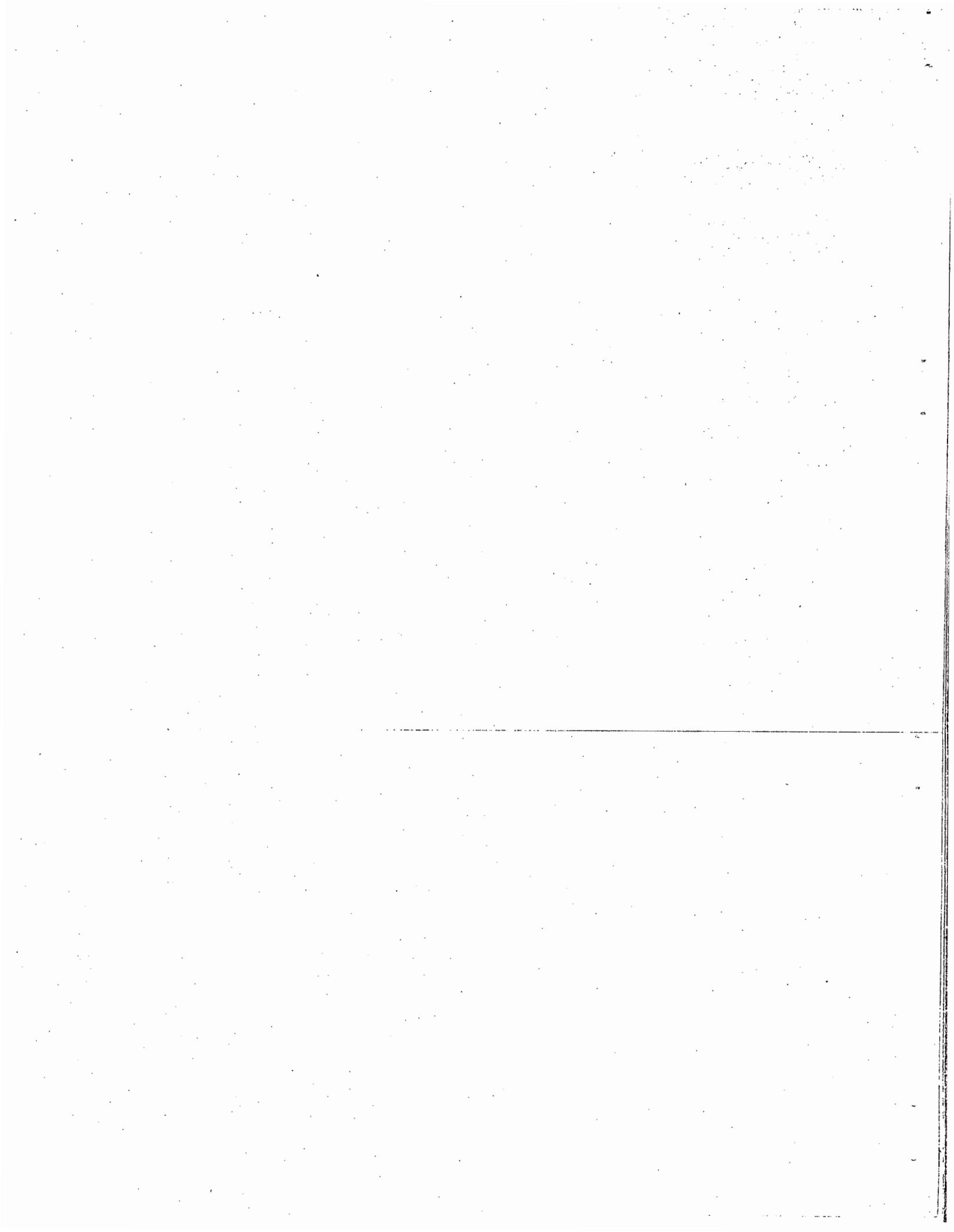
SEPTEMBER 1973

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ORNL ISOTOPIC POWER FUELS QUARTERLY REPORT  
FOR PERIOD ENDING JUNE 30, 1973

*Eugene Lamb*

SUMMARY

Compatibility tests of  $^{244}\text{Cm}_2\text{O}_3$  with 18 materials in 67 couples began and will run at 900, 1100, 1400°C for periods of 2500 and 5000 hr. A thorough analysis of the fuel used in the compatibility couples was completed. Short-term compatibility tests of  $^{244}\text{Cm}_2\text{O}_3$  with  $\text{ThO}_2$ ,  $\text{ZrO}_2$ , and  $\text{Al}_2\text{O}_3$  are in progress.

Free energy calculations for the reaction of  $^{244}\text{Cm}_2\text{O}_3$  with C, Be,  $\text{ZrO}_2$ , and  $\text{Al}_2\text{O}_3$  were made.

Equipment for  $^{244}\text{Cm}_2\text{O}_3$  property characterization (helium release, vapor pressure, x-ray diffraction, emissivity, heat capacity, dimensional stability) is being prepared and checked for operability. The measurements of solution rates of  $^{244}\text{Cm}_2\text{O}_3$  in distilled water were completed. A comparison of vapor pressures of curium, plutonium, and americium oxides was made based on literature values.

Modifications to the impact gun for hot cell operation are in progress.

Neutron and gamma radiation measurements were made on a 61.3-W<sub>t</sub>  $^{244}\text{Cm}_2\text{O}_3$  source.

The  $^{240}\text{Pu}$  decay product will be removed from a 50-g batch of  $^{244}\text{Cm}_2\text{O}_3$  to provide material for baseline characterization studies.

Pellets for characterization and compatibility experiments were prepared in the Curium Source Fabrication Facility (SFF); variations in pellet quality were noted. Tamped powder densities in the range of 5.5 to 6.0 g/cm<sup>3</sup> were measured. The self-sintering behavior of  $^{244}\text{Cm}_2\text{O}_3$  powder in layers of different thicknesses was noted; up to 0.5-cm-deep layers formed only light crusts, but a 2-cm-diam by 2-cm-deep body of powder formed into a hard mass.

## CURIUM-244 FUEL DEVELOPMENT

(Division of Space Nuclear Systems Program 04 30 05 03)

 $^{244}\text{Cm}_2\text{O}_3$  Compatibility ProgramCharacterization of  $^{244}\text{Cm}_2\text{O}_3$  for Compatibility Tests*(T. A. Butler and J. R. DiStefano)*

Examination of a hot-pressed pellet typical of those prepared for use in the current 900, 1100, and 1400°C compatibility test matrix was completed. The bulk density of the pellet was calculated from averaged measurements of the diameter, thickness, and weight. The data are:

Thickness (in.)	Diameter (in.)	Weight (g)	Volume (cm <sup>3</sup> )	Density (g/cm <sup>3</sup> )
0.0796	0.1783	0.2915	0.03257	8.9502
0.0797	0.1782	0.2915	0.03257	8.9490
0.0798	0.1784	0.2915	0.03269	8.9178
0.0797	0.1784	0.2915	0.03265	8.9289
Av 0.0797	0.1783	0.2915	0.03262	8.9365

The percent of theoretical density is 76.4 based on a maximum value of 11.7 g/cm<sup>3</sup> for B-type  $\text{Cm}_2\text{O}_3$  calculated from x-ray diffraction data.

The top surface temperature of the pellet when resting on a thermally insulated surface was measured by a thermocouple to be 230°F (110°C).

The specific power was determined by calorimetry to be 2.06 W/g; thus the power and power density of the pellet was 0.6 W and 18.4 W/cm<sup>3</sup>, respectively.

Optical photomicroscopy of a cross section of the pellet indicated areas of variable density and considerable void volume which would be anticipated from the density measurement.

Characterization by x-ray diffraction did not show peaks that could be identified with any of the curium or plutonium oxides. Electron-beam microprobe analysis of the polished surface of the pellet indicated only Al, Fe, and Ca other than curium and does not appear to be a useful characterization method for this purpose.

The composition of batch SRL-18  $^{244}\text{Cm}$  oxide powder used in the 900, 1100, and 1400°C compatibility test matrix was determined. The  $^{244}\text{Cm}$  and  $^{240}\text{Pu}$  contents were determined by radiochemical analysis and the elemental impurities by spark source mass spectrometry. The radiochemical results are shown in Table 1.

Table 1. Radiochemical Analysis of Batch SRL-18 Curium Oxide

Sample No.	Weight (mg)	$^{244}\text{Cm}$ (mg/g)	$^{240}\text{Pu}$ (mg/g)	Total Cm <sup>a</sup> (mg/g)	Total Pu <sup>a</sup> (mg/g)
Cm18-2	13.57	638	99.1	676.4	99.1
Cm18-3	10.66	626	99.5	663.7	99.5
Cm18-4	15.88	628	102.4	665.8	102.4
Average		630.7	100.3	668.6	100.3
Adjusted Average <sup>b</sup>		738	115.8	771.8	115.8

<sup>a</sup>Based on 94.32% abundance of  $^{244}\text{Cm}$  and >99% abundance of  $^{240}\text{Pu}$ .

