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ORNL ISOTOPIC POWER FUELS
QUARTERLY REPORT FOR
PERIOD ENDING DECEMBER 31, 1973

Eugene Lamb

MASTER

OAK RIDGE NATIONAL LABORATORY

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Eugene Lamb

Isotopes Division

FEBRUARY 1974

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CONTENTS

	<u>Page</u>
SUMMARY	1
CURIUM-244 FUEL DEVELOPMENT	2
$^{244}\text{Cm}_2\text{O}_3$ Compatibility Program	2
Compatibility Couple Tests	2
FY 1974 Compatibility Matrix	5
Compatibility With Ceramic Materials	7
Free Energy Calculations — Cm_2O_3 With Haynes 188, Ta-10% W, and TZM	9
Effect of Oxygen Solubility in Container Alloys on Interaction With Curium Oxide Heat Sources	14
Free Energy Calculations for the Reaction of Beryllium With Cm_2O_3 Heat Source Product	16
Data Needed for Calculation of Cm_2O_3 Free Energy	17
Summary of Thermodynamic Calculation	18
$^{244}\text{Cm}_2\text{O}_3$ Property Characterization	19
Helium Release	19
Vapor Pressure	22
X-Ray Diffraction	24
Emissivity	26
Heat Capacity	26
Dimensional Stability	26
Behavior of Hot-Pressed Cm_2O_3 Pellet in Boiling Distilled Water	27
$^{244}\text{Cm}_2\text{O}_3$ Radiation Measurements	29
Curium From Power Reactor Fuels	29
Criticality of $^{244}\text{Cm}_2\text{O}_3$ Fuels	33
REFERENCES	33

LIST OF FIGURES

	<u>Page</u>
Fig. 1. Electron-Beam Scanning Images of Cm_2O_3 -Pt Compatibility Test	4
Fig. 2. $^{244}\text{Cm}_2\text{O}_3$ Mechanical Properties Compatibility Test System	6
Fig. 3. Temperature Versus Time for Helium Release Tests	19
Fig. 4. Helium Release Rate Versus Time (0-152 hr)	19
Fig. 5. Helium Release Rates Versus Time (0-900 hr)	21
Fig. 6. Comparison of Calculated and Measured Yield of ^{244}Cm in Enriched-Uranium-Fueled PWR	30

LIST OF TABLES

Table 1. Status of Compatibility Tests	3
Table 2. $^{244}\text{Cm}_2\text{O}_3$ /Ceramic Compatibility Experiments	7
Table 3. Partial Molar Free Energies of Mixing of the Constituents in Haynes 188 Alloy	9
Table 4. Gibbs Free Energy of Formation of Oxides Involved in Haynes 188/Curium Fuel Reactions	10
Table 5. Free Energies of Reaction for Haynes 188/ Cm_2O_3 Reactions	11
Table 6. Free Energies of Reaction for Haynes 188/ PuO_2 Reactions .	11
Table 7. Partial Molar Free Energies of Mixing of Tungsten and Tantalum in Ta-10% W Alloy	12
Table 8. Gibbs Free Energy of Formation of Oxides Involved in Ta-10% W/Curium Fuel Reactions	12
Table 9. Free Energies of Reaction for Ta-10% W/Curium Fuel Reactions	12
Table 10. Partial Molar Free Energies of Mixing of Constituents in TZM	13
Table 11. Gibbs Free Energy of Formation of Oxides Involved in TZM/Curium Fuel Reactions	13

	<u>Page</u>
Table 12. Free Energies of Reaction for TZM/Curium Fuel Reactions	14
Table 13. Equilibrium Oxygen Partial Pressures Above PuO_2 and Average Equilibrium Oxygen Pressure for PuO_2 Decomposition to Pu_2O_3	15
Table 14. Gibbs Free Energy as a Function of Temperature	17
Table 15. Disintegration of Cm_2O_3 Pellet in Boiling Distilled Water	28
Table 16. Isotopic Abundance of Curium Recovered From Boiling Water Reactor Fuel	29
Table 17. Influence of ^{235}U Enrichment on Isotopic Abundance of ^{244}Cm at 33 GWD/MTU and Four-Year Decay Period	31
Table 18. Influence of Burnup on Isotopic Abundance of ^{244}Cm at 3.3% ^{235}U Reactor Fuel Enrichment and Four-Year Decay Period	32
Table 19. Power Density of $^{244}\text{Cm}_2\text{O}_3$ Fuel Forms	32

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SUMMARY

Heat soak periods were completed on the remaining 49 couples of the compatibility matrix which was put on test in May 1973. Examination of the 18 couples which were taken off test in August 1973 was completed. The planned short-term ceramic compatibility matrix exposures were completed, and optical microscope examinations were done. Experiments were started to develop fabrication techniques for preparation of an 11-couple compatibility matrix scheduled for FY 1974. The design of this matrix was finalized, and component procurement was started. Free energy calculations were made for the reactions of Cm_2O_3 fuel with Haynes 188, Ta-10% W, and TZM alloys and with beryllium.

Measurements of the helium release characteristics of hot-pressed $^{244}\text{Cm}_2\text{O}_3$ were started at 500, 650, and 800°C. A vapor pressure determination was run on curium metal in the temperature range from 900 to 1800°C. Vapor pressure measurements of $^{244}\text{Cm}_2\text{O}_3$ in the range from 1200 to 1600°C were begun; these will continue next quarter. Examinations by x-ray diffraction were made on materials resulting from exposures of $^{244}\text{Cm}_2\text{O}_3$ to flowing seawater, boiling seawater, and dry air as well as on $^{244}\text{Cm}_2\text{O}_3$ samples taken at various stages in the fuel forming process. The equipment which will be used for emissivity measurements was improved and checked with nonradioactive samples.

Observations of two 25-W $^{244}\text{Cm}_2\text{O}_3$ pellets stored in argon and one 25-W $^{244}\text{Cm}_2\text{O}_3$ pellet stored in dry air continued. A $^{244}\text{Cm}_2\text{O}_3$ pellet was leached with boiling distilled water and the rate of ^{244}Cm loss measured. Computational studies of ^{244}Cm yields and isotopic abundances from various types of reactor operations were continued. A study of the potential for criticality problems in ^{244}Cm assemblies was initiated.

CURIMUM-244 FUEL DEVELOPMENT

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

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

The compatibility test matrix outlined in Table 1 has been completed, and examination of the couples exposed at 1100°C for 2500 hr (sets 3, 4, 5, and 6) has also essentially been completed. Results of these tests can be summarized as follows:

Platinum — Reacted with Cm_2O_3 . Microprobe analysis indicates that the reaction phase contains ~72% platinum and 14% curium.

Pt-2608M — Surface roughening and grain boundary attack up to maximum of 1 to 2 mils. Oxygen pickup from 10 to 90 ppm.

Pt₃Ir — Generally no attack. Isolated areas of grain boundary attack from 1 to 3 mils.

Iridium — Generally no attack. Attack to <1 mil depth noted in one specimen.

Graphite — No attack.

Of the five materials tested only unalloyed platinum appears unsuitable for containing $^{244}\text{Cm}_2\text{O}_3$ at 1100°C. Test conditions included both helium and vacuum and under both conditions capsules were in contact with graphite or not. It was not apparent that these variables significantly affected the test results. The inner capsules were in contact with graphite in sets 3 and 6 but no reaction with the graphite was observed. Samples heat treated under the test conditions, but in the absence of $^{244}\text{Cm}_2\text{O}_3$ showed no unusual microstructural changes.

Unalloyed platinum reacted with Cm_2O_3 under all conditions after 2500 hr at 1100°C. Electron beam microprobe data from one of the tests is summarized in Fig. 1. The reaction product is the area labeled "b" in the upper left hand picture. It contains 72% platinum and 14% curium from x-ray intensity data. If these data are normalized to 100% (i.e., assume no other constituents were present), the values would be 84% Pt-16% Cm. This corresponds stoichiometrically to a compound of the type CmPt_5 . In addition to the reaction product there was a darker gray phase around the outer edge of $^{244}\text{Cm}_2\text{O}_3$ pellet as indicated by the backscattered electron picture in the lower left of Fig. 1. However, x-ray intensity data indicated it to contain the same curium concentration as the bulk pellet.

Sets 1, 2, 7, 8, 9, and 10, shown in Table 1, have now completed their exposures at 900 to 1400°C. Examination of these materials is scheduled to begin in early February.

