DP-1137

AEC RESEARCH AND DEVELOPMENT REPORT

YIELDS OF TRANSCURIUM NUCLIDES IN THE SRP HIGH FLUX REACTOR

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Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.65

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DP-1137

Chemistry
(TID-4500)

YIELDS OF TRANSCURIUM NUCLIDES IN THE SRP HIGH FLUX REACTOR

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December 1967

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CONTRACT AT(07-2)-1 WITH THE
UNITED STATES ATOMIC ENERGY COMMISSION

ABSTRACT

Yields of the transcurium nuclides (through mass 255) produced in four $^{242}PuO_2$ targets that were irradiated in the Savannah River Plant high flux reactor were measured by chemical analysis and radioassay. Total integrated exposure of individual targets ranged from 3.8×10^{22} to 9.3×10^{22} n/cm^2.

INTRODUCTION

Eight target slugs, each containing 33.8 g of ^{242}Pu (as PuO_2), were irradiated in a Savannah River reactor to provide the High Flux Isotopes Reactor (HFIR) at Oak Ridge with target material for further irradiation to produce ^{252}Cf and higher actinides.

Wafer sections from four of these Savannah River Plant target slugs were analyzed for actinide yields after high flux irradiations of varying duration. This report presents these yields and the methods used for analysis of nuclides from ^{249}Bk through ^{255}Es. This study is part of a program to develop an improved set of cross sections for use in predicting actinide yields in the Savannah River reactors.

SUMMARY

Four target slugs, each containing 33.8 g of ^{242}Pu as the oxide compacted with aluminum powder, were analyzed for productivity of transcurium nuclides through mass 255 following high flux irradiation. Integrated neutron exposures ranged from 3.8×10^{22} to 9.3×10^{22} n/cm^2.

Wafers sectioned from each slug were dissolved, and the solutions were purified by ion exchange and solvent extraction. The individual nuclides were determined by appropriate combinations of alpha counting and pulse height analysis, neutron counting, or mass spectrometry. ^{249}Bk was determined through alpha counting and pulse height analysis of its low-abundance alpha branch. Mass spectrometry was used to measure ratios of the californium isotopes; alpha counting and pulse height analysis were used to confirm the 250/252 ratio. The reference isotope in the californium series was ^{252}Cf, which was measured by alpha counting and confirmed by neutron counting. ^{254}Cf was

estimated through neutron counting. Alpha particle measurement techniques were used for ^{254}Es and for ^{253}Es, the daughter of ^{253}Cf. Alpha particles of ^{255}Fm were observed in the alpha spectrum of einsteinium isolated from the two most highly irradiated targets.

Interpretation of these data in terms of cross sections for predicting actinide productivity in the Savannah River reactors will be the subject of a later report.

DISCUSSION

DESCRIPTION OF SLUGS AND DISSOLUTION OF WAFERS

The four target slugs that were examined were designated P-3, P-4, P-5, and P-6 in order of increasing neutron exposure. A description of Slug P-3, and the methods used in cutting, measuring, and dissolving wafers from it have been reported.[1] The other three slugs were sampled similarly.

ANALYTICAL METHODS

General

Fifty milliliters of dissolver solution of a wafer from each slug was purified by chromatographic and solvent extraction procedures. The purification plan for each slug differed slightly because the priority of interest in certain nuclides varied or because experience with the initial targets suggested improvements in subsequent treatments. Whenever possible, chemical yields were corrected on the basis of gross alpha counting of solutions before and after a purification step.