<sup>b</sup>Based on assumption of incomplete sample transfer (see text).

Conversion of the total curium and plutonium amounts to weights of  $\text{Cm}_2\text{O}_3$  and  $\text{PuO}_2$  yields 734.4 mg/g and 113.7 mg/g, respectively (848.1 mg/g total). However, the radiochemical assay for  $^{244}\text{Cm}$  is inconsistent with a calorimetrically measured value of 2.06 W/g which is equivalent to 728 mg of  $^{244}\text{Cm}$  based on 2.83 W/g for 100%  $^{244}\text{Cm}$  isotope. A review of the sampling and radiochemical procedures did not reveal any identifiable systematic errors; therefore, it was concluded that the powder samples were incompletely recovered from the transfer containers and resulted in the low radiochemical assays. An adjustment factor  $728/630.7 = 1.154$  was applied to the reported  $^{244}\text{Cm}$  and  $^{240}\text{Pu}$  assay values. The adjusted values for the weights of  $\text{Cm}_2\text{O}_3$  and  $\text{PuO}_2$  are 847.8 mg/g and 131.2 mg/g, respectively (979 mg/g total).

Five determinations of contaminants in batch SRL-18 curium oxide were made at ORNL and one by the Savannah River Laboratory which was supplied with the batch shipment. From these assays, the values shown in Table 2 were taken as representative of the oxide used in the pellet fabrication.

Conversion of the principal (>10 ppm) impurities to fully oxidized forms yields 12.2 mg total weight per gram of curium oxide.

The composition of the  $^{244}\text{Cm}$  oxide used in the compatibility couples can be summarized as follows:

	<u>mg/g</u>	<u>Percent</u>
$\text{Cm}_2\text{O}_3$	847.8	84.8
$\text{PuO}_2$	131.2	13.1
Impurities	12.2	1.2
Unknown	8.8	0.9

Table 2. Impurities in Batch SRL-18 Curium Oxide

Element	Wt (ppm of Cm)	Element	Wt (ppm of Cm)
Aluminum	15	Lead	20
Arsenic	<10	Platinum	1000
Boron	<5	Silicon	300
Barium	<10	Strontium	<10
Calcium	130	Tantalum	100
Niobium	<10	Titanium	<15
Cadmium	<10	Tungsten	400
Cobalt	<5	Zinc	100
Chromium	400	Zirconium	400
Copper	100	Americium	7500
Iron	2000	Yttrium	30
Iridium	100	Bromine	<10
Potassium	<5	Chlorine	20
Magnesium	<10	Fluorine	10
Manganese	15	Sulfur	15
Molybdenum	100	Lanthanum	15
Sodium	<10	Cerium	30
Nickel	150	Other rare earths	100

Compatibility Couple Fabrication (*T. A. Butler, J. R. DiStefano, and C. L. Ottinger*)

Previous work on the fabrication of couples for compatibility testing at 900, 1100, and 1400°C was reported previously.<sup>1</sup> Test conditions for these couples are summarized in Table 3. Details of the procedure used in fabricating the couples are given below.

The Cm<sub>2</sub>O<sub>3</sub> pellets were prepared by first calcining CmO<sub>2</sub> at 1000°C in vacuum for 4 hr, loading the graphite dies in argon, hot-pressing at 1450°C in vacuum, and ejecting the pellets in an argon atmosphere. The completed pellets were then heated to 1000°C in argon and held at this temperature while the oxygen content in the argon was increased to ~2000 ppm. These conditions were maintained 4 hr to oxidize any residual carbon from the pellet surfaces. Then the oxygen content was reduced to <100 ppm and the pellets cooled to cell temperature. The pellet ends were polished on a diamond-impregnated steel plate; then the pellets were stored (in argon or vacuum) prior to encapsulation.

Loading of the inner capsule components was carried out in a hot cell under argon atmosphere (<150 ppm oxygen). After welding, the inner capsules were transferred to an adjacent cell (argon atmosphere) where they were enclosed in a secondary capsule (Hastelloy X or tantalum). Those couples to be tested in dynamic vacuum were sealed in the secondary capsule under argon to allow transfer and handling of the containers under sealed conditions prior to testing. The other couples which were to be

run under a static helium atmosphere were inserted in secondary capsules having an open 1/4-in.-OD tube protruding from the top end plug (nickel from Hastelloy X and tantalum from the tantalum end plug). This tube allowed the container to be evacuated and back-filled with helium. With the helium line still attached, the protruding tube was first crimped closed and then pinched off as near the end as possible. (If the tube were not completely leak-tight after the pinch-off, there would be but little exchange prior to seal welding between helium at 1 atm inside the tube and argon at 1 atm on the outside.) The pinched end of the tube was then tungsten arc welded. Sealing of these secondary capsules with an internal helium pressure of 1 atm provided positive assurance that the surrounding atmosphere was helium.

The ten secondary capsules containing the 67 compatibility couples were visually examined and photographed through a Kollmorgan periscope. The appearance of the welds on the secondary capsules was satisfactory, with

Table 3. Test Conditions for  $^{244}\text{Cm}_2\text{O}_3$  Compatibility Couples<sup>a</sup>

Material <sup>b</sup>	Conditions		
	Static Helium (5000 hr at 900°C)	Static Helium and Dynamic Vacuum (2500 hr at 1100°C)	Static Helium and Dynamic Vacuum (5000 hr at 1400°C)
Iridium	✓	✓	✓
Graphite	✓	✓	✓
Platinum	✓	✓	
Hf-1% Pt-0.5% Pd	✓		
Hastelloy C-276	✓		
Haynes alloy No. 25	✓		
Haynes alloy No. 188	✓		
ThO <sub>2</sub>	✓		
Pt-20% Rh	✓		
Pt-26% Rh-8% W	✓		
Pt-26% Rh-8% W-0.5% Ti		✓	
Pt <sub>3</sub> Ir		✓	
Molybdenum			✓
Mo-46% Re			✓
Tantalum			✓
T-111			✓
Tungsten			✓
W-26% Re			✓

<sup>a</sup>All couples except graphite to be tested both singly and in graphite intermediate capsule surrounding inner capsule.

<sup>b</sup>Inner capsule and disk specimen.

some areas of roll-over and indentations noted. A helium leak test of the individual secondary capsules in an evacuated chamber indicated that all were sealed. The secondary capsules for the 1100°C tests were transferred to argon-atmosphere glove boxes attached to Brew furnaces, and the secondary capsules containing the couples for exposure to dynamic vacuum were penetrated.

In addition to the test couples, control capsule tests under similar time-temperature-atmosphere conditions are being conducted as summarized in Table 4.

Table 4. Control Tests for  $^{244}\text{Cm}_2\text{O}_3$  Compatibility Couples<sup>a</sup>

Material	Conditions				
	5000 hr at 900°C Static Helium	2500 hr at 1100°C		5000 hr at 1400°C	
		Helium	Vacuum	Helium	Vacuum
Iridium	✓		✓	✓	✓
Graphite	✓ <sup>b</sup>	✓	✓	✓	✓
Platinum	✓	✓	✓		
Hf-1% Pt-0.5% Pd	✓ <sup>b</sup>				
Hastelloy C-276	✓ <sup>b</sup>				
Haynes alloy No. 25	✓ <sup>b</sup>				
Haynes alloy No. 188	✓ <sup>b</sup>				
ThO <sub>2</sub>	✓				
Pt-20% Rh	✓ <sup>b</sup>				
Pt-26% Rh-8% W	✓ <sup>b</sup>				
Pt-26% Rh-8% W-0.5% Ti		✓			
Pt <sub>3</sub> Ir		✓	✓		
Molybdenum				✓	✓
Mo-46% Re					✓
Tantalum				✓	✓
T-111				✓	✓
Tungsten				✓	✓
W-26% Re				✓	✓

<sup>a</sup>Inner capsules to be tested in graphite intermediate capsules except where noted.

<sup>b</sup>Couple also to be tested singly.

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

All compatibility couples in the 900, 1100, and 1400°C test matrix were completed and the heat soak period started. The status of the tests as of June 28, 1973, is summarized in Table 5. The column headed "thermal cycles" indicates the number of times furnaces lost power for various reasons.