Table 1. Status of Compatibility Tests

Set ^a	Start of Test	Thermal Cycles	Cumulative Hours	End of Test
1	5-3-73	1	5021	12-3-73
2	5-3-73	1	5021	12-3-73
3	4-16-73	3	2560	8-6-73
4	4-16-73	3	2560	8-6-73
5	4-17-73	2	2510	8-9-73
6	4-17-73	2	2510	8-9-73
7	5-3-73	2	4892	11-27-73
8	5-3-73	2	4892	11-27-73
9	5-3-73	3	5016	12-4-73
10	5-3-73	3	5016	12-4-73

^aSet identifications:

1. 900°C; 5000 hr; helium atmosphere; Ir, C, Pt, Hf-1% Pt-0.5% Pd, Hastelloy C-276, Haynes 25, Haynes 188, ThO₂, 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, ThO₂, Pt-20% Rh, and Pt-26% Rh-8% W.
3. 1100°C; 2500 hr; graphite, helium atmosphere; Ir, Pt, Pt₃Ir, and Pt-26% Rh-8% W-0.5% Ti.
4. 1100°C; 2500 hr; dynamic vacuum; Ir, Pt, Pt₃Ir, C, and Pt-26% Rh-8% W-0.5% Ti.
5. 1100°C; 2500 hr; helium atmosphere; Ir, Pt, Pt₃Ir, C, and Pt-26% Rh-8% W-0.5% Ti.
6. 1100°C; 2500 hr; graphite, dynamic vacuum; Ir, Pt, Pt₃Ir, and Pt-26% Rh-8% W-0.5% Ti.
7. 1400°C; 5000 hr; graphite, helium atmosphere; 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.

Backscattered Electrons

Cm M_{α} Pt M_{α}

Backscattered Electrons

Cm M_{α} Pt M_{α} Fig. 1. Electron-Beam Scanning Images of Cm_2O_3 -Pt Compatibility Test.

	<u>Area A</u>	<u>Area B</u>	<u>Area C</u>	<u>Area D</u>
% Concentration	95 Cm 0 Pt	72 Pt 14 Cm	95 Cm 0 Pt	100 Pt

FY 1974 Compatibility Matrix

(T. A. Butler, J. R. DiStefano, C. L. Ottinger)

A test system that will incorporate mechanical properties specimens for evaluation with $^{244}\text{Cm}_2\text{O}_3$ has been designed (Fig. 2). A test package consisting of four sheet tensile specimens and two fuel frames containing two rectangular $^{244}\text{Cm}_2\text{O}_3$ samples is held together by two nuts and bolts made from the material being evaluated. Test conditions selected are as indicated below:

5000 hr — 900°C <u>Argon-Graphite</u>	5000 hr — 1100°C <u>Vacuum-Graphite</u>	10,000 hr — 1400°C <u>Vacuum-Graphite</u>
Pt-3008/TZM ^a	Pt-3008	W-26% Re
Haynes 188/Ta-10% W ^a	Mo-46% Re	Mo-46% Re
Mo-46% Re	Ta-10% W	Ta-10% W
TZM		
Ta-10% W		

^aOuter sealed container surrounding vented inner capsule.

Procurement and fabrication of test components are now underway. The alloys Haynes 188, Ta-10% W, TZM, and W-26% Re are being obtained from commercial sources. The alloys Mo-46% Re and Pt-3008 are not commercially available, and material is being prepared by melting, casting, and sheet rolling procedures. Procedures to form cylindrical tubes from sheet are also being developed for these materials.

Tensile specimens of all of the materials except W-26% Re are to be die-punched from sheet at room temperature. The W-26% Re alloy tends to develop edge cracks when sheared, and, therefore, tensile specimens of this material will have to be prepared by machining and grinding operations.

In addition to the mechanical properties tests, two ceramic materials, ZrO_2 and Si_3N_4 , will be tested with $^{244}\text{Cm}_2\text{O}_3$ for 5000 hr at 900°C. Capsules will be vented to an argon atmosphere and will be in contact with graphite. The ZrO_2 test specimen is rigidized felt, and the container material is Y_2O_3 stabilized ZrO_2 . The Si_3O_4 for these tests is to be supplied by Gulf General Atomic. Three disk specimens have been received, but containers have not yet been fabricated.

Fabrication procedures for the $^{244}\text{Cm}_2\text{O}_3$ fuel pieces are being developed. The planned procedure uses a slotted die body and rectangular punch to form a $^{244}\text{Cm}_2\text{O}_3$ fuel piece $1/2 \times 1/8 \times \sim 0.04$ in. by hot pressing. Test pressings with Gd_2O_3 have given mixed results, but the problems are mechanical in nature and the technique should prove usable.

Fig. 2. $^{244}\text{Cm}_2\text{O}_3$ Mechanical Properties Compatibility Test System.

Compatibility With Ceramic Materials (*P. Angelini*)

The short-term ceramic compatibility experimental matrix started last quarter is given in Table 2. The results of experiments A, B, E, and F have been reported previously.¹ The other exposures were completed and examinations were started. After a 401-hr exposure of couples C, G, and I, a second Cm_2O_3 pellet/specimen set was placed on top of the original set in each capsule; then the exposure was continued for an additional 512 hr. In experiments D, H, and J, the fuel pellets were fabricated, loaded into the couples along with the proper specimens, and taken to temperature within an elapsed time of two days. The time was held to a minimum in order to minimize any possible deformation of pellets due to helium generation, thus providing better solid/solid contact between the pellets and the inserts. In addition to the fuel pellet, ~150 mg of powder obtained by crushing hot-pressed fuel pellets was placed in each capsule. Additional solid/solid contact area was expected to occur between the powder and the ceramic insert.

Table 2. $^{244}\text{Cm}_2\text{O}_3$ /Ceramic Compatibility Experiments

Experiment	Material	Temperature (°C)	Time (hr)	
			Planned	Actual
A	ThO_2	1500	24	24
B	ThO_2	1300	100	100
C	ThO_2	1500	500	512/913
D	ThO_2	1300	500	733
E	ZrO_2	1500	24	24
F	ZrO_2	1300	100	100
G	ZrO_2	1500	500	512/913
H	ZrO_2	1300	500	733
I	Al_2O_3	1500	500	512/913
J	Al_2O_3	1300	500	733

Optical microscopy was completed on couples C, G, H, and I; preliminary examination of couples D and J was done.

Couple C (ThO_2 , 512/913 hr, 1500°C)

This diffusion couple was seen to be cracked when it was removed from the heating furnace. When it was unloaded at HRLEL, the capsule of the diffusion couple had separated into three pieces. Two mounts were made of the most important pieces. Viewing these mounts from a low power microscope, one sees that extensive reaction did occur. Where the curium had reacted with the thoria, the thoria had a bluish color, and a concentration gradient was clearly evident. The thoria inserts in contact with the lower curia fuel pellet which had been exposed for a total of 913 hr at 1500°C showed curium had diffused into the thoria to a distance >30 mils. The inserts in contact with the upper pellet which had been exposed

at 1500°C for 512 hr showed curium had diffused into the thoria to an extent of 15 mils. Also the lower fuel pellet was completely broken up. The upper fuel pellet was still in one piece; however, a reaction product could be seen in the fuel pellet.

Couple D (ThO₂, 733 hr, 1300°C)

A reaction occurred in the experiment with diffusion couple D. Again the reaction occurred to a greater extent between the Cm₂O₃ powder and insert than between the pellet and insert. A reaction zone >6 mils was seen in the ThO₂ capsule and insert. The zone appeared to be blue in color.

Couple G (ZrO₂, 512/913 hr, 1500°C)

No extensive reaction areas could be seen in this diffusion couple as viewed at this time.

Couple H (ZrO₂, 733 hr, 1300°C)

The optical microscopy was completed on diffusion couple H. A dark area ~3 mils in thickness completely surrounding the ²⁴⁴Cm₂O₃ pellet was recorded. This effect was noted previously in the other ZrO₂ diffusion couples. Since this phenomenon seems to occur only in the Cm₂O₃ pellets exposed to ZrO₂, it is an indication of a reaction.

Couple I (Al₂O₃, 512/913 hr, 1300°C)

A reaction occurred in this diffusion couple. There were surface regions on both the curium oxide and aluminum oxide where extensive reaction occurred. Throughout the curium oxide pellet there was an interconnected linear second phase; the concentration of this phase within the fuel pellet decreased as the distance from the reaction area increased. This second phase did not exist in the as-fabricated pellet or in any of the previous diffusion couples which have been analyzed. Another concentration gradient seen as a varying grayish tone was observed in the aluminum oxide inserts and capsule. It appears as if there were almost thorough interdiffusion throughout the fuel pellet, inserts, and capsule.

Couple J (Al₂O₃, 733 hr, 1300°C)

The preliminary data from diffusion couple J indicated complete interdiffusion between inserts and fuel pellets. A definite discoloration of the Al₂O₃ inserts and parts of the Al₂O₃ container could be seen. Some cracking of the capsule also occurred. The ²⁴⁴Cm₂O₃ powder placed at the bottom of the container was found to interact much more with the container than did the Cm₂O₃ pellet. This signifies that a much better contact area occurred between the Cm₂O₃ powder and Al₂O₃ insert than between the contact area of the pellet and insert.