Slug P-3

The aliquot of dissolver solution was treated with 6M NaOH to separate the actinides by precipitation from aluminum. The actinide precipitate was dissolved in 8M HNO_3, and plutonium was removed from the solution by anion exchange. Next, the acidity

of the effluent was adjusted, and the ^{144}Ce-^{144}Pr and a transcurium fraction were separated from most of the ^{244}Cm by cation exchange chromatography with 0.4M ammonium α-hydroxyisobutyrate[2] as the elutriant. The transcurium fraction in 10M HNO_3 was extracted with thenoyltrifluoroacetone to remove ^{95}Zr-^{95}Nb.[3] After addition of $KBrO_3$ to oxidize berkelium, extraction chromatography with a column of di(2-ethylhexyl) phosphoric acid on diatomaceous earth[4] was used to remove ^{249}Bk. The remainder of the curium was removed by two cycles of closely controlled cation exchange chromatography with a heated column.[5]

^{250}Cf, ^{252}Cf, ^{253}Es, and ^{254}Es were determined by alpha counting and pulse analysis (Figure 1) of an aliquot of the purified fraction. ^{249}Bk was determined in the purified berkelium

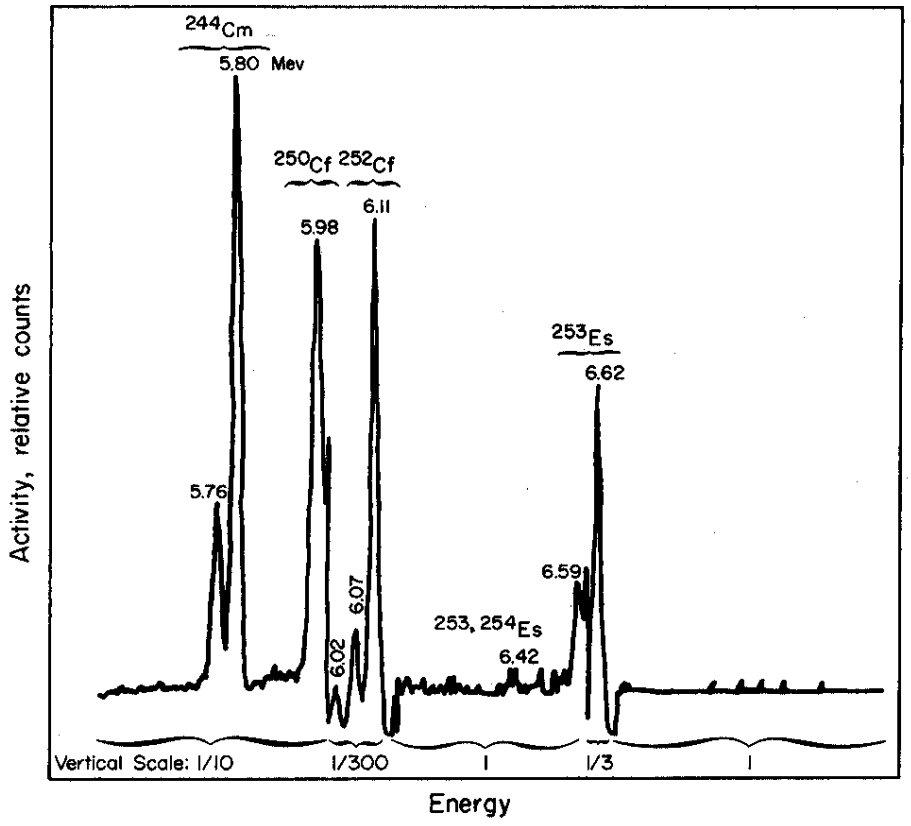

FIG. 1 ALPHA SPECTRUM OF Es-Cf FRACTION OF SLUG P-3

fraction by counting the 1.41 x 10^{-3}%-abundant alpha branch[6] (Figure 2). The remainder of the einsteinium-californium fraction was analyzed by mass spectrometry after further purification by a laboratory-scale extraction with tertiary amine[7] to remove $^{152-154}$Eu. The yield of ^{251}Cf was obtained from the mass spectral analysis; the yield of ^{253}Cf was determined from its daughter, ^{253}Es. An upper limit for ^{254}Cf was obtained by counting[8] the spontaneous fission neutrons from the einsteinium-californium fraction and correcting for those from ^{252}Cf.