Table 5. Status of Compatibility Tests

Set <sup>a</sup>	Start of Test	Thermal Cycles	Cumulative Hours	Projected End of Test
1	5-3-73	None	1344	11-28-73
2	5-3-73	None	1344	11-28-73
3	4-16-73	3	1697	8-4-73
4	4-16-73	3	1697	8-4-73
5	4-17-73	2	1574	8-9-73
6	4-17-73	2	1574	8-9-73
7	5-3-73	2	1231	12-3-73
8	5-3-73	2	1231	12-3-73
9	5-3-73	2	1224	12-3-73
10	5-3-73	2	1224	12-3-73

<sup>a</sup>Set identifications:

- 900°C; 5000 hr; helium atmosphere; Ir, C, Pt, Hf-1% Pt-0.5% Pd, Hastelloy C-276, Haynes 25, Haynes 188, ThO<sub>2</sub>, Pt-20% Rh, and Pt-26% Rh-8% W.
- 900°C; 5000 hr; graphite, helium atmosphere; Ir, Pt, Hf-1% Pt-0.5% Pd, Hastelloy C-276, Haynes 25, Haynes 188, ThO<sub>2</sub>, Pt-20% Rh, and Pt-26% Rh-8% W.
- 1100°C; 2500 hr; graphite, helium atmosphere; Ir, Pt, Pt<sub>3</sub>Ir, and Pt-26% Rh-8% W-0.5% Ti.
- 1100°C; 2500 hr; dynamic vacuum; Ir, Pt, Pt<sub>3</sub>Ir, C, and Pt-26% Rh-8% W-0.5% Ti.
- 1100°C; 2500 hr; helium atmosphere; Ir, Pt, Pt<sub>3</sub>Ir, C, and Pt-26% Rh-8% W-0.5% Ti.
- 1100°C; 2500 hr; graphite, dynamic vacuum; Ir, Pt, Pt<sub>3</sub>Ir, and Pt-26% Rh-8% W-0.5% Ti.
- 1400°C; 5000 hr; graphite, helium atmosphere; Ir, C, Mo, Mo-46% Re, Ta, T-111, W, and W-26% Re.
- 1400°C; 5000 hr; dynamic vacuum; Ir, Mo, Mo-46% Re, Ta, T-111, W, and W-26% Re.
- 1400°C; 5000 hr; helium atmosphere; Ir, Mo, Mo-46% Re, Ta, T-111, W, and W-26% Re.
- 1400°C; 5000 hr; graphite, dynamic vacuum; Ir, C, Mo, Mo-46% Re, Ta, T-111, W, and W-26% Re.

### Curium Prototype Test Facility (*K. W. Haff*)

Removal of the old barricades in preparation for installation of shielding and test equipment is complete, and the facility has been prepared for painting. Design drawings of the water shielding tanks are complete and fabrication of the tanks is ~50% complete. Installation of the concrete block shielding on the upper part of the enclosure is ~20% complete. Shielding work is expected to be completed by the end of July. Shipment of the surplus equipment which is to be used in these tests was delayed until mid-August. We expect to receive the equipment in late August.

### $\text{Cm}_2\text{O}_3$ Compatibility With Ceramic Materials (*P. Angelini*)

The equipment needed for this experiment was gathered and bagged into the glove box. The curium oxide pellets needed for the experiments have been fabricated and those not used have been placed in storage.

The present plan of experiments is presented in Table 6.

Table 6.  $^{244}\text{Cm}_2\text{O}_3$  Compatibility Experiments

Experiment	Material	Temperature (°C)	Time (hr)
A	$\text{ThO}_2$	1500	24
B	$\text{ThO}_2$	1300	100
C	$\text{ThO}_2$	1500	$t_3$
D	$\text{ThO}_2$	1300	$t_4$
E	$\text{ZrO}_2$	1500	24
F	$\text{ZrO}_2$	1300	100
G	$\text{ZrO}_2$	1500	$t_3$
H	$\text{ZrO}_2$	1300	$t_4$
I	$\text{Al}_2\text{O}_3$	1500	24

Experiments A and B (see Table 6) have been performed, and the couples have been transferred to HRLEL for examination. Experiments E and F are being readied. The design for fabricating the capsules and inserts for Experiment I has been given to the shops for fabrication. One of the as-fabricated  $^{244}\text{Cm}_2\text{O}_3$  pellets has been sent to HRLEL for optical microscopy study.

The compatibility couples will be examined using an electron-beam microprobe. This examination will yield data, such as concentration of  $^{244}\text{Cm}$  versus distance in the oxide to be tested. From that data the diffusion coefficient of curium in those oxides can be calculated. Optical microscopy will also be performed to define areas in the compatibility couple to be examined using the electron-beam microprobe.

Experiments C, D, G, and H will be performed after the preliminary analysis of Experiments A and B. Thus, different exposure times,  $t_3$  and  $t_4$ , at temperature may be chosen for the remaining couples in order to derive more information from the experiment.

### Free Energy Calculation for the Reaction of Carbon with $\text{Cm}_2\text{O}_3$ (E. E. Ketchen)

The Gibbs free energy was calculated for the reaction of carbon with  $\text{Cm}_2\text{O}_3$  to produce  $\text{CmO}$  between 1700 and 2400°K.

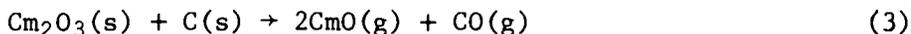
The free energy was calculated for Eq. 1 using Eq. 2 which had been developed by P. K. Smith and D. E. Peterson.<sup>2</sup> The authors have presented indirect evidence that  $\text{Cm}_2\text{O}_3$  will vaporize to  $\text{CmO}$  and oxygen as the main products. The formation of  $\text{CmO}$  as the major vaporization product needs to be verified by mass spectrometer measurements.



$$\Delta G = -RT \ln k = -2,303 RT (\log 0.335 + 3[7.32 - (29,050/T)]) \quad (2)$$

where

$k$  = equilibrium constant for equation,  
 $T$  = temperature in °K, and  
 $R$  = 1.9872 cal/deg·mole.



The free energy values calculated for Eq. 1 combined with data from the JANAF tables<sup>3</sup> were used to calculate the free energy of Eq. 3. The results are shown in Table 7. Since the free energies for the reaction are positive up to 2400°K (the highest temperature for which the calculations have been made), this reaction should not proceed unless the products of the reaction are removed. Since the free energy becomes more positive as the temperature is lowered, this reaction becomes less likely to proceed.

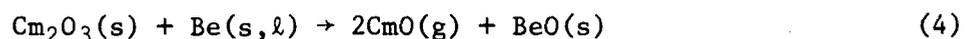
Table 7. Gibbs Free Energy as a Function of Temperature for Eq. 3

$\Delta G$ (kcal/mole)	Temperature (°K)
135.5	1700
125.3	1800
115.0	1900
104.8	2000
94.5	2100
84.3	2200
74.1	2300
63.9	2400

The free energy of other potential reactions of curium sesquioxide and carbon, such as those to form  $\text{CmC}_{0.5}\text{O}_{0.5}$ ,  $\text{CmC}_{0.9}$ ,  $\text{CmC}_{1.5}$ , and  $\text{CmC}_2$  will be investigated next. Since thermodynamic data are not available for these compounds, the data for the plutonium compounds may be used as reasonable substitutes for curium.

#### Free Energy Calculations for the Reaction of Beryllium with $\text{Cm}_2\text{O}_3$ (E. E. Ketchen)

The thermodynamics of the reaction of  $\text{Cm}_2\text{O}_3$  with beryllium to form  $\text{CmO}$  or curium metal have been investigated to evaluate the possibility of the reaction of  $\text{Cm}_2\text{O}_3$  with beryllium metal.

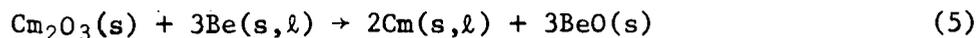


The free energy values calculated for Eq. 1 combined with data from the JANAF tables<sup>3</sup> and a compilation of data by Alvin Glassner<sup>4</sup> were used to calculate the free energy of Eq. 4. The results are shown in Table 8. No reaction would be expected in the 600-1800°C temperature range.

Table 8. Gibbs Free Energy as a Function of Temperature for Eq. 4

$\Delta G$ (kcal/mole)	Temperature (°C)
143.98	600
131.98	800
120.04	1000
108.17	1200
96.03	1400
84.10	1600
72.10	1800

The free energy was calculated for Eq. 5 using the free energy of formation of  $\text{Pu}_2\text{O}_3$  (ref. 5) as a stand-in for  $\text{Cm}_2\text{O}_3$  and the free energy of formation of  $\text{BeO}$  from a compilation of data by Alvin Glassner.<sup>4</sup> The free energy values are shown over a range of 600 to 1800°C in Table 9.



The Gibbs free energy values for Eq. 5 are negative over the range of temperatures considered, hence the reaction should occur. However, a protective coating of  $\text{BeO}$  or  $\text{BeO}\cdot\text{Cm}_2\text{O}_3$  may be formed over the metal which will inhibit the reaction.

Table 9. Gibbs Free Energy as a Function of Temperature for Eq. 5

$-\Delta G$ (kcal/mole)	Temperature ( $^{\circ}\text{C}$ )
14.89	600
13.65	800
12.21	1000
10.55	1200
8.74	1400
6.29	1600
3.72	1800

Reaction of  $\text{Cm}_2\text{O}_3$  With  $\text{ZrO}_2$  and  $\text{Al}_2\text{O}_3$  (E. E. Ketchen)

In the application of  $^{244}\text{Cm}_2\text{O}_3$  as a heat source, it may be desirable to have the curium compact in contact with ceramic materials such as  $\text{ZrO}_2$  or  $\text{Al}_2\text{O}_3$ . Will the  $\text{ZrO}_2$  or  $\text{Al}_2\text{O}_3$  interact with  $\text{Cm}_2\text{O}_3$  in the 600-1000 $^{\circ}\text{C}$  temperature range?