Free Energy Calculations — Cm_2O_3 With
Haynes 188, Ta-10% W, and TZM (E. E. Ketchen)

Reactions between Cm_2O_3 and the constituents of the alloys Haynes 188, Ta-10% W, and TZM were investigated from a thermodynamic viewpoint in the temperature range of 600-1300°C. In all cases, the partial molar free energies of mixing for the constituents of the alloy were calculated using the method reported previously¹ for the reaction of Cm_2O_3 and PuO_2 in certain alloys. The free energies of formation for the oxides were calculated from a data compilation by Glassner² and a review article by Oetting.³ The free energies of Pu_2O_3 were used as stand-ins for those of Cm_2O_3 . In calculating the heat of mixing, it was assumed that there is no compound formation between the constituents of the alloy.

Reaction of Cm_2O_3 With Haynes 188 — The results of the calculations of the partial molar free energies of mixing for the constituents in Haynes 188 are shown in Table 3. The composition of Haynes 188 is 41.87 atom % Co, 26.50 atom % Cr, 23.47 atom % Ni, 4.77 atom % W, 1.68 atom % Fe, 0.86 atom % Mn, 0.44 atom % Si, 0.41 atom % C, and 0.00004 atom % La.

Table 3. Partial Molar Free Energies of Mixing of
the Constituents in Haynes 188 Alloy

Component	$-\Delta\bar{G}_M$ (cal/mole)				
	600°C	800°C	1000°C	1200°C	1300°C
Co	1,460	1,800	2,150	2,500	2,670
Cr	1,980	2,510	3,040	3,570	3,830
Ni	2,510	3,080	3,660	4,230	4,520
W	5,030	6,240	7,450	8,660	9,260
Fe	7,090	8,710	10,340	11,960	12,770
Mn	8,230	10,120	12,020	13,910	14,860
Si	9,320	11,470	13,620	15,770	16,840
C	9,460	11,650	13,830	16,020	17,110
La	17,750	21,820	25,890	29,950	31,990

The free energies of formation of CoO , Cr_2O_3 , NiO , WO_2 , Fe_3O_4 , MnO_2 , SiO_2 , CO_2 , La_2O_3 , Pu_2O_3 , and PuO_2 were calculated; results are shown in Table 4.

Table 4. Gibbs Free Energy of Formation of Oxides
Involved in Haynes 188/Curium Fuel Reactions

Constituents	ΔG_f (kcal/mole)				
	600°C	800°C	1000°C	1200°C	1300°C
CoO	-39.46	-35.37	-31.02	-27.08	-24.96
Cr ₂ O ₃	-203.92	-189.81	-175.72	-161.67	-154.63
NiO	-39.06	-35.01	-31.01	-27.10	-25.15
WO ₂	-101.16	-93.85	-86.78	-79.96	-76.80
Fe ₃ O ₄	-199.79	-185.20	-170.88	-156.61	-149.47
MnO ₂	-76.82	-73.30	-69.76	-66.23	-64.27
SiO ₂	-172.92	-164.60	-156.33	-148.10	-144.00
CO ₂	-94.62	-94.66	-94.70	-94.73	-94.74
La ₂ O ₃	-373.84	-361.82	-349.69	-337.23	-331.04
Pu ₂ O ₃	-352.12	-339.32	-326.94	-314.98	-309.15
PuO ₂	-215.55	-207.05	-198.70	-190.53	-186.50

The free energies from Tables 3 and 4 were used to calculate the free energies for Eqs. 1-18. Results are shown in Tables 5 and 6.

Table 5. Free Energies of Reaction for Haynes 188/Cm₂O₃ Reactions

Equation	ΔG_r (cal/mole of Cm ₂ O ₃)				
	600°C	800°C	1000°C	1200°C	1300°C
1	238,500	238,620	240,320	241,240	242,380
3	152,170	154,530	157,300	160,440	162,380
5	242,460	243,540	244,880	246,390	247,270
7	207,920	207,910	207,940	300,030	207,840
9	218,220	220,020	222,040	224,430	225,780
11	146,340	149,790	153,710	158,020	160,930
13	106,720	109,620	112,870	116,480	118,410
15	224,380	214,800	205,560	196,910	192,710
17	13,780	21,140	29,020	37,660	42,080

Table 6. Free Energies of Reaction for Haynes 188/PuO₂ Reactions

Equation	ΔG_r (cal/mole of PuO ₂)				
	600°C	800°C	1000°C	1200°C	1300°C
2	20,490	20,600	20,800	20,750	20,780
4	6,110	6,590	6,960	7,280	7,470
6	21,210	21,420	21,560	21,610	21,620
8	15,450	15,480	15,400	15,210	15,060
10	17,170	17,500	17,750	17,950	18,050
12	5,190	5,800	6,370	6,880	7,230
14	-1,420	-900	-440	-40	150
16	18,200	16,610	15,020	13,360	12,540
18	-16,900	-15,650	-14,420	-13,180	-12,580

Since the free energies of Eq. 14 and Eq. 18 are negative, it appears that the silicon and lanthanum of the Haynes 188 alloy should react with the PuO₂ of the ²⁴⁴Cm₂O₃ product over the 600-1300°C temperature range. It should be noted that it was assumed there would be no compound formation between the components of the alloy or with the curium metal formed. Also no account was taken for possible oxygen solubility in the alloy. Experimental verification of the interaction of Haynes 188 alloy with the curium product is needed.

Reaction of Cm₂O₃ With Ta-10% W Alloy — The results of the calculations of the partial molar free energies of mixing for tantalum and tungsten are shown in Table 7. The composition of Ta-10% W is 90.14 atom % Ta and 9.86 atom % W.

Table 7. Partial Molar Free Energies of Mixing of Tungsten and Tantalum in Ta-10% W Alloy

Constituents	$-\Delta\bar{G}_M$ (cal/mole)				
	600°C	800°C	1000°C	1200°C	1300°C
Ta	170	220	260	300	320
W	3,960	4,880	5,800	6,720	7,180

The free energies of formation of Ta_2O_5 , WO_2 , Pu_2O_3 , and PuO_2 were calculated; results are shown in Table 8.

Table 8. Gibbs Free Energy of Formation of Oxides Involved in Ta-10% W/Curium Fuel Reactions

Compound	$-\Delta G_f$ (kcal/mole)				
	600°C	800°C	1000°C	1200°C	1300°C
Ta_2O_5	395.69	374.82	354.18	333.75	323.63
WO_2	101.16	93.85	86.78	79.96	76.80
$Pu_2O_3^a$	352.12	339.32	326.94	314.98	309.15
PuO_2	215.55	207.05	198.70	190.53	186.50

^aUsed as a stand-in for Cm_2O_3 .

The free energies from Tables 7 and 8 were used to calculate the free energies for Eqs. 19-22. Results are shown in Table 9.

Table 9. Free Energies of Reaction for Ta-10% W/Curium Fuel Reactions

Equation	ΔG_r (kcal/mole of Cm_2O_3 or PuO_2)				
	600°C	800°C	1000°C	1200°C	1300°C
19	114.91	114.96	114.74	115.09	115.35
20	-0.49	-0.54	-1.32	-2.76	-3.57
21	206.31	205.86	205.47	205.12	204.72
22	15.18	15.14	14.99	14.73	14.54

Since the free energies for Eq. 20 are negative it appears that the PuO_2 of the curium product will react with the tantalum of the Ta-10% W alloy over the 600-1300°C temperature range. It should be noted that it was assumed there would be no compound formation between the tantalum and tungsten of the alloy or between the tantalum and tungsten with the curium metal that might be formed. Also no account was taken for possible oxygen solubility in the alloy. Experimental verification of the interaction of Ta-10% W alloy with the curium product is needed.

Reaction of Cm_2O_3 With TZM Alloy — The partial molar free energies of mixing of the constituents in TZM (99.002 atom % Mo, 0.997 atom % Ti, and 0.0008 atom % Zr) were calculated. The results are shown in Table 10.

Table 10. Partial Molar Free Energies of Mixing of Constituents in TZM

Constituents	$-\Delta\bar{G}_M$ (cal/mole)				
	600°C	800°C	1000°C	1200°C	1300°C
Mo	16	20	24	28	30
Ti	7,870	9,710	11,540	13,370	14,290
Zr	20,280	24,930	29,580	34,320	36,550

The free energies of formation of MoO_2 , TiO , ZrO_2 , Pu_2O_3 , and PuO_2 were calculated; results are shown in Table 11.

Table 11. Gibbs Free Energy of Formation of Oxides Involved in TZM/Curium Fuel Reactions

Compound	$-\Delta G_f$ (kcal/mole)				
	600°C	800°C	1000°C	1200°C	1300°C
MoO_2	95.60	87.71	80.36	72.94	69.27
TiO	103.78	99.38	95.08	90.87	88.81
ZrO_2	221.71	212.82	203.85	194.96	190.66
Pu_2O_3^a	352.12	339.32	326.94	314.98	309.15
PuO_2	215.55	207.05	198.70	190.53	186.50

^a Pu_2O_3 used as a stand-in for Cm_2O_3 .

The free energies from Tables 10 and 11 were used to calculate the free energies for Eqs. 23-28. Results are shown in Table 12.