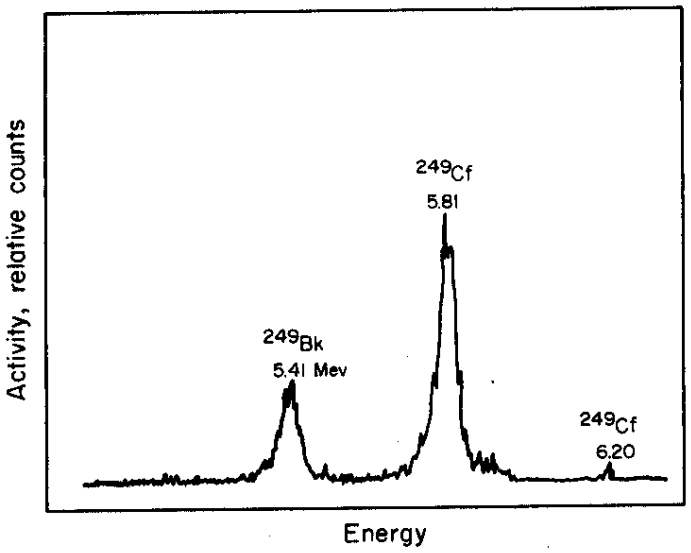

FIG. 2 ALPHA SPECTRUM OF Bk FRACTION OF SLUG P-3

Slug P-4

Aluminum, plutonium, ^{144}Ce-^{144}Pr, and most of the ^{244}Cm were removed as described for Slug P-3. ^{249}Bk was extracted with di(2-ethylhexyl) phosphoric acid[9] from a solution of the transcurium fraction in HNO_3-$KBrO_3$, and was determined by alpha counting. A purified einsteinium-californium fraction was obtained after one cycle of heated-column ion exchange chromatography. The alpha pulse spectrum of this fraction is shown in Figure 3. The einsteinium-californium fraction was analyzed by mass spectrometry after purification from lanthanides by tertiary amine extraction.

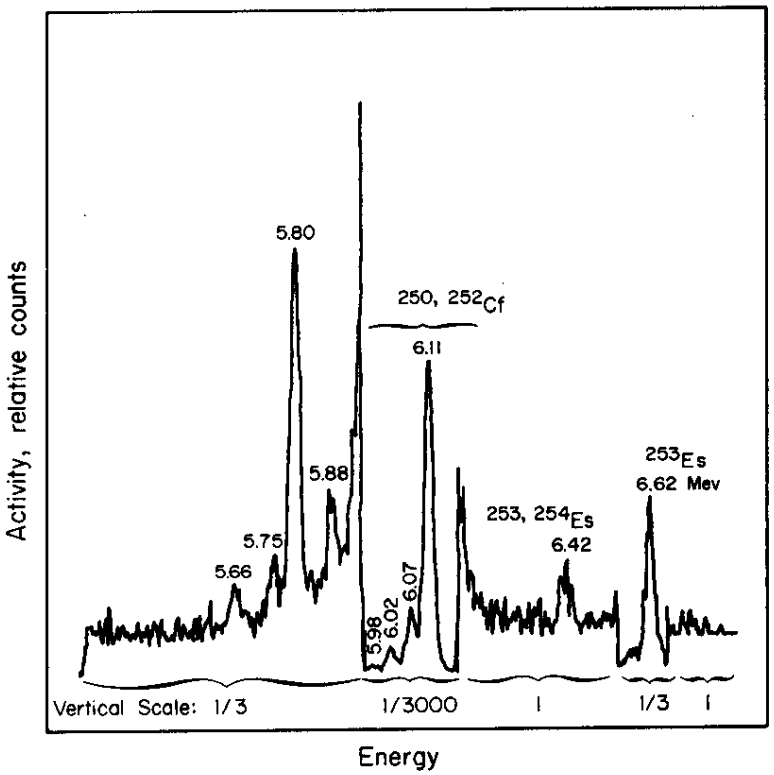

FIG. 3 ALPHA SPECTRUM OF Es-Cf FRACTION OF SLUG P-4

Slugs P-5 and P-6

Dissolver solution aliquots of these two slugs were treated in the same manner except that after removing the aluminum, the separate plutonium removal step (anion exchange) was omitted for Slug P-6. Following removal of ^{144}Ce-^{144}Pr and ^{244}Cm by cation exchange chromatography, two cycles of tertiary amine extraction yielded a partially purified transcurium fraction. The actinides were purified further and then separated by ion exchange chromatography on heated columns. Berkelium was separated by solvent extraction, as for Slug P-4.