Reaction With  $\text{ZrO}_2$  - The thermodynamic data needed to calculate the free energies of reaction between  $\text{ZrO}_2$  and  $\text{Cm}_2\text{O}_3$  are not available for either curium sesquioxide or the rare-earth sesquioxides. The rare-earth sesquioxides react with  $\text{ZrO}_2$  to form  $\text{RE}_2\text{Zr}_2\text{O}_7$  as one of the major compounds. The heat of reaction at 25 $^{\circ}\text{C}$  for Eq. 6 is shown in Table 10.<sup>6</sup> The heat capacities of these compounds have not been measured, hence it is not possible to calculate the heat of reaction for Eq. 6 at 600-1000 $^{\circ}\text{C}$ .



where

RE = one of the rare-earth elements.

Table 10. Enthalpy of Formation of  $\text{RE}_2\text{Zr}_2\text{O}_7$  From  $\text{RE}_2\text{O}_3$  and  $\text{ZrO}_2$  at 25 $^{\circ}\text{C}$ 

Compounds	$-\Delta H_f$ (kcal/mole)
$\text{La}_2\text{Zr}_2\text{O}_7$	30.1
$\text{Pr}_2\text{Zr}_2\text{O}_7$	28.7
$\text{Nd}_2\text{Zr}_2\text{O}_7$	26.5
$\text{Sm}_2\text{Zr}_2\text{O}_7$	25.5
$\text{Eu}_2\text{Zr}_2\text{O}_7$	19.1
$\text{Gd}_2\text{Zr}_2\text{O}_7$	18.1
$\text{Y}_2\text{Zr}_2\text{O}_7$	11.4

V. A. Krzhizhanovskaya and V. B. Glushkova<sup>7</sup> have performed experiments to study the reaction of  $\text{Nd}_2\text{O}_3$  with  $\text{ZrO}_2$  at various temperatures. They have studied the kinetics of the reaction as low as  $1200^\circ\text{C}$ . The reaction rate constants decrease as the temperature is lowered from  $1500$  to  $1200^\circ\text{C}$ . The activation energy of formation is  $112$  kcal/mole for the reaction of  $\text{Nd}_2\text{O}_3$  with  $2\text{ZrO}_2$  to form  $\text{Nd}_2\text{Zr}_2\text{O}_7$ . Since the activation energy of solid solutions of  $\text{ZrO}_2$  and  $\text{Nd}_2\text{O}_3$  is  $80$  kcal/mole, the solid solution will be formed as an intermediate product before the neodymium zirconate is formed.

The  $\text{Cm}_2\text{O}_3$  would be expected to form a curium zirconate ( $\text{Cm}_2\text{Zr}_2\text{O}_7$ ) similar to the rare earths. There would probably be little reaction between  $\text{Cm}_2\text{O}_3$  and  $\text{ZrO}_2$  at  $600$  to  $1000^\circ\text{C}$ , but the rate of reaction at the  $\text{Cm}_2\text{O}_3$ - $\text{ZrO}_2$  interface would be expected to increase with temperature.

Reaction With  $\text{Al}_2\text{O}_3$  - The thermodynamic data needed to calculate the free energy of formation of  $\text{CmAlO}_3$  have not been measured. However, the reactions to produce  $\text{CmAlO}_3$  (ref. 8),  $\text{CeAlO}_3$  (ref. 9), and  $\text{NdAlO}_3$  (ref. 10) have been investigated.

The  $\text{CmAlO}_3$  was prepared by the reaction of  $\text{Cm}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3$  powder at  $1380^\circ\text{C}$  in a 4% H-96% He atmosphere. Both  $\text{CeAlO}_3$  and  $\text{NdAlO}_3$  were prepared at temperatures above  $1600^\circ\text{C}$ . These aluminates are stable in the region below  $1000^\circ\text{C}$ , when under an inert atmosphere. In the absence of free energy data, the reaction of  $\text{Cm}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3$  below  $1000^\circ\text{C}$  cannot be predicted, but it is conceivable that a slow reaction may occur. Compatibility experiments are needed to determine the extent of  $\text{Cm}_2\text{O}_3$  reaction with  $\text{Al}_2\text{O}_3$ .

### $^{244}\text{Cm}_2\text{O}_3$ Property Characterization

#### Helium Release (*P. Angelini*)

The helium release furnaces, manifolds, and vacuum system were installed in a glove box. Also the measuring instruments including the helium leak detector, recorders, and vacuum meters were integrated into the system.

The three furnaces to be used in this experiment were tested at  $800^\circ\text{C}$ .

Preliminary vacuum and temperature checks were performed on the manifolds. Minor repairs were made and a final vacuum check was made on the manifolds.

Block diagram drawings of the equipment to be used in this experiment with procedures for operating the equipment and the glove box are being prepared.

The operating temperature at which the three experiments will be conducted are  $800$ ,  $650$ , and  $500^\circ\text{C}$ . The pellets needed for this experiment have been fabricated and are in storage. The experimental procedure for the helium release experiment will be basically the same as that of the previous

steady-state helium release experiments performed with  $^{244}\text{Cm}_2\text{O}_3$  (ref. 11) and  $^{238}\text{PuO}_2$  (ref. 12). The pellets will be heated in vacuum to the required temperature. The helium released from each pellet will be stored in their respective sample container tubes. At prescribed intervals, the helium from the storage tubes will be measured. When the helium release rate from a pellet is >95% of the helium generation rate, the initial part of the experiment will be completed and the sample assembly will be heated to  $1900^\circ\text{C}$  in a different furnace to determine the helium inventory in that sample.

#### Vapor Pressure (*P. Angelini and J. C. Posey*)

The high-temperature vacuum furnace has been installed in the glove box. Most of the pipe fitting and electrical work has been completed. The high-temperature vacuum furnace was helium leak checked. A leak was located in the liquid nitrogen cold trap, and a new trap was ordered.

The glove box windows have been installed, and checking of the glove box for air leaks has been completed. Various leaks were found and eliminated. The oxygen concentration in the glove box with a normal purge flow of  $0.5\text{ ft}^3/\text{min}$  argon is less than 50 ppm during operations.

The block diagrams and procedure for this experiment are being prepared.

#### Vapor Pressures of Curium, Plutonium, and Americium Oxides (*P. Angelini*)

Some of the reported vapor pressures of plutonium, curium, and americium oxides published in the literature are presented in Table 11.

A graph of the vapor pressure data listed in Table 11 is presented in Fig. 1. The identification numbers in Table 11 coincide with the identification numbers in Fig. 1.

In all of the above mentioned data the vapor specie was not determined experimentally but was inferred by calculation and vaporization trends in various other oxides. The experiments were all of the Knudsen cell type, and the vapor pressures were calculated using the Knudsen equation. However, the vapor specie has to be known so that a true vapor pressure can be calculated.

From the data presented it can be seen that the reported vapor pressure for  $\text{AmO}_2$  is greater than that of  $\text{PuO}_2$  or  $\text{Cm}_2\text{O}_3$ , and that the vapor pressure for  $\text{Am}_2\text{O}_3$  is much higher than the vapor pressure of either  $\text{Cm}_2\text{O}_3$ ,  $\text{PuO}_2$ , or  $\text{Pu}_2\text{O}_3 \cdot \text{Pu}_{1.6}$ . Thus a large content of americium oxide in curium oxide fuel may be detrimental from the vaporization standpoint in relationship to various capsule designs and time at temperature.

The vapor pressure reported for the  $\text{Pu}_2\text{O}_3 \cdot \text{Pu}_{1.6}$  system by Ackerman *et al.*<sup>13</sup> and Phipps *et al.*<sup>14</sup> agree quite well. The vapor pressure for this system is also higher than that of  $\text{Cm}_2\text{O}_3$  or  $\text{PuO}_2$ . The vapor pressure of pure

Table 11. Vapor Pressure of Plutonium, Curium, and Americium Oxides

Number	Solid Phase	Vapor Phase <sup>a</sup>	log P <sub>e</sub> (atm) = A + (B/T)		Temperature Range (°C)	Method (effusion cell)	Reference
			A	B			
1	PuO <sub>1.6</sub> ·Pu <sub>2</sub> O <sub>3</sub>	PuO	8.13	-27,910	1790-2075	Tantalum	b
2	PuO <sub>~1.6</sub> ·Pu <sub>2</sub> O <sub>3</sub>	PuO	8.09	-27,800	1645-2105	Tantalum	c
3	Cm <sub>2</sub> O <sub>3</sub>	Cm <sub>2</sub> O <sub>3</sub>	7.32	-29,050		Tungsten	d
4	Am <sub>2</sub> O <sub>3</sub>	AmO	8.19	-25,650	1645-2105	Tantalum	c
5	PuO <sub>~1.87</sub>	PuO <sub>2</sub>	7.82	-29,700	1990-2380	Tungsten	e
6	PuO <sub>~1.92</sub>	PuO <sub>2</sub>	7.50	-29,260	1640-2070	Tungsten	c
7	AmO <sub>2</sub>	AmO <sub>2</sub>	7.28	-28,260	1640-2070	Tungsten	c

<sup>a</sup>Calculated for log P<sub>e</sub> equation.