Table 12. Free Energies of Reaction
for TZM/Curium Fuel Reactions

Equation	ΔG_r (kcal/mole of Cm_2O_3 or PuO_2)				
	600°C	800°C	1000°C	1200°C	1300°C
23	208.84	207.49	206.44	205.62	205.29
24	15.59	15.41	15.15	14.81	14.63
25	66.42	70.54	76.32	81.89	85.59
26	-8.45	-7.45	-6.54	-5.71	-5.33
27	49.98	57.49	65.53	73.88	77.98
28	-10.87	-9.59	-8.33	-7.14	-6.59

Since the free energies for Eqs. 26 and 28 are negative, the PuO_2 of the $^{244}\text{Cm}_2\text{O}_3$ product would be expected to react with the titanium and zirconium in TZM alloy over the 600–1300°C temperature range. It should be repeated again that it was assumed there is no compound formation between the Mo, Ti, or Zr of the alloy or between them and the curium metal. Also, no account was taken for the oxygen solubility in the TZM. Experimental verification of the interaction of the product with TZM is needed.

Effect of Oxygen Solubility in Container Alloys on Interaction With Curium Oxide Heat Sources (*E. E. Ketchen*)

Thermodynamic calculations have been made to investigate the reaction of curium oxide heat source material (5% PuO_2) with a number of possible container alloys. Since the free energy of decomposition of PuO_2 to Pu_2O_3 and oxygen is less than the free energy of the decomposition of Cm_2O_3 to curium and oxygen, the PuO_2 decomposition process is the major process that would furnish oxygen to react with the container material. For example, all of the constituents of TZM (Ti, Zr, Mo) are expected to react with PuO_2 but not with Cm_2O_3 in the temperature range from 800 to 1300°C.

Haynes 188 alloy (41.87 atom % Co, 26.50 atom % Cr, 23.47 atom % Ni, 4.77 atom % W, 1.68 atom % Fe, 0.86 atom % Mn, 0.44 atom % Si, 0.41 atom % C, and 0.00004 atom % La) was found to be one of the alloys, by free energy calculations, that has low percentage components (Si and La) that should react with PuO_2 . The silicon and lanthanum should react with the PuO_2 if not protected by other elements. This reaction could occur if oxygen can be transmitted in the alloy to the lanthanum and silicon atoms. The solubility of oxygen and its diffusion rate in the alloy are the two factors that determine the speed of the reaction of oxygen with the low

percentage components. Generally it is assumed that the oxygen diffuses through the metal as a monoatomic gas.

When PuO_2 is heated it loses oxygen and passes through a series of intermediate compositions to Pu_2O_3 . The first oxygen is easiest to remove; at each succeeding composition the oxygen is more difficult to remove as the Pu_2O_3 composition is approached. This effect can be seen by comparing the equilibrium partial oxygen pressure over PuO_2 with the average equilibrium oxygen pressure for the decomposition of PuO_2 to Pu_2O_3 . The pressures, which are shown in Table 13, were calculated from data by F. L. Oetting³ and R. A. Kent.⁴ For example, the oxygen pressure at 1000°C in equilibrium with PuO_2 (9.42×10^{-19} atm) is much higher than the average decomposition pressure calculated from the free energy of reaction of PuO_2 to give Pu_2O_3 (6.40×10^{-25} atm). Also, it is evident from the low pressures in Table 13 that any mechanism by which a small quantity of oxygen is removed from the atmosphere above the PuO_2 helps to decompose the PuO_2 . The PuO_2 decomposition is aided by the oxygen solubility and oxygen diffusion in the container material.

Table 13. Equilibrium Oxygen Partial Pressures Above PuO_2 and Average Equilibrium Oxygen Pressure for PuO_2 Decomposition to Pu_2O_3

Temperature (°C)	Equilibrium Oxygen Pressure (atm)	
	Partial Above PuO_2	Average for PuO_2 to Pu_2O_3
600	1.88×10^{-41}	2.96×10^{-40}
800	5.56×10^{-28}	3.50×10^{-31}
1000	9.42×10^{-19}	6.40×10^{-25}
1200	4.93×10^{-12}	2.49×10^{-20}
1300	2.56×10^{-9}	1.81×10^{-18}

Since Table 13 shows that there is a marked difference between the equilibrium partial pressure of oxygen above PuO_2 and the pressure of oxygen due to the free energy change going from PuO_2 to Pu_2O_3 , it is evident that the calculations made in past reports on the free energy of reaction between the alloys and PuO_2 may be too conservative in predicting some reaction between PuO_2 and the alloy. However, since the initial equilibrium partial pressure above PuO_2 decreases so rapidly as it loses oxygen,⁴ the estimates are probably still good.

Even though the free energy for the reaction of PuO_2 with the container material is not favorable, the oxygen solubility and oxygen diffusion process can cause the reduction of plutonium oxide. That is, the equilibrium pressure of oxygen in the container above PuO_2 may be below the concentration of oxygen needed to react with the container material, but the oxygen solubility and oxygen diffusion through the container wall can result in some PuO_2 reduction.

In general, more information is needed on the solubility of oxygen in the container material. The reported solubility of oxygen in the alloys may not be the true oxygen solubility in the metal but a combination of the true solubility and the solubility of a metal oxide. These oxygen and metal oxide solubility phenomena have been reported in tantalum metal by S. Stecura.⁵ Actual tantalum oxides were identified in the material after the metal had been dissolved. This occurred both at the reported solubility limit and well below this limit.

In conclusion, the term solubility of oxygen in the alloy may not be well defined. Although it is implied that a maximum solubility of oxygen in the alloy has been found, this reported solubility may also include the oxygen in the metal oxide which is soluble in the alloy as well as the oxygen solubility alone. It would be difficult if not impossible to calculate the free energy change due to the oxygen solubility in the alloy.

Free Energy Calculations for the Reaction of Beryllium with Cm₂O₃ Heat Source Product (*E. E. Ketchen*)

The thermodynamics of the reaction of Cm₂O₃ with beryllium to form CmO or curium metal have been investigated to evaluate the possibility of the reaction of Cm₂O₃ with beryllium metal. Since the Cm₂O₃ heat source product will contain some PuO₂ at the time of fabrication, the reaction of beryllium with PuO₂ was investigated. The Cm₂O₃ heat source material may be in contact with beryllium in the capsule.

The free energies of reaction of Cm₂O₃ with beryllium as noted in Eqs. 29 and 30 were reported previously.⁶ Table 14 shows that the formation of curium metal is favored but CmO should not be formed. As noted previously⁶ the reaction should not be too great since the BeO or BeO·Cm₂O₃ should form a protective coating over the beryllium up to the melting point.

The free energies for the reaction of PuO₂ with beryllium (Eq. 31) were calculated and are shown in Table 14. Since the free energies are negative, it is evident that beryllium should react with the PuO₂. However, as mentioned before the BeO and BeO·Cm₂O₃ compounds should form a protective coating over the metal to prevent further reaction. Since the melting point of beryllium is 1283°C, the beryllium will be a liquid at 1300°C and probably would present more problems at this temperature.

Table 14. Gibbs Free Energy as a Function of Temperature

Temperature (°C)	ΔG (kcal/mole of Cm_2O_3 or PuO_2)		
	Eq. 29	Eq. 30	Eq. 31
600	143.98	-14.89	-21.68
800	131.98	-13.65	-21.44
1000	120.04	-12.21	-21.90
1200	108.17	-10.55	-21.72
1300	101.89	-10.07	-21.28

Data Needed for Calculation of Cm_2O_3 Free Energy (*E. E. Ketchen*)

Experiments will be conducted soon to measure the specific heat (C_p) of Cm_2O_3 as a function of temperature over the 400-1300°C range. Using these data and Eqs. 32 and 33 below, it should be possible to calculate the change in heat content and entropy for Cm_2O_3 over the 400-1300°C temperature range. It may be feasible to extrapolate the specific heat data to 298°K, thus permitting calculation of $H_T - H_{298^\circ\text{K}}$ and $S_T - S_{298^\circ\text{K}}$. It should be noted that these two functions are for Cm_2O_3 and not for the formation of Cm_2O_3 from the elements.

$$H_T - H_{673^\circ\text{K}} = \int_{673^\circ\text{K}}^T C_p \, dT \quad (32)$$

$$S_T - S_{673^\circ\text{K}} = \int_{673^\circ\text{K}}^T (C_p/T) \, dT \quad (33)$$

From the heat content and entropy it is possible to calculate the free energy of Cm_2O_3 using Eq. 34 for temperatures up to 1300°C.

$$F = H - TS \quad (34)$$

Again, this free energy will be for Cm_2O_3 , not the free energy of formation of Cm_2O_3 . If the free energy of curium were known, it could be used, along with the known free energy of oxygen, to calculate the free energy of formation of Cm_2O_3 . At present the experimental free energy functions $[(F_T - H_{298^\circ\text{K}})/T]$ are available for solid curium up to the melting point, but $H_{298^\circ\text{K}}$ is not known. When ^{248}Cm is available, the specific heat should be measured to as near 0°K as possible. These data could then be used to calculate $H_{298^\circ\text{K}}$ for curium and hence the free energy (F_T) for curium. This information would permit calculation of the free energy of formation for Cm_2O_3 over the desired range.