An alpha pulse height scan of the purified transcalifornium fraction shown in Figure 4 was the first detection of element 100, fermium, at Savannah River. Calculation of the yield of ^{255}Es was based on the activity of its daughter, ^{255}Fm. The

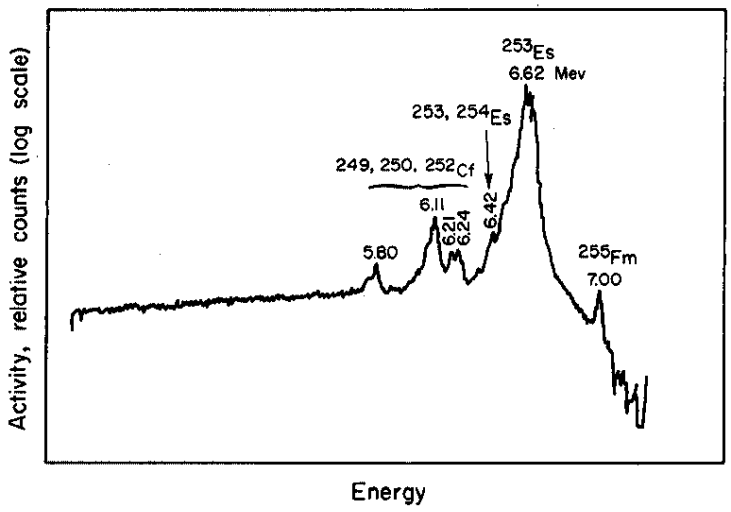

FIG. 4 ALPHA SPECTRUM OF Es FRACTION OF SLUG P-5

alpha scan from the corresponding fraction from Slug P-6 is shown in Figure 5. ^{254}Es was determined after allowing the shorter-lived, more-alpha-abundant ^{253}Es to decay sufficiently to prevent interference by a 0.1%-abundant alpha branch of this activity at the same energy as alpha particles from ^{254}Es. The californium isotopes in the purified californium fractions were analyzed mass spectrometrically.

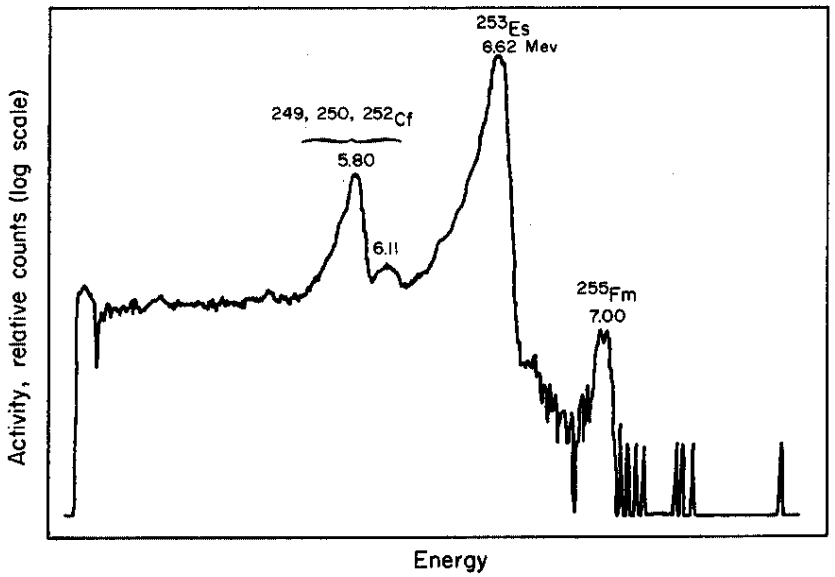

FIG. 5 ALPHA SPECTRUM OF Es FRACTION OF SLUG P-6

RESULTS

The transcurium nuclide