<sup>b</sup>T. E. Phipps, C. W. Learns, and O. C. Simpson, *J. Chem. Phys.* **18**, 724 (1950).

<sup>c</sup>R. J. Ackerman, R. L. Faircloth, and M. H. Rand, *J. Phys. Chem.* **70**(11), 3698-3706 (1966).

<sup>d</sup>P. K. Smith and D. E. Peterson, *J. Chem. Phys.* **52**(10), 4963-72 (1970).

<sup>e</sup>N. R. Mulford and L. E. Lamar, *Plutonium*, Cleaver-Hume Press, London, 1970, p. 19611,411.

Pu<sub>2</sub>O<sub>3</sub> has not yet been measured and may be quite different from that of the Pu<sub>2</sub>O<sub>3</sub>·PuO<sub>1.6</sub> system.

Theoretically Pu<sub>2</sub>O<sub>3</sub> would be formed as the <sup>244</sup>Cm decays in an initially pure Cm<sub>2</sub>O<sub>3</sub> fuel form in an environment where excess oxygen would not be available. The vapor pressure data for the Pu<sub>2</sub>O<sub>3</sub>·Pu<sub>1.6</sub> system were obtained using a tantalum Knudsen cell in both the work of Ackerman *et al.*<sup>13</sup> and Phipps *et al.*<sup>14</sup> The initial material loaded into the tantalum cell was PuO<sub>2</sub>. Thus, reduction of the PuO<sub>2</sub> occurred during the experiment, and the final stoichiometry of the vaporizing sample could not be determined with great accuracy in this method.

From work reported by Ackerman *et al.*<sup>13</sup> and Mulford and Lamar,<sup>15</sup> it is seen that the vapor pressure of PuO<sub>2</sub> tends to increase as the stoichiometry of the sample decreases. Also the activation energy for the vapor pressure of both the PuO<sub>1.92</sub> reported by Ackerman *et al.*<sup>13</sup> and

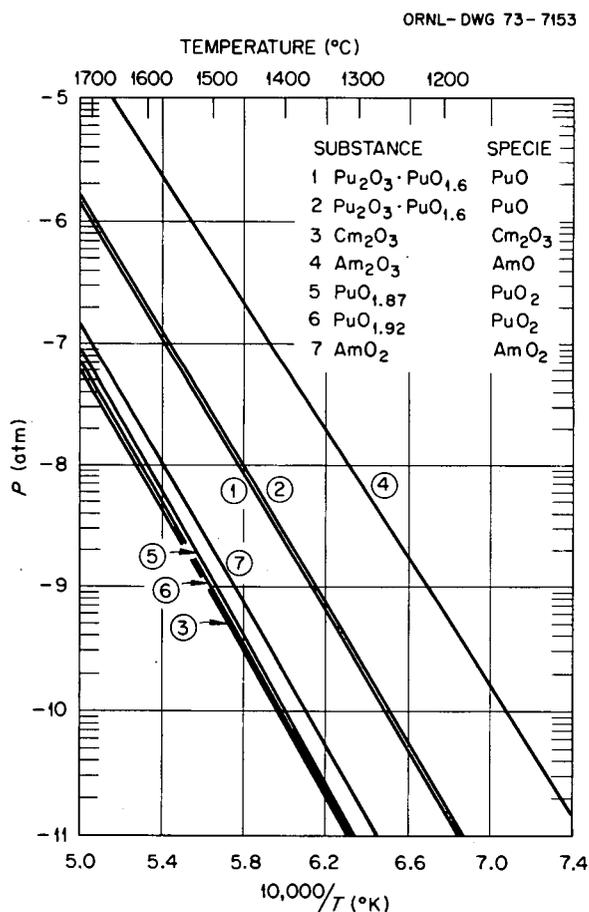


Fig. 1. Vapor Pressure Versus 1/T (°K).

that for  $\text{PuO}_{1.87}$  by Phipps *et al.*<sup>14</sup> is greater than that reported by Smith and Peterson<sup>2</sup> for the activation energy for the vapor pressure of  $\text{Cm}_2\text{O}_3$ . In using the vapor pressures reported by Ackerman *et al.*<sup>13</sup> for  $\text{PuO}_{1.92}$  and by Smith and Peterson<sup>2</sup> for that of  $\text{Cm}_2\text{O}_3$ , the temperature at which the vapor pressure for  $\text{PuO}_{1.92}$  becomes less than that for  $\text{Cm}_2\text{O}_3$  is 1166°K. This is a temperature which is nearly 500°C lower than the lowest temperature at which the vapor pressures of  $\text{PuO}_{1.92}$  and  $\text{Cm}_2\text{O}_3$  were experimentally determined. A 500°C extrapolation may result in rather large errors. The vapor pressure equation for  $\text{Cm}_2\text{O}_3$  was calculated using  $\text{Cm}_2\text{O}_3$  as the vaporizing specie. The vapor pressure for  $\text{Cm}_2\text{O}_3$  would be increased if the vaporizing specie is in reality  $\text{CmO}$ . If this were the case, the vapor pressure for  $\text{Cm}_2\text{O}_3$  would be higher than that of  $\text{PuO}_2$  at almost all significant temperatures.

It appears from the published data that the true vapor pressure of  $\text{PuO}_2$  and  $\text{Cm}_2\text{O}_3$  are somewhat similar in magnitude and activation energy. Also the vapor pressure of  $\text{Pu}_2\text{O}_3$  will probably be larger than that of  $\text{Cm}_2\text{O}_3$  although literature references for the vapor pressure of  $\text{Pu}_2\text{O}_3$  have not been found. The amount of  $\text{AmO}_2$  or  $\text{Am}_2\text{O}_3$  initially present in the fuel form and the ingrown  $\text{Pu}_2\text{O}_3$  (if its vapor pressure is near that of the  $\text{Pu}_2\text{O}_3 \cdot \text{PuO}_{1.6}$ ) may have significant effect on the amount of alpha-emitting material volatilized from the fuel.

#### X-Ray Diffraction Unit (*P. Angelini*)

The air leaks in the x-ray process glove box have been sealed. The oxygen content in the box can be held to less than 50 ppm with an argon purge flow of 0.5 ft<sup>3</sup>/min. For the initial purge, up to 4 ft<sup>3</sup>/min of argon may be used to lower the oxygen concentration to 50 ppm in a period of less than 1 hr. The glove box is of the stainless steel type with safety glass windows.

A Plexiglas entry glove box could not be adequately sealed from air leaks. In addition, safety regulations now require stainless glove boxes with safety glass windows. Thus, a new entry box also satisfying the new regulations will be needed. Design of the new glove box has been completed and fabrication of the new unit will begin July 1, 1973.

The x-ray diffraction system was partly aligned. The unit is now ~0.03° off in alignment. The final aligning procedure with the furnace hot stage in place has yet to be performed.

The first experiment planned with the system is to perform a scan on the first pellet used in the <sup>244</sup>Cm<sub>2</sub>O<sub>3</sub> solubility experiments. This may indicate whether the pellet has undergone any chemical change during those experiments.

#### Emissivity of <sup>244</sup>Cm<sub>2</sub>O<sub>3</sub> (*P. Angelini*)

A special flange which will greatly simplify sample loading onto the spectroradiometer has been fabricated and tested on the high-temperature vacuum furnace. The new flange will also allow the unit to be operated

in either of two furnaces available for this experiment. The flange operates as expected and will enable easier loading of the emittance assembly onto the furnace.

The spectroradiometer has been modified so that AC amplification can be used for signal recovery. Testing of the signal recovery system is continuing. The spectroradiometer unit will be tested before the glove box becomes contaminated with radioactive materials.

#### Heat Capacity (*P. Angelini*)

Curium sesquioxide ( $^{244}\text{Cm}_2\text{O}_3$ ) prepared from material freshly separated from the  $^{240}\text{Pu}$  daughter will be utilized in this determination in order to define the heat capacity of pure  $\text{Cm}_2\text{O}_3$ . The heat capacity of various mixtures of  $^{244}\text{Cm}_2\text{O}_3$  and  $^{240}\text{Pu}_2\text{O}_3$  can then be calculated. The thermocouple material to be pressed in the  $^{244}\text{Cm}_2\text{O}_3$  fuel pellet is W5Re-W26Re.

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

The dimensional stability of  $^{244}\text{Cm}_2\text{O}_3$  fuel forms will be determined to supply information relative to three principal conditions.

1. The shelf life of fabricated fuel pieces as functions of storage temperature, atmosphere, and time. The physical integrity and size changes will be determined.
2. Dimensional and structural changes of  $^{244}\text{Cm}_2\text{O}_3$  fuels in sealed containers that may influence the performance of the power system.
3. The effect of sudden exposure of fuel pieces to the atmosphere or bodies of water such as might occur upon reentry of fueled systems from space.