As data for the C_p of Cm_2O_3 become available, estimates of the free energy of formation for Cm_2O_3 can be made which are probably better than the

estimated values now used. In the event that Cm_2O_3 is shown to form only curium and oxygen, not CmO , it may be possible to calculate the free energy function $[(F_T - H_{298^\circ\text{K}})/T]$ for Cm_2O_3 . This function along with data now available would permit the calculation of the free energy function for the formation of Cm_2O_3 from its elements. The possibility of using this approach depends on the results of the vapor pressure/vapor species experiments now underway.

Summary of Thermodynamic Calculation (*E. E. Ketchen*)

During the past few months several alloys which have potential use for Cm_2O_3 encapsulation have been investigated from a thermodynamic viewpoint considering possible reactions between the fuel and the alloy. In all cases a reference fuel consisting of 95% Cm_2O_3 and 5% PuO_2 was assumed. The reactions considered were only those of the type

where

F = element in the fuel (Cm or Pu)

M = constituent in alloy

M_zO_y = most favorable oxide of M

The calculations did not consider possible solubility effects or other compound formations. Free energy data for Pu_2O_3 were used as stand-ins for those of Cm_2O_3 which are not available. For the nine alloys investigated, no reactions of Cm_2O_3 (or Pu_2O_3) with any constituents were predicted. Calculations for three alloys also indicated no reactions with PuO_2 ; two indicated reactions of PuO_2 with major constituents; and four indicated reactions of PuO_2 with minor constituents. A summary of the PuO_2 /alloy calculations is shown below:

<u>Alloy</u>	<u>Constituent(s) Predicted to React With PuO_2</u>
Pt-26% Rh-8% W	None
Pt-30% Rh-8% W	None
Ir-2% W	None
Hafn alloy 20-20	Hf (95.78 atom %)
Ta-10% W	Ta (90.14 atom %)
Haynes 188	Si (0.4 atom %) and La (4×10^{-5} atom %)
Type 316 stainless steel	Si (1.97 atom %)
T-111	Hf (2.03 atom %)
TZM	Ti (0.997 atom %) and Zr (8×10^{-4} atom %)

It should be noted that these calculations are limited in scope and that they are estimates at best. However, taken at face value, they seem to indicate that the plutonium oxides present in the fuel may influence the compatibility situation more than the curium oxide does.

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

Helium Release (*P. Angelini*)

Three 0.46-cm-diam by 0.14-cm-thick $^{244}\text{Cm}_2\text{O}_3$ pellets were hot-pressed using powder from batch SRL-31, and the residual carbon from the die body was removed by oxidation using the procedure which maintains the $^{244}\text{Cm}_2\text{O}_3$ form. The pellets were then transferred, in an inert atmosphere container, to the balance glove box for mass and dimensional measurements.

The pellets were loaded into separate sample containers, and each container was placed in its respective furnace. The furnace temperatures were then increased to the predetermined values shown in Fig. 3; the initial heat-up helium release curves are shown in Fig. 4. In each case there was an extremely large amount of helium evolved during the initial evacuation of the sample tube. This helium, which could not be quantitatively measured, is probably that which is very close to the surface of the releasing unit or that which is held in open microcracks or grains.

Fig. 3. Temperature Versus Time for Helium Release Tests.

Fig. 4. Helium Release Rate Versus Time (0-152 hours).

The release characteristics of samples A and C during their initial temperature increase were very similar. The release rate measured from sample B was different; it increased to larger values above 750°C after decreasing from its first burst at a temperature. This second burst could be due to the annealing of radiation damage in the fuel. Thus, the microstructure of the fuel pellet maintained at 800°C could be very different from that of the pellets maintained at 500 or 650°C. Also if this theory is true, the data reported previously in the helium release report on $^{244}\text{Cm}_2\text{O}_3$ could be extrapolated to a minimum temperature of 750°C. Below this temperature another mechanism would predominate.

The plots of helium release rate versus time for samples A (650°C), B (800°C), and C (500°C) presented in Fig. 5 are based on experimental points which are average values of release rates. Individual points represent integrated release rates based on incremental measuring times ranging from 5 min to 3 days. Data are usually taken on a particular sample during a 1-day period every 3 days. During each incremental measuring time the release rate is recorded then integrated such that the amount of helium emitted over that incremental time is obtained. The abscissa of Fig. 5 represents the time from initial heating of the sample to the end of the incremental measuring time for the release rate.

The release rate curve for sample A (650°C) was proceeding as expected up to 332 hr; during this period the burst-type helium emission was increasing. Beyond 332 hr the burst-type helium emission increased tremendously. During the time period from 400 to 730 hr the sample was emitting above its production rate. Thus, the inventory accumulated during the first 400 hr at temperature was emitted during the 400- to 730-hr period. Since the areas of the two regions are approximately equal, a very small helium inventory should remain in the sample. The data to be obtained after the 730-hr period will show whether this phenomenon is cyclic or whether the release rate will level off, as expected, at the helium production rate for the sample. The sample will remain at this temperature for ~500 additional hours. If the helium release rate is equal to the production rate during this time the fuel pellet will be removed under inert atmosphere and observed. Part of the pellet will be sent for optical microscopy and another part studied by x-ray diffraction. The majority of the sample will be returned to the furnace, and the experiment continued for an additional 200 hr, after which the experiment will be terminated.

Sample C (500°C) has undergone the same phenomenon as sample A; however, sample B (800°C) has not undergone this phenomenon. Since the helium release rates for this sample have changed very little in 750 hr, equilibrium was probably reached in a short period. The experiment will be continued to prove this. Part of this pellet will also be studied by optical microscopy and x-ray diffraction analysis such that the differences between this pellet and sample A can be recorded.

The release of stored helium at 500 and 650°C may be caused by alpha particles destroying the releasing units in the fuel. At these temperatures the damage caused by the alpha particles may be cumulative until the releasing unit can no longer accept any further damage and begins to be destroyed. The final helium inventory measurement at the completion of the long-term test should determine the amount of damage, and the studies by optical microscopy and by x-ray diffraction analysis should determine the type of damage which occurred. At a temperature of 800°C, radiation damage is annealed to some equilibrium level and the releasing unit is not destroyed, and release by this mechanism does not occur.

Fig. 5. Helium Release Rates Versus Time (0-900 hr).

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

The vapor pressure collection assembly was used in measuring the vapor pressure of curium metal. This provided a test to check both the unit and the range of agreement when an experiment was being performed on a radioactive element as reported in the literature. The equipment and procedure used by Smith *et al.*⁷⁻⁸ in performing their experiments on ^{244}Cm metal were the same as those of Smith and Peterson⁸ in their experiments on $^{244}\text{Cm}_2\text{O}_3$.

In our experiments a tungsten Knudsen cell was used. The temperature was measured by using a disappearing filament pyrometer. The black-body hole was located at the base of the Knudsen cell. The temperature values were corrected for pyrometer error, furnace window, and glove box window effects according to a calibration of the pyrometer and glass performed by the Standards Group of the Instrumentation and Control Division. The experiment was performed in the temperature range from 900 to 1600°C. The vapor pressure collection assembly remained cool during the experiment. The rotary seal worked very well, the turning was smooth and could be performed easily. A technique for handling the platinum metal sample collection discs was worked out, and the radiation background was kept to a very minimum. The individual collection discs were counted with an alpha spectrometer by personnel in the Analytical Chemistry Division. The least square curve with corresponding standard error of coefficients for the vapor pressure of ^{244}Cm metal is

$$\log_{10} P = (3.834 \pm 0.411) - (17,939 \pm 641)/T$$

where

P = vapor pressure in atmospheres
T = absolute temperature in Kelvin

The vapor species is assumed to be ^{244}Cm . The vapor pressure as reported above is lower than the vapor pressure obtained by Smith *et al.*⁷ for the vapor pressure of curium. In the temperature region of 1100°C the difference could be explained by experimental temperature differences of ~30°C. Accurate temperature measurements and uniform Knudsen cell temperature are important factors in obtaining accurate vapor pressure data. The design of the furnace heat zone and Knudsen cell temperature measurement capability in our vapor pressure furnace is such that a minimum correction factor need be applied to the actual measurements.

Vapor pressure data are required for the prediction of the volatile migration of Cm_2O_3 in heat sources. Smith and Peterson⁸ have determined the vapor pressure of Cm_2O_3 in the range of 1570-2336°C; however, current interest is in the range of about 800-1400°C. The use of their data in the lower range involves a long extrapolation. Furthermore, the crystalline form of Cm_2O_3 changes at 1600°C, which is at the lower edge of their range of measurement. The slope of the pressure-versus-temperature curve will be influenced to some degree by this transition.

