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YIELD OF CURIUM-244 IN THE SRP HIGH FLUX REACTOR

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by

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ABSTRACT

Comprehensive destructive analysis of a ^{242}Pu target slug that was irradiated to 3.7×10^{22} n/cm^2 in the Savannah River Plant high flux reactor confirmed the predicted curium yield. The measured curium yield was 0.276 g for each gram of ^{242}Pu irradiated. Curium yield was determined by counting the gross alpha activity in solutions of dissolved wafers cut from the target slug (alpha pulse height analysis verified that the alpha activity was >>99% ^{244}Cm). The ^{243}Am (0.136 g for each gram of ^{242}Pu irradiated) and residual ^{242}Pu (0.511 g for each gram irradiated) were determined by mass spectrometry using the isotope dilution technique. Overall mass balance for the target slug was obtained to within 1%.

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INTRODUCTION

A total of 520 g of ^{242}Pu was irradiated at high flux in a Savannah River reactor[1] to provide the High Flux Isotopes Reactor (HFIR) at Oak Ridge with target material of higher mass.

To corroborate the curium yield under irradiation at a flux comparable to that planned for the production of 4.5 kg of ^{244}Cm,[2] one target slug of Savannah River Plant (SRP) design, initially containing about 34 g of ^{242}Pu, was analyzed after an exposure of 3.7×10^{22} n/cm^2. This report describes the analytical methods used to assay the SRP target slug and summarizes the results of the analyses.

SUMMARY

Chemical analysis of a $^{242}PuO_2$-Al target slug confirmed the calculated production of ^{244}Cm during 20 cycles of irradiation (3.7×10^{22} n/cm^2) in the high flux charges. The measured yield of ^{244}Cm was 0.276 g for each gram of ^{242}Pu irradiated; the predicted yield was 0.255 g of ^{244}Cm per gram of ^{242}Pu.

Curium, americium, and residual plutonium were determined in solutions of dissolved wafers cut from the target slug. Curium was determined by counting the alpha activity in the dissolver solution. Pulse height analysis verified that the alpha activity was >>99% ^{244}Cm. The ^{243}Am (0.136 g per gram of ^{242}Pu irradiated) and residual ^{242}Pu (0.511 g per gram of ^{242}Pu irradiated) were determined by mass spectrometry using the isotope dilution technique.

Overall mass balance for the actinide target was obtained to within 1%. Americium and curium represented ~41% of the starting ^{242}Pu; fission products, ~9%; and residual ^{242}Pu, ~51%. The 49% burnup of initial plutonium is equivalent to an effective burnup cross section of about 18 barns for ^{242}Pu.

DISCUSSION

Description of Target Slug

The target slug consisted of aluminum-jacketed compacts of a blend of PuO_2 in aluminum powder similar to the targets used for irradiation of NpO_2.[3] (The target slug was identified as SRP-P3). Two wafers ($\sim\frac{1}{2}$ inch thick) were cut from the midregion of the irradiated slug. Weights and thicknesses of the wafers are listed in Table I.

TABLE I
PROPERTIES OF TARGET SLUG

	Target Slug Before Irradiation	Wafers Cut from Irradiated Target Slug	
		Wafer A	Wafer B
Total wafer weight, g		19.94(a)	15.99(a)
Core weight, g	171.45(b)	17.30(c)	13.85(c)
Thickness, in.		0.558(d)	0.45(d,g)
Core volume, cm^3	52.5(e)	5.30_6(f)	4.3(f)
Pu content, g	34.90(b)		
^{242}Pu content, g	33.82		
Fraction of total target slug			
Core weight		0.100_9	0.0808
Core volume		0.101_1	0.082(g)

(a) Average of four weighings. Rel Std Dev: Wafer A, 0.075%; Wafer B, 0.036%.
(b) From production records.
(c) Calculated from total wafer weight by subtraction of 0.038-in. Al cladding.
(d) Average of 12 measurements. Rel Std Dev: Wafer A, 0.79%; Wafer B, 0.73%.
(e) Calculated on the basis of 99.3% of theoretical density, the value experienced in the Savannah River Plant for actinide oxide—aluminum mixtures after pressing at 620°C and 19 tons/in^2.
(f) Calculated for 0.862-inch-diameter core. This diameter was determined by measuring an X-ray of the target slug before irradiation and correcting for 0.64% expansion during irradiation.
(g) Values not used in calculations because of irregular thickness of Wafer B.