In preparation for initiation of experimental measurements of the dimensional stability of hot-pressed  $^{244}\text{Cm}_2\text{O}_3$  pellets, a resistance furnace, measuring test stand, and die bodies are being assembled for installation in the SFF hot cells.

#### Rate of Solution of $^{244}\text{Cm}_2\text{O}_3$ (*J. C. Posey*)

Measurements were completed on the rate of loss of  $^{244}\text{Cm}$  from the surface of a  $\text{Cm}_2\text{O}_3$  pellet exposed to flowing distilled water saturated with air. This water is considered to be equivalent to the natural rain in regions of low atmospheric pollution.

The experimental equipment is shown in Fig. 2. A steady stream of water flows from the constant head tank through the solution tube and into the water collection tank. The flow rate is measured by means of a rotometer. Samples are taken by placing the sample bottle under the outlet to the collection tank.

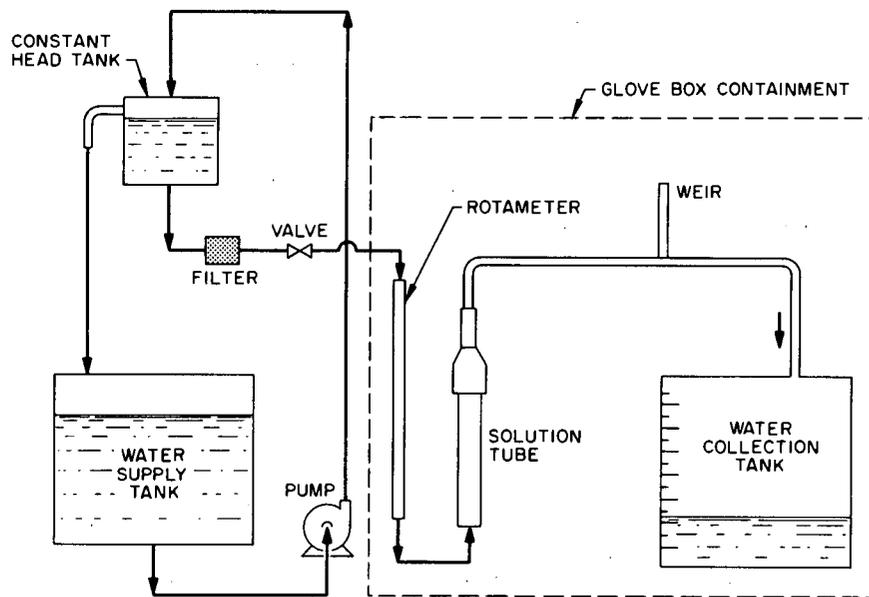


Fig. 2. Apparatus for Solution Rate Measurement.

Details of the solution tube are shown in Fig. 3. The water flows upward around the  $^{244}\text{Cm}_2\text{O}_3$  pellet. The maximum linear flow rate in the sample cavity occurs in the annular space between the cylindrical pellet and the container. It is 0.126 cm/sec when the normal flow rate of 100 ml/hr is used. The Reynold's number is less than 10. Thus, the flow is in the nonturbulent range.

All samples were acidified to reduce adsorption of curium by the sample bottle and to bring suspended oxide, if any, into solution. Some of the samples were filtered through a 0.1- $\mu\text{m}$  pore filter before acidification. An alpha-counting procedure was used to determine the  $^{244}\text{Cm}$  concentration.

The  $\text{Cm}_2\text{O}_3$  pellet, 0.46 cm in diameter and 0.158 cm thick, was hot-pressed 6 days and 15 hr before the beginning of the solution rate measurements.

The results are shown graphically in Fig. 4. The initial unfiltered sample indicated a much higher rate than the subsequent samples. This is attributed to the washing of loose particulate material from the surface of the pellet. The rate then became relatively constant. After 345 hr, a light grey material was observed to be settling in the outlet tube. After 432 hr, the pellet was observed to be disintegrating. Much higher rates of solution were then observed as might be expected from the large increase in surface area. The rates of solution shown on the graph are based on the original surface area of the pellet. The upward trend of the rate suggests progressing breakup of the pellet.

Because of an electrical power outage over a weekend, the flow of water was interrupted for a considerable period of time prior to the first

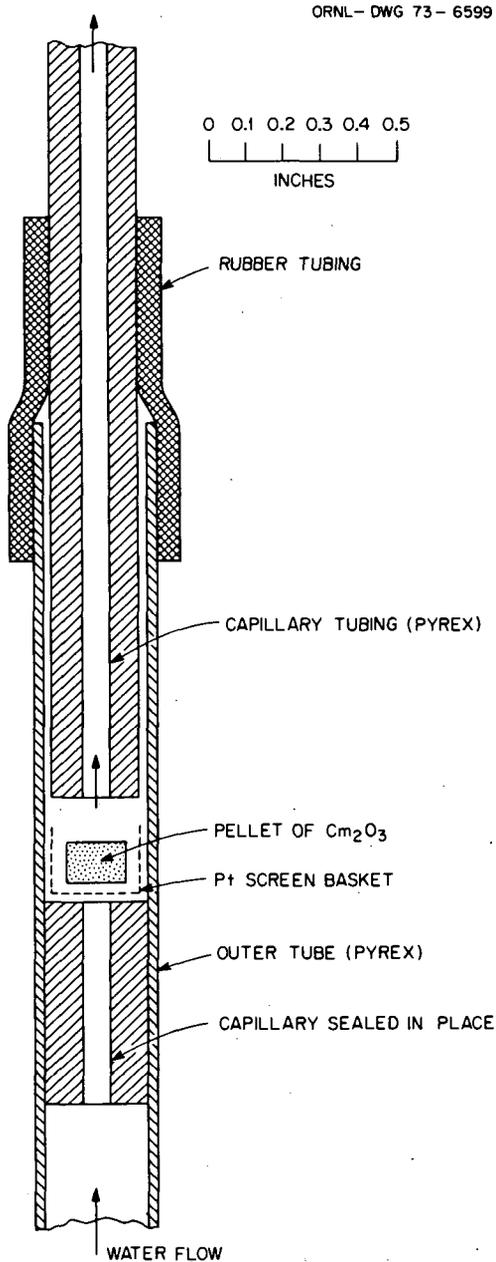


Fig. 3. Solution Tube.

The average rate of solution of  $^{244}\text{Cm}$  from the intact pellet was  $9.3 \mu\text{g}/\text{cm}^2\cdot\text{hr}$ . The standard deviation of points from the average was  $1.8 \mu\text{g}/\text{cm}^2\cdot\text{hr}$ . This value is based on 18 rates observed before the first appearance of sediment in the outlet tube. The first rate and four rates based on filtered samples were excluded in calculating the average. These samples are believed to have been contaminated by particulate material from the first sample which was deposited on the filter and in the filtration equipment. These rates are not shown in Fig. 4. Their inclusion in the average would raise the value to  $11.0 \mu\text{g}/\text{cm}^2\cdot\text{hr}$ .

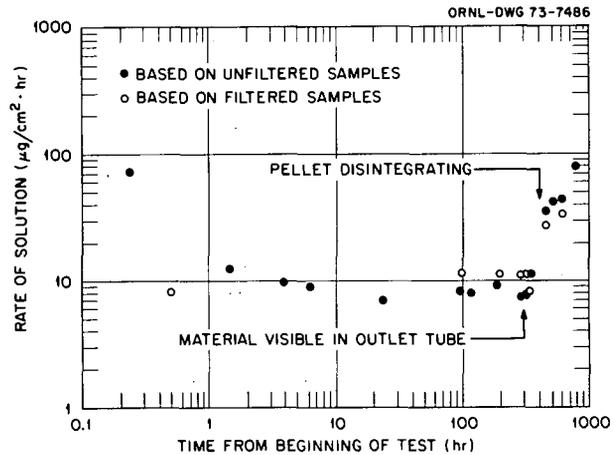


Fig. 4. Rate of Solution of Curium from Hot-Pressed  $\text{Cm}_2\text{O}_3$  Pellet.

observation of disintegration. This may have allowed an increase in the pellet temperature and hastened the reaction; however, the appearance of material in the outlet tube suggests that decomposition may have started before the power outage.

The conditions under which a pellet disintegrates are needed in addition to the absolute rate of solution from an intact surface for safety review information. Therefore, a series of aging experiments is planned in which pellets will be placed in beakers of water and held at constant elevated temperature. The times to visible disintegration will be noted. Exploratory work will be started with a pellet in distilled water at a temperature near  $100^\circ\text{C}$ . If rapid disintegration occurs, a need for further testing of the effect of temperature would be indicated. A more complete program would also involve the effects of pellet age, density, plutonium content, and water impurities.

After these filtered samples of high  $^{244}\text{Cm}$  content were observed, a policy of using a new filter for each sample was adopted. The absence of a consistent bias between the filtered samples and the unfiltered samples indicates that particulate material was not present in the samples. Any colloidal material leaving the surface of the pellet would, however, dissolve rapidly since the total curium concentration was less than the saturation concentration.