The Knudsen cell method will be used for the greater part of our measurements. Later, the Langmuir method will be used for the measurements at the lowest temperatures. This work is not a routine application of these methods of pressure measurement. The Knudsen cell method is used ordinarily at pressures down to 10^{-9} atm and the Langmuir method is used at pressures as low as 10^{-11} atm. We hope to go to 10^{-14} atm at 1100°C . Extrapolation of the data of Smith and Peterson⁸ indicates that below about 1100°C the pressure will be too low for determination.

The optical pyrometer system used for temperature measurement required calibration because the light used for measurement passes through two pieces of glass: the furnace window and the glove box window. The effect of the resulting light absorption on the pyrometer reading was measured by the Instrument Standards Group of ORNL using pieces of glass identical to the two windows. The pyrometer was also checked against a Pt-Pt-10% Rh thermocouple in the base of the Knudsen cell. The two groups of data were in good agreement. (The thermocouple vaporized during this work. It can be replaced if measurements below the optical pyrometer range are needed.)

In our application of the Knudsen cell method, the rate at which material leaves the orifice of the Knudsen cell is determined as follows: 1) Small stainless steel targets are mounted in a holder at the top of the vacuum furnace. The holder can be rotated to bring any one of ten targets over a hole leading into the furnace. 2) When in position, the target is struck by a known fraction of the total molecular beam of curium oxide leaving the Knudsen cell. 3) The amount of curium deposited on the targets is determined by an alpha-counting procedure.

It cannot be assumed with certainty that all molecules which strike the surface stick. In some similar reported cases, the condensation coefficient is much less than one.^{9,10} (The condensation coefficient is the fraction of the molecules striking the surface which stick.) The size of the condensation coefficient is strongly influenced by the nature of the surface. High values occur when the condensing molecules readily fit into the crystalline lattice of the surface.

A series of targets with differing surfaces has been exposed in the vapor pressure apparatus. The surfaces were stainless steel, oxidized stainless steel, Nd_2O_3 deposited on stainless steel, lead oxide on stainless steel, and platinum. The curium deposits on these targets will be determined by alpha counting. A statistically significant variation in the observed rates of deposit will indicate the need of further investigation.

The problem of error because of accidental contamination of the targets is serious. The deposits on the targets are of the order of only 10^{-10} g. Deposits of this size can be accurately measured by alpha counting, but an extremely small amount of contamination will cause a significant error. Furthermore, the targets are used in equipment which has been previously exposed to curium.

The first targets exposed were leached by acid, and measurements were made of the total curium in the resulting solutions. The resulting data were badly scattered and were higher than expected. In two cases control targets which had not been exposed to the molecular beam showed greater curium deposits than any of the targets which had been exposed. The problem has been largely solved by counting the deposits while on the target and by the use of an appropriate masking procedure during the counting. Only the part of the surface which was exposed to the molecular beam is exposed during the counting. In one series of measurements with Cm_2O_3 , six stainless steel targets were exposed at Knudsen cell temperatures which varied from 1202 to 1587°C. These targets were counted using the mask. The logarithms of the vapor pressures calculated from these targets were plotted as a function of the reciprocal of the absolute temperature. An excellent straight line was obtained with five of the six points. The other point was high. The target on which this point was based showed appreciable contamination on the unexposed surfaces. It is likely that some contamination was also present on the exposed surface.

Numerical values will not be reported until questions of systematic error are resolved.

X-Ray Diffraction (*P. Angelini*)

The goniometer was realigned with the graphite crystal monochromator in place. The monochromator introduces a small loss in signal strength but increases the signal-to-noise ratio. After the final alignment procedure, scans of silicon and gold were obtained for use as standards under identical experimental settings for future experiments.

In October an x-ray diffraction scan was made on some pieces of a pellet which had been used in a seawater leach test, samples of which were scanned by x ray in August, and stored in a seawater-filled container. The material was removed from the container, prepared on its mount under argon, and immediately inserted into the x-ray diffraction assembly. The system was evacuated and the scan was made. The original scan on this material had shown two patterns, a monoclinic pattern, probably Cm_2O_3 , and a more intense fcc pattern with a cell parameter of $a = 5.369 \text{ \AA}$. In the scan made after two months' storage in seawater, only an fcc pattern was seen; the calculated cell parameter was $a = 5.391 \text{ \AA}$. Also, the peaks were broader and less intense than had been the case in the first scan. Comparison of the two scans indicates that additional oxidation had occurred while the material was in storage in seawater, since the monoclinic pattern was not present in the second scan. The broader peaks in the second scan probably indicate additional radiation damage; and, the much larger fcc cell parameter of the second scan indicates that the sample had also hydrated.

A pellet which had been exposed to dry air for approximately four months was transferred to the x-ray diffraction process glove box. The pellet was crushed and a scan of this sample was performed under an air atmosphere. Only an fcc pattern was obtained. The calculated lattice parameter was $a = 5.377 \text{ \AA}$. This lattice parameter is larger than the lattice parameter of freshly prepared and annealed CmO_2 sample ($a = 5.369 \text{ \AA}$).

A monoclinic pattern was not observed. Thus the original Cm_2O_3 pellet had oxidized to CmO_2 in the length of time that it had been exposed to air. The larger lattice parameter and broad peaks indicate radiation damage to the crystal structure.

Some of the remaining solid matter from the first boiling distilled water solubility experiment pellet was transferred to the x-ray diffraction system, and the sample was mounted under an argon atmosphere. The material seemed to be both in solution and in suspension in the distilled water. A scan was performed under vacuum immediately after the mount was placed in the x-ray system. Two patterns were observed: a very strong fcc pattern and a strong hexagonal pattern. The fcc pattern had a calculated lattice parameter of $a = 5.398 \text{ \AA}$. The calculated lattice parameter of the hexagonal pattern was $a = 6.396 \text{ \AA}$ and $c = 3.691 \text{ \AA}$. The peaks were rather sharp and intense in both patterns. The fcc pattern is probably a hydrated form of CmO_2 and the hexagonal pattern is attributed to $\text{Cm}(\text{OH})_3$. The lattice parameter for the hexagonal structure calculated from the data is very similar to the lattice parameter of various lanthanide hydroxides. Also the intensity and the number and position of the peaks reinforce the conclusion that the pattern was that of $\text{Cm}(\text{OH})_3$. This was the only sample of the leach test materials in which the hydroxide compound was identified.

Part of a pellet from the first set of pellets scheduled for use in the vapor pressure experiment was transferred to the x-ray diffraction process glove box (these fuel pellets were made from the purified curium oxide powder). A scan was performed over $22^\circ\text{--}85^\circ 2\theta$, and the lines recorded showed the material to have two phases; both a monoclinic and an fcc structure were observed. The monoclinic structure was attributed to Cm_2O_3 monoclinic form and the fcc structure to a slightly reduced CmO_2 form. Since the presence of CmO_2 showed that some oxidation of the Cm_2O_3 had occurred, the pellets were rejected, and a second calcination/pressing cycle using the purified material was made. A scan over $10^\circ\text{--}85^\circ 2\theta$ was performed on a sample soon after the calcining procedure. Very strong lines were present, all of which were attributed to the cubic C form of Cm_2O_3 . The material should be in the C form since the oxide had been taken to 1000°C in vacuum. Pellets were hot-pressed at 1450°C from the batch of calcined powder. A scan performed on one of the as-pressed pellets showed that the material had a monoclinic structure, with one line at $26.38^\circ 2\theta$ attributable to graphite. Another pellet was processed through the carbon burn-off procedure; a scan of samples from this pellet showed that this sample also had a monoclinic form. There was one unaccounted for line at $20.86^\circ 2\theta$; possibly some impurity oxidized during the carbon burn-off. The $26.38^\circ 2\theta$ line was not detected, showing that carbon surface deposits had been burnt off the pellet. The lines of the scan on the pellet which had been through the carbon burn-off process were less intense than the lines of the scan on the as-pressed pellet. The monoclinic structures of both the pellets are attributable to the B form of Cm_2O_3 , which forms irreversibly at the hot-pressing temperature. The lower intensity of the lines in the scan of the pellet which had received the carbon burn-off treatment indicate a distortion of the crystal lattice relative to the as-pressed pellet. Both pellets were pressed at the same

time, but the as-pressed pellet scan was made two days before the scan of the other pellet after the carbon removal procedure. The crystal distortion probably resulted from radiation damage, possibly influenced by helium effects rather than from the carbon burn-off process itself.

The results of these measurements indicate that the procedure used for pellet fabrication and carbon removal result in the desired fuel form. They also demonstrate the value of x-ray diffraction analysis as a tool for identification of structure; this is useful for proper description of processes occurring in various experiments. The CmO_2 indicated by the scan of the material which was rejected probably resulted from some exposure to air during the post-pressing handling or transfer operations. This again points up the necessity of exclusion of oxygen in Cm_2O_3 handling operations.