Dissolution of Wafers

The aluminum cladding and aluminum matrix of the wafers were dissolved in hot $NaOH-NaNO_3$, and the solution was adjusted to 12M HNO_3—0.1M HF to dissolve the actinides.[4] Dissolution was complete in 9 hours except for a small quantity of siliceous residue. The residue was filtered from the solution, and the filtrate was diluted to 2 liters with 1M HNO_3.

The residue from the dissolution of Wafer A (about 3 cc of gray flocculent solid) was washed with 1M HNO_3. About 95% of the residue was dissolved in 25% NaOH; the remaining residue dissolved upon acidification of the caustic solution with 16 HNO_3—0.1M HF. Analysis of the acidified solution showed that the residue contained only 0.93 mg of ^{242}Pu and 0.56 mg of ^{244}Cm (less than 0.1% of either product).

Analytical Methods

The ^{242}Pu remaining after irradiation was determined by mass spectrometry with isotopic dilution; a solution of NBS-certified ^{239}Pu was used as the standard. Persulfate oxidation was used to ensure exchange between the plutonium isotopes, and the plutonium was purified by extraction with TTA (thenoyl trifluoroacetone).[5] Samples without added ^{239}Pu were purified by TTA extraction for determination of isotopic compositions.

The concentration of ^{244}Cm was determined by measurement of its alpha activity on counting plates prepared from dilutions of the dissolver solution. The calculation of ^{244}Cm concentration was based on an 18.11-year half-life determined by W. C. Bentley as reported by D. Metta.[6] Pulse height analyses verified that ^{244}Cm contributed >>99% of the alpha activity.

The isotopic composition of the curium was determined by mass spectrometric measurements on samples free from americium, rare earths, plutonium, fission products, and aluminum. These samples were obtained by the following treatment. A $La(OH)_3$ precipitation was used to concentrate the actinides and lanthanides and separate them from aluminum, cesium, and other components that were soluble in 3N NaOH. The $La(OH)_3$ precipitate was dissolved, adjusted to 0.05N HNO_3, and equilibrated with "Dowex" 50X-4 resin.* Curium was eluted with ammonium alphahydroxyisobutyrate by the technique of Smith and Hoffman.[7] Three ion exchange purifications were necessary to obtain high purity curium.

* "Dowex" is a trademark of Dow Chemical Co.

Americium was determined by mass spectrometry with isotopic dilution; ^{241}Am tracer was used as the standard.[8] The americium was concentrated and purified by oxidizing it to the VI state and precipitating curium and rare earths as trivalent fluorides.[9] Plutonium was removed by absorption on "Dowex" 1X-4 resin in the chloride form.

Results

Results of the analyses of the target slug are summarized in Table II. The isotopic compositions of the plutonium and curium are given in Table III.

The plutonium mass analysis data suggest an effective burnup cross section of about 18 barns for ^{242}Pu and an effective cross section for the production of ^{244}Pu of about 35 barns in the Savannah River high flux spectrum. The yields of curium isotopes of masses 245 through 248 are lower than those predicted from reactor operation at lower flux and harder neutron spectra.

TABLE II

ANALYSIS OF IRRADIATED TARGET SLUG

	Weight, g per g of ^{242}Pu irradiated			
	Wafer A	Wafer B	Mean	Predicted[1]
^{242}Pu (99.53% isotopic purity)	0.520	0.520	0.511	0.502
^{243}Am (99.95% isotopic purity)	0.137	0.134	0.136	0.176
^{244}Cm (99.63% isotopic purity)	0.277	0.275	0.276	0.255
Total actinides	0.946	0.923	0.935	-
Fission products[a]	0.091	0.085	0.088	-
	Weight, g per g of total Pu irradiated			
	Wafer A	Wafer B	Mean	
Total products	1.005	0.977	0.991	

(a) Assuming mean effective mass of 242 for fissioning nucleus and 6.0% yield of ^{137}Cs from fissions. ^{137}Cs was determined by the tetraphenyl boron method.[10]

TABLE III

ISOTOPIC COMPOSITION OF PLUTONIUM AND CURIUM IN TARGET SLUG

	Plutonium, wt %					
	238	239	240	241	242	244
Before irradiation	0.44	0.35	1.71	0.62	96.89	(a)
After irradiation[b]	$\leq$0.005	0.017	0.112	0.023	99.53	0.321

	Curium, wt %						
	242	243	244	245	246	247	248
Found	<0.004[c]	0.016[c]	96.63	0.580	2.71	0.044	0.022
Predicted	0.001	$<10^{-4}$	93.33	0.839	5.40	0.17	0.26

(a) Not measured. Estimated to be 0.045% from prior irradiation history and distribution of masses 239 through 242.
(b) Avg of data on 9 samples.
(c) Measured by alpha pulse energy analysis.

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