The  $\text{Cm}_2\text{O}_3$  solution rates are similar to those of  $\text{Pm}_2\text{O}_3$  as measured by Fullam and Roberts,<sup>16</sup> who observed rates which varied from 3.0 to 4.5  $\mu\text{g}/\text{cm}^2\cdot\text{hr}$  with low density  $^{147}\text{Pm}_2\text{O}_3$  pellets. With high density  $^{147}\text{Pm}_2\text{O}_3$  pellets they observed rates varying from 11.7 to 12.6  $\mu\text{g}/\text{cm}^2\cdot\text{hr}$ .

A test was made of the effect of water flow rate. Solution rates of 7 and 9  $\mu\text{g}/\text{cm}^2\cdot\text{hr}$  were observed at flow rates of 29 and 239 mg/hr, respectively. The difference between these values is not significant in view of the scatter of the data.

A steady stream of very fine bubbles could be seen rising from the surface of the pellet throughout the test. The bubbles are believed to be caused by the decomposition of water by alpha radiation. The rate of production, about 0.2  $\text{cm}^3/\text{hr}$ , greatly exceeds the rate of helium production. The accumulation of bubbles in the outlet tube caused some variation in the flow rate.

A test of the rate of solution of  $\text{Cm}_2\text{O}_3$  in seawater is in progress. The procedure is the same as that used with distilled water.

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

Modifications to the impact gun for hot cell operation were begun this quarter and are ~95% complete. An optical pyrometer is used at the present time for temperature measurements of the sample in the sabot heater. The best system for hot cell work will probably consist of a thermocouple attached to the sabot heat sink. The thermocouple attached to the heat sink will sense a temperature that will be calibrated to the sample temperature inside the impact cup. Other modifications to the gun include a holder for the target granite block that can be operated remotely.

A glove box large enough to contain the  $\pi\text{MC}$  microscope and all equipment needed for slide preparations has been obtained. Modifications to make the glove box compatible with all operations of the  $\pi\text{MC}$  system are ~75% complete. The light path housing of the microscope has been sealed with RTV silicon rubber to provide a leak-free glove box system. The microscope has been placed in the glove box and calibrated. Microscope slide preparation equipment has been designed and will be fabricated when funds become available.

Two samples were impacted during this report period. One sample was a 6.3764-g Mo-82%  $\text{ThO}_2$  pellet and the other a 2.9-g  $\text{Gd}_2\text{O}_3$  pellet. Analytical equipment to aid in the analysis of the samples in a glove box will

be designed and built. Weight distribution **analyses** on the Mo-ThO<sub>2</sub> and Gd<sub>2</sub>O<sub>3</sub> samples impacted during April have been partially completed. Analysis of the particles >45 μm was performed during this report period. Analysis of those particles ≤45 μm with the πMC particle analyzer will be performed after the slide preparation equipment and microscope have been installed in the glove box.

Europium oxide (Eu<sub>2</sub>O<sub>3</sub>, theoretical density 7.42 g/cm<sup>3</sup>) will be used as a nonradioactive fuel simulant for Cm<sub>2</sub>O<sub>3</sub> (theoretical density 11.67 g/cm<sup>3</sup>) in the next series of impact tests. The selection of a proper fuel simulant can best be made by comparing impact test results of the actual fuel (Cm<sub>2</sub>O<sub>3</sub>) with several candidate simulants. The selection of Eu<sub>2</sub>O<sub>3</sub> as the fuel simulant in the out-of-cell testing of the impact gun was based on europium oxide being of Type A (monoclinic) lattice structure. A second reason for the selection of Eu<sub>2</sub>O<sub>3</sub> was the existence of a program in the ORNL Metals and Ceramics Division in which they are presently fabricating Eu<sub>2</sub>O<sub>3</sub> pellets of varying size and density. The Metals and Ceramics Division has been requested to fabricate 18 pellets of Eu<sub>2</sub>O<sub>3</sub> ranging in size and density as shown in Table 12.

Table 12. Eu<sub>2</sub>O<sub>3</sub> Pellet Size and Density Specifications

Variable	Length (in.)	Diameter (in.)	% Theoretical Density	Number of Pellets
Length	1/8	1/4	90	3
	1/4	1/4	90	3
	3/8	1/4	90	3
Density	1/4	1/4	75	3
	1/4	1/4	87.5	3
Diameter	1/2	1/2	90	3

#### Curium Data Sheets (*T. A. Butler*)

Final revisions were made to the draft copy of the *Curium Data Sheets*. The major change resulted from a decision to use the isotopic composition of curium expected from the operation of light-water power reactors as a reference heat source rather than the Savannah River product. The effect of this change is to reduce the maximum heat content of the product by ~3.5%.

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

The following information was supplied by C. W. Kee of the Chemical Technology Division, Engineering Coordination and Analysis Section, concerning calculated yields of <sup>244</sup>Cm versus MWD/MT for enriched <sup>235</sup>U fueled and plutonium recycle fueled PWR reactors. Conditions of reactor operation are:

	Uranium Fueled	Plutonium Recycle
$^{235}\text{U}$ Enrichment (%)	3.3	
Power (MW/MT)	30	30
Flux ( $\text{n}/\text{cm}^2\cdot\text{sec}$ )	$2.92 \times 10^{13}$	$1.5 \times 10^{13}$
Flux Factors:		
Thermal	0.632	0.5
Resonance	0.333	0.55
Fast	2.0	4.0
$^{244}\text{Cm}$ (g/MT, discharge):		
<u>Burnup (MWD/MT)</u>		
3,300	$8.4 \times 10^{-5}$	19.1
6,600	$4.4 \times 10^{-3}$	76.6
9,900	$4.3 \times 10^{-2}$	172.3
13,200	0.21	305.
16,500	0.74	471.
19,800	2.02	671.
23,100	4.66	898.
26,400	9.49	1147.
29,700	17.6	1418.
33,000	30.26	1703.

The study of  $^{244}\text{Cm}$  availability from power reactor fuel reprocessing will be continued with emphasis on light water moderated reactors. The effect of  $^{235}\text{U}$  fuel enrichment on  $^{244}\text{Cm}$  yield at constant power levels will be examined next.

#### $^{244}\text{Cm}_2\text{O}_3$ Radiation Measurements (*K. W. Haff and T. A. Butler*)

The encapsulated source containing 61.3  $W_t$  was transferred to the Tower Shielding Facility in June, and the radiation measurements were completed. Analysis of the data is expected to be completed next quarter.

A complete assay was made of  $^{244}\text{Cm}$  oxide from batch SRL-17 which was used to prepare the source for the radiation measurements.

The isotopic abundances of the two principal elements, curium and plutonium, are given in Table 13.

Table 13. Isotopic Abundances of Curium and Plutonium in SRL-17 Oxide

Curium		Plutonium	
Mass No.	Isotopic Abundance (atom %)	Mass No.	Isotopic Abundance (atom %)
244	94.28	238	0.083
245	0.87	239	0.024
246	4.73	240	99.87
247	0.078	241	0.002
248	0.044	242	0.019
		244	0.005

The curium and plutonium content of SRL-17 was determined by radiochemical analysis for  $^{244}\text{Cm}$  and  $^{240}\text{Pu}$  and the total element weight calculated by use of the isotopic abundances given in Table 13. The results are shown in Table 14.

Table 14. Curium and Plutonium Content in SRL-17 Oxide

Sample No.	Weight (mg)	Curium (mg/g)	Plutonium (mg/g)	$\text{Cm}_2\text{O}_3$ (mg/g)	$\text{PuO}_2$ (mg/g)
Cm17-1	8.50	738	101	811	114
Cm17-2	9.61	738	103	811	117
Average		738	102	811	116
Adjusted Average <sup>a</sup>		791	109	869	124

<sup>a</sup>Based on calorimetric value of 2.11 W of  $^{244}\text{Cm}$  per gram.

The radioactive impurities were determined by standard germanium crystal gamma spectrometry and are listed in Table 15.

Table 15. Radioactive Impurities in SRL-17 Oxide

Nuclide	Curies/g
Ruthenium-103	$\leq 5 \times 10^{-6}$
Ruthenium-106	$\leq 2 \times 10^{-5}$
Cesium-134	$\leq 4 \times 10^{-6}$
Cerium-144	$\leq 4 \times 10^{-4}$
Praseodymium-144	$\leq 4 \times 10^{-4}$
Zirconium-95	$\leq 8 \times 10^{-6}$
Niobium-95	$\leq 3 \times 10^{-6}$
Europium-154	$3.1 \times 10^{-5}$
Americium-243	$6 \times 10^{-3}$

The nonradioactive impurities were determined by spark source mass spectrometry and are listed in Table 16. Conversion of the major metallic impurities (>10 ppm) to fully oxidized forms equates to 7.7 mg/g.