Emissivity (*P. Angelini*)

The new furnace heat shields were fabricated and installed in the furnace which will be used for the emittance experiment. A graphite pellet was used for initial calibration experiments of the spectroradiometer. The new heat shield enabled the attainment of 1375°C as the maximum sample temperature at maximum furnace temperature.

The test of the spectroradiometer with its readout instruments demonstrated satisfactory operation of the unit. The mechanical chopping frequency was changed from 16 to 2.42 hz, and the lock-in amplifier was used in its tuned circuit mode. These two changes have greatly enhanced the signal-to-noise ratio from the instrument. Excellent readings could be obtained from the system with sample temperatures from 700 to 1375°C and spectral range of 3.0, 3.5, 4.0, 4.5, 5.0, and 5.5 microns in addition to the total radiation receiving mode.

Heat Capacity (*E. Angelini*)

The system has been checked. Final preparations are being made before the cell becomes radioactive.

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

Observations of the three 25-W test pellets made in September continued. The two 25-W Cm_2O_3 pellets — one of purified Cm_2O_3 and the other of production-grade material — which had been stored under argon at $\sim 650^\circ\text{C}$ were checked on November 20. The pellet of purified Cm_2O_3 (PCM-P2) was intact and no dimensional changes had occurred; this pellet was fabricated on September 18. Inspection of the pellet of production-grade Cm_2O_3 (batch SRL-31) revealed that one large piece weighing 0.71 g and several small chips also totaling 0.71 g had broken off from one end of the pellet. This pellet was fabricated on September 27 and had last been inspected on October 30, at which time it was still intact. The pellet

of purified Cm_2O_3 (PCM-P1) which had been stored in air was also inspected; there was no significant change. During the two-month storage of PCM-P1 in dry air it lost a total of 10.7% of its original weight. Essentially all of this loss occurred during the first week, with the first observable loss occurring 70 hr after the start of the air exposure. The loss was in the form of small chips flaking from the edges of the pellet ends. The body of the pellet has shown no evidence of swelling.

Based on the observations made on these three pellets, it would appear that $^{244}\text{Cm}_2\text{O}_3$ pellets of this size can be stored at ambient temperature ($\sim 650^\circ\text{C}$ average surface temperature) in argon for several weeks without significant physical change. However, exposure to air should be prevented or held to a minimum to preclude deterioration of the pellet. The break-age of the SRL-31 pellet was probably caused by mechanical or thermal stress since the container in which it is stored has to be handled frequently.

Behavior of Hot-Pressed Cm_2O_3 Pellet in Boiling Distilled Water (*J. C. Posey*)

A test was made of the effect of boiling distilled water on a hot-pressed Cm_2O_3 pellet containing 13.1 wt % of $^{240}\text{PuO}_2$. The procedure was the same as that used with boiling seawater. The pellet was held in a basket of platinum gauze (370- μm holes) immersed in 175-ml of water contained in a Pyrex glass flask. The pellet density was 7.8 g/cm^3 and its area was 0.561 cm^2 .

Samples of the water were taken in pairs from time to time and analyzed for ^{244}Cm . One sample was withdrawn through a sintered glass filter stick; the other was not filtered. At the end of the test the heat was turned off, boiling ceased, and all suspended material was allowed to settle for 24 hr. The water was again sampled.

After about 8 hr the pellet started to disintegrate slowly. At first most of the material was present as large pieces which broke into progressively smaller and smaller pieces until at 144 hr all of the material had sifted through the 370- μm holes in the basket. At the end of the test, the Cm_2O_3 was present as a powder, partly on the bottom of the flask and partly suspended in the water.

The schedule of the disintegration of the pellet is given in Table 15. The concentrations of curium in the water illustrate the progressive break-up of the pellet. The curium concentrations in the unfiltered samples are dependent on the number of particles small enough to remain in suspension under the conditions of mild agitation as caused by the boiling water. The concentrations in the filtered samples are dependent on the number of particles capable of passing through the filter. The observation that both values increased with time indicates an increase in the amount of curium in the form of very small particles.

Table 15. Disintegration of Cm_2O_3 Pellet in Boiling Distilled Water

Time From Beginning Test (hr)	Observations	^{244}Cm Concentration ($\mu\text{g}/\text{ml}$)	
		Filtered	Unfiltered
1	Pellet intact; water clear	0.037	0.060
4	Pellet intact; water clear	0.044	0.076
8.4	Pellet intact; one particle at bottom of flask; water clear	—	—
24	Large irregular pieces in basket; many particles at bottom of flask; water clear	0.24	1.2
48	Less material in basket; water murky	13	28
76	One very small piece remaining in basket; water very murky	28	54
144	No material in basket; water very murky	32	59

The concentrations observed after 24 hr settling at room temperature were $8.7 \mu\text{g}/\text{ml}$ for the filtered sample and $7.8 \mu\text{g}/\text{ml}$ for the unfiltered sample. These values are much lower than the final values for the boiling solution. The solution was clear at this time.

The disintegration of the pellet was expected. Similar disintegration had been observed with air-saturated distilled water at room temperature although a much longer time, 300–400 hr, passed before visible damage took place.⁶ Furthermore, Fullam and Roberts¹¹ had observed a similar disintegration of pellets of Nd_2O_3 , Pm_2O_3 , and Sm_2O_3 in water. They showed that a hydration reaction occurred and that a hydrated oxide or hydroxide was produced. Because of the similarity of Cm_2O_3 to these oxides, it is likely that the reaction of Cm_2O_3 was the same. Free atmospheric oxygen is not present in boiling water, but some exposure to oxygen occurred afterward. In some of the powder examined using x-ray diffraction the presence of $\text{Cm}(\text{OH})_3$ and hydrated CmO_2 were indicated. This work is described in greater detail in the "X-Ray Diffraction" section of this report.

To date, tests have been carried out using flowing air-saturated seawater and distilled water at room temperature and using boiling seawater and distilled water. All of these tests used Cm_2O_3 pellets of comparatively low density and high ^{240}Pu content; tests with low ^{240}Pu content and higher density are planned.

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

The gamma spectrum of the 69.1-W source was rechecked and the data evaluated. The source will be defueled early in 1974. A report on the radiation measurements from the source was drafted; the final report should be completed in January.

Curium From Power Reactor Fuels (*T. A. Butler*)

The variations in isotopic composition of curium recovered from power reactor fuels was discussed in a previous report.¹ Calculations and one set of experimental data indicated that significant increases in the abundance of the fissile nuclei, ^{243}Cm and ^{245}Cm , can be anticipated relative to the product obtained from irradiation of plutonium targets in the Savannah River Plant reactors. Additional experimental evidence was found¹² which supports the previous discussion. In this case, UO_2 fuel rods were irradiated in the Vallecitos Boiling Water Reactor to ~ 10 GWD/MT(U) and then transferred to the Dresden boiling water reactor for additional irradiation. Curium isotopic contents of full-cross-section fuel rod samples were measured by mass spectrometry. The data are given in Table 16 with values normalized to a decay period of five years after fuel discharge.

Table 16. Isotopic Abundance of Curium Recovered From Boiling Water Reactor Fuel

Sample Number	^{235}U Enrichment (atom %)	Burnup [GWD/MT(U)]	Isotopic Abundance (atom %)			
			^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm
68	3.53	28.5	2.27	92.96	4.26	0.52
69	2.79	25.3	2.55	93.57	3.42	0.46
70	2.79	25.3	2.28	93.53	3.76	0.43

The calculated production of ^{244}Cm in enriched-uranium-fueled PWR's as functions of burnup and ^{235}U enrichment has been reported.^{1,6} The calculated yields for a 3.3% ^{235}U fuel enrichment given in Table 40 of reference 1 are in rather good agreement with measured data¹³ of ^{244}Cm yields obtained from analyses of Yankee-Rowe reactor fuels. The Yankee-Rowe is a commercial PWR and the ^{235}U fuel enrichment was 3.4% for the Core I from which the ^{244}Cm yield data were obtained as a function of burnup. The two sets of data are shown in Fig. 6.

The information obtained to date on the yield of curium isotopes and their isotopic abundances in power reactor fuels is summarized and interpreted, particularly with respect to the power density of $^{244}\text{Cm}_2\text{O}_3$ fuels, in the following discussion.

Few experimental data have been reported on the curium content of spent power reactor fuels and even less on the isotopic abundances. However, the available data are in sufficiently close agreement with calculated values derived from assumed average operating conditions of reference reactor types to allow predictions by calculations to be made with reasonable confidence. Reference reactor models include the light-water moderated reactor (LWR), high-temperature, gas-cooled graphite moderated reactor (HTGR), and liquid-metal-cooled fast breeder reactor (LMFBR). The LWRs are of two types: the boiling water reactor (BWR) and pressurized water reactor (PWR).

Fig. 6. Comparison of Calculated and Measured Yield of ^{244}Cm in Enriched-Uranium-Fueled PWR.