Table 16. Impurities in Batch SRL-17 Curium Oxide

Element	Wt (ppm of Cm)	Element	Wt (ppm of Cm)
Aluminum	50	Lead	40
Arsenic	<10	Platinum	500
Boron	10	Silicon	100
Calcium	100	Tantalum	200
Cobalt	10	Titanium	10
Chromium	400	Zinc	10
Copper	50	Zirconium	250
Iron	2000	Americium	3600
Potassium	10	Chlorine	20
Magnesium	30	Fluorine	10
Manganese	50	Sulfur	80
Molybdenum	100	Lanthanum	40
Sodium	50	Cerium	40
Nickel	100	Other rare earths	100

In summary, the composition of SRL-17 curium oxide is:

<u>Material</u>	<u>mg/g</u>	<u>Percent</u>
Cm <sub>2</sub> O <sub>3</sub>	869	86.8
PuO <sub>2</sub>	124	12.4
Impurities	7.7	0.8

Radioactive impurities are at or below the limits of detection with the exception of <sup>154</sup>Eu ( $3.1 \times 10^{-5}$  Ci/g) and <sup>243</sup>Am ( $6 \times 10^{-3}$  Ci/g).

#### Curium Oxide Purification (*T. A. Butler*)

Arrangements were made with the ORNL TRU facility to reprocess a batch (No. 72-Cm-4) of curium oxide recovered from the Kilowatt Heat Source Test containing about 50 g of <sup>244</sup>Cm. The purpose of the reprocessing is to remove the ~15% PuO<sub>2</sub> content which has accumulated as a result of decay of <sup>244</sup>Cm. This will provide ~45 g of initial product-quality Cm<sub>2</sub>O<sub>3</sub> on which to make characterization studies and relate them to property measurements made with variable levels of PuO<sub>2</sub> content. The reprocessed material is expected to be available by August.

#### Curium Source Fabrication Facility Operation

(*C. L. Ottinger*)

A total of 84 pellets were pressed during the preparation of the compatibility couples in March and April. The average density was 8.5 g/cm<sup>3</sup>, but one batch of nine pellets was considerably below average, ranging

from 5.9 to 7.0 g/cm<sup>3</sup>. Neglecting the low density batch, the average density of the other 75 pellets was 8.8 g/cm<sup>3</sup>. All of these pellets had been pressed at 4000 psi and 1450°C with pressure being held for about 1/2 hr. Another pressing was done in which all three conditions were varied. The pressure was increased to 5000 psi; the temperature was raised to 1525°C; and the pressing time was increased to 1 hr. Unusually high ejection pressure was required to remove the punches, and no usable pellets were obtained. A second pressing was done using the original procedure and nine intact pellets were recovered (one punch broke). The average density of the pellets was 8.4 g/cm<sup>3</sup>. After the residual carbon had been oxidized two of the pellets were transferred for solubility studies; the others were stored.

The reasons for the relatively low densities of the pellets are not known. The use of the multiple die and the small size of the pellets probably contribute to the results since minor variations in conditions can have significant effects. The fuel material used for both test pressings was from the same batch (SRL-18) used for the compatibility pellets, but it had been recalcined at 1325°C in vacuum.

Some excess compatibility pellets plus a set of smaller pellets which had been made for characterization work were stored in an argon-purged vessel for 10 days while the compatibility samples were being encapsulated. Nearly all of these pellets were found to be broken when the vessel was opened. It is thought that air leaked into the vessel, causing partial re-oxidation of the Cm<sub>2</sub>O<sub>3</sub> to CmO<sub>2</sub>.

Material from batch SRL-17 was pressed into two pellets for use in a shielding experiment. This material was similar in age and chemical composition to the compatibility fuel. The 32-g fraction was broken into small grains which were spread in a thin layer in a 2-in.-diam platinum dish for calcination at 1000°C in vacuum for 4 hr. There was no change in the appearance of the fuel except for the formation of some small agglomerates which were easily broken up. Two 1.35-cm-diam pellets were pressed at 1450°C, one at 4000 psi and one at 4500 psi. The full pressure was applied when the die temperature was ~1100°C and maintained until there was no additional movement of the ram at 1450°C (30 min in each case). Most of the ram travel occurred in the temperature range of 1300 to 1350°C. The two pellets were similar, with densities of 9.8 and 9.7 g/cm<sup>3</sup>.

During the die loading operations for the 1.35-cm pellets some tamping experiments were done in which increments of fuel were compacted by manipulator-handled tools in a 1.1-cm-diam tube. Although the method used for packed height measurement was not really accurate, the indicated densities for three increments were in the range of 5.5 to 6.0 g/cm<sup>3</sup>. Also the loaded die assemblies were measured carefully, and both showed pre-pressed density of 5.9 g/cm<sup>3</sup>. Some source designers are considering the use of lower density fuel. These preliminary tests indicate that densities on the order of 60% of the hot-pressed density could be achieved by simple procedures.

The two 1.35-cm-diam  $^{244}\text{Cm}$  pellets were encapsulated into a test source, and the source was delivered to the Tower Shielding Facility. Significant data for these pellets are given below:

	Pellet <u>SE-1</u>	Pellet <u>SE-2</u>	Total <u>(average)</u>
Height (cm)	1.11	1.15	2.26
Weight (g)	15.62	16.21	31.83
Density (g/cm <sup>3</sup> )	9.8	9.9	(9.85)
Watts (direct assay)	32.5	34.8	67.3
Power Concentration (W/g)	2.08	2.14	(2.11)
Power Density (W/cm <sup>3</sup> )	20.4	21.1	(20.75)

During the pellet preparation and source assembly, the cell atmospheres were argon with oxygen content <200 ppm except for the residual carbon removal operation, which was done in argon at 2500 ppm oxygen by heating at 1000°C for 8 hr. When pellet SE-2 was removed from the furnace after the carbon removal, a hairline crack was noted across the diameter of the top surface. The pellets were assayed individually by calorimetry. Inspection of the pellets after calorimetry showed that pellet SE-2 had separated into two pieces and that pellet SE-1 had developed a hairline crack across the diameter of the top surface. The pellets, still in their platinum calorimeter cups, were stored overnight in vacuum, and pellet SE-1 also separated into two pieces.

Each pellet piece was visually inspected with a 6X monocular through the cell window. The exposed surfaces were smooth, uniform, and fine-grained; there was no evidence of chipping or flaking. In both cases the fracture was along the vertical axis, dividing the pellet into essentially equal pieces. The two halves of each pellet were fitted together with forceps and inserted into the 1.41-cm-diam inner capsule without difficulty. No material loss was detected either by weighing or by calorimetry of the empty storage cups. The inner capsule was welded by the GTA process in argon; then it was inserted into the outer capsule which was welded by GTA in air.

Fuel batch SRL-17 was used for these test pellets. This material was processed in November 1969. Assays in June 1973 showed a power concentration of 2.04 W/g in the  $\text{CmO}_2$  form and 2.11 W/g in the  $\text{Cm}_2\text{O}_3$  form (from pellet data); the  $^{244}\text{Cm}$ /total curium ratio is 0.943. From these data the pellet composition would be 86.8%  $\text{Cm}_2\text{O}_3$ , 11.6%  $\text{PuO}_2$ , and 1.6% other impurities (analytical results are reported in  $^{244}\text{Cm}_2\text{O}_3$  Radiation Measurement section, p. 21). These results are in good agreement with the preparation history of this fuel batch and the analytical results.

The most recently processed fuel batch, SRL-31 (January 1971), was opened and some of the material removed for tests. The fuel, in the form of granular powder, came out of the annular shipping can easily. Some fractions of the powder were stored in platinum containers for observation. A 30-g fraction in a layer ~7.5-cm diameter by 0.5-cm deep remained in the powder form when stored in air for five days. A similar fraction,

20 g in a layer 5-cm diameter by 0.5-cm deep, was calcined to the  $\text{Cm}_2\text{O}_3$  form and only a light crust formed. However, a 12-g fraction in a layer ~2-cm diameter by 2-cm deep formed into a hard mass while being assayed on the calorimeter. These observations are in accord with previous experience in that the fuel must be handled in relatively thin layers to prevent agglomeration.

Two runs of pellets, pressed from batch SRL-31 on the 0.46-cm, ten-place multiple die, yielded 19 pellets. These were stored overnight in vacuum and one batch of ten pellets disintegrated to powder. Both batches had been pressed under the same conditions. The batch which disintegrated had been held in the die body for 10 hr prior to ejection; also, these pellets had been stored in a platinum wire basket and the others in a ceramic dish. It is not known whether either of these differences could account for the breakage. Since the breakage occurred before density measurements were made, it may be that this was originally a poor quality batch, and the breakage was simply mechanical. Another pressing was made which yielded ten good pellets. After residual carbon oxidation, the ends of the pellets were polished and the pellets stored in individual containers. The average density of the 19 pellets was  $9.9 \text{ g/cm}^3$ . This is significantly better than the average densities realized on pellets of the same nominal size made from batches SRL-17 ( $8.4 \text{ g/cm}^3$ ) and SRL-18 ( $8.8 \text{ g/cm}^3$ ). Samples of batch SRL-31 were submitted for analysis. Five SRL-31 pellets and one SRL-17 pellet were transferred to experimental facilities.

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