A projection of the growth of the nuclear power industry in the United States¹⁴ indicates that the LWRs will predominate at least through 1985; therefore, the power density of $^{244}\text{Cm}_2\text{O}_3$ fuels available from reprocessing of these reactor fuels is of most practical interest. The characteristics of curium recovered from BWR fuel are expected to closely approximate those from PWR fuel based on equal burnup of ^{235}U -enriched uranium and are not considered separately. However, beginning about the end of this decade, some of the PWRs are expected to be fueled with recycled plutonium. The effect of plutonium recycle is to increase the yield (about 50 fold) of curium and to reduce the abundance of the ^{244}Cm isotope.

Predictions of the isotopic composition of $^{244}\text{Cm}_2\text{O}_3$ fuels obtained from commercial reprocessing of fuels from various power reactors are speculative at this time. Several factors contribute to the uncertainty. It is assumed that high-level waste streams which contain the curium fraction will be accumulated as solutions in common waste storage tanks for periods of three to six years before curium recovery operations begin. Thus, curium originating in reactor fuels of differing composition, reactor flux spectra, and burnup will have an opportunity to homogenize during storage and average the differences in isotopic composition. The recent trend toward standardization of reactor designs in a mature nuclear power industry will make future predictions of curium isotopic composition more certain.

Using the predicted nuclear power growth,¹⁴ it can be assumed that the majority of reactor fuels for reprocessing will derive from PWRs fueled with enriched uranium, with gradually increasing numbers being fueled by plutonium recycle. It is further assumed that chemical recovery and purification of curium will be done after a decay period of four years to reduce the ^{242}Cm to negligible amounts, and that $^{244}\text{Cm}_2\text{O}_3$ fuels will be fabricated one year after the chemical recovery. Chemical processing removes essentially all of the plutonium residual and decay product but leaves an estimated maximum 3 wt % of other contaminants.

The isotopic composition of curium in enriched-uranium PWR fuels varies somewhat depending on the amount of ^{235}U enrichment and the burnup level attained. During power reactor start-up periods, lasting for two to three years, portions of the reactor cores are discharged with relatively low burnup values. The calculated effects of ^{235}U enrichment and burnup are shown in Tables 17 and 18.

Table 17. Influence of ^{235}U Enrichment on Isotopic Abundance of ^{244}Cm at 33 GWD/MTU and Four-Year Decay Period

Enrichment (Weight %)	Isotopic Abundance of ^{244}Cm (Weight %)
1.5	89.76
2.5	91.09
3.3	92.08

Table 18. Influence of Burnup on Isotopic Abundance of ^{244}Cm at 3.3% ^{235}U Reactor Fuel Enrichment and Four-Year Decay Period

Burnup (GWD/MTU)	Isotopic Abundance of ^{244}Cm (Weight %)
3.3	98.18
16.5	95.41
33.0	92.08

The information in Tables 17 and 18 shows that the ^{244}Cm content of the curium fraction increases with increased enrichment of the reactor fuel and decreases with increased burnup. These two effects tend to be compensating if it is assumed that waste streams from the reprocessing of a variety of reactor fuels are mixed in aqueous waste storage tanks. Most PWRs are expected to be fueled with ~3% ^{235}U enriched uranium,¹⁴ therefore, it is recommended that 92.08 wt % isotopic abundance of ^{244}Cm at a four-year decay period be used as a most probable value. The corresponding value for a PWR fueled with recycled plutonium and burnup of 33 GWD/MTU is 86.92 wt % ^{244}Cm at the four-year decay period.

In calculation of the probable power density of $^{244}\text{Cm}_2\text{O}_3$ fuels obtained from power reactor operations, previous assumptions must be borne in mind: the ^{242}Cm content of the curium fraction is decayed to negligible amounts, 3 wt % of impurity oxides are present in the fuel but plutonium has been removed at the end of a four-year decay period, fuel form fabrication is done one year after chemical recovery, the other principal isotopes of curium (^{243}Cm , ^{245}Cm , and ^{246}Cm) do not contribute significantly to the power of the fuel, and fuel density of 10.3 g/cm³ is attainable in hot-pressed pellets. The specific power of the $^{244}\text{Cm}_2\text{O}_3$ fuel will decrease by 3.74% during the holding period of one year between chemical processing and fuel fabrication due to decay of ^{244}Cm to ^{240}Pu . Mass loss due to the escape of helium is negligible in this length of time. The power density of the two sources of $^{244}\text{Cm}_2\text{O}_3$ isotopic power fuels is summarized in Table 19.

Table 19. Power Density of $^{244}\text{Cm}_2\text{O}_3$ Fuel Forms

Source of Curium-244	Specific Power (W/g) ^a	Power Density (W/cm ³) ^a
PWR, ^{235}U enriched	2.215	22.8
PWR, Pu recycle	2.09	21.5

^aAt five-year decay after discharge of the reactor fuel, density of 10.3 g/cm³.

The calculations given in Table 19 indicate that $^{244}\text{Cm}_2\text{O}_3$ fuels used for heat sources about 1980 should approximate fuel power densities of 22.8 W/cm^3 and decrease to a probable limit of 21.5 W/cm^3 around 1990 as plutonium recycle fuels for the PWR come into greater use.

Criticality of $^{244}\text{Cm}_2\text{O}_3$ Fuels (*T. A. Butler*)

Criticality of the actinide elements has been reviewed by Clayton and Bierman.¹⁵ Of the curium nuclei which are present in power reactor fuels, those containing an odd number of neutrons, ^{243}Cm , ^{245}Cm , and ^{247}Cm , are classified as fissile and those containing an even number of neutrons, ^{244}Cm , ^{246}Cm , and ^{248}Cm , are classified as fissionable. Fissionable nuclei are those for which a chain reaction is possible with most of the fissions caused by high-energy neutrons. Fissile nuclei are those for which a chain reaction is possible with the fissions caused by slow, or thermal, neutrons. The only data on critical experiments with curium nuclei is a material-replacement experiment with ^{244}Cm reported by Barton.¹⁶ He estimated that the critical mass of a bare metal sphere of ^{244}Cm (density 13.5 g/cm^3) is $27.7 \pm 2.5 \text{ kg}$. Clayton and Bierman¹⁵ calculated the critical mass of a similar sphere of ^{244}Cm to be 23.2 kg .

Preliminary calculations of the criticality of four representative heat source systems indicate that there should be no problem with these systems even if the $^{244}\text{Cm}_2\text{O}_3$ contains 12% of the fissile isotopes ^{243}Cm plus ^{245}Cm . The study will be completed in January 1974 and a detailed report made.

REFERENCES

1. Eugene Lamb, *ORNL Isotopic Power Fuels Quarterly Report for Period Ending September 30, 1973*, ORNL-4932, Oak Ridge National Laboratory.
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).
4. R. A. Kent, *Thermodynamic Analyses of MHW Space Electric Power Generator*, LA-5202MS, Los Alamos Scientific Laboratory (March 1973).
5. S. Stecura, *Observation of Oxide Particles Below the Apparent Oxygen Solubility Limit in Tantalum*, NASA TMX-2889, Lewis Research Center (September 1973).
6. Eugene Lamb, *ORNL Isotopic Power Fuels Quarterly Report for Period Ending June 30, 1973*, ORNL-4917, Oak Ridge National Laboratory.

7. P. K. Smith, W. H. Hale, and M. C. Thompson, "Vapor Pressure and Crystal Structure of Curium Metal," *J. of Chem. Physics* 30, 5066-76 (1969).
8. P. K. Smith and D. E. Peterson, "High-Temperature Evaporation and Thermodynamic Properties of Cm_2O_3 ," *J. Chem. Phys.* 52(10), 4963-72 (May 1970).
9. John L. Margrave, *Physiochemical Measurements at High Temperatures*, J. O'M. Bockris, J. L. White, and J. D. McKenzie (eds.), Butterworths Scientific Publications, London, 1959, pp. 230-31.
10. Saul Dushman and J. M. Lafferty, *Scientific Foundations of Vacuum Technique*, 2nd Ed., John Wiley & Sons, Inc., New York, 1962, pp. 408-10.
11. H. T. Fullman and F. P. Roberts, *Reactions of the Sesquioxides of Pm, Nd, and Sm With Water*, BNWL-1421, Pacific Northwest Laboratories (June 1970).
12. F. H. Megerth, Compiler, *Zircaloy-Clad UO_2 Fuel Rod Evaluation Program Quarterly Progress Report No. 9*, GEAP-10160, General Electric Company (February 1970).
13. R. P. Matsen, *An Analysis of Yankee-Rowe Burnup Data*, BNWL-1122, Pacific Northwest Laboratory (July 1969).
14. *The Growth of Nuclear Power 1972-1985*, WASH-1139 (Rev. 1), December 1971.
15. E. D. Clayton and S. R. Bierman, "Criticality Problems of Actinide Elements," *Actinides Reviews* 1, 409-32 (1971).
16. D. M. Barton, "Central Reactivity Contributions of ^{244}Cm , ^{239}Pu , and ^{235}U in a Bare Critical Assembly of Plutonium Metal," *Nucl. Sci. Eng.* 33, 51 (1968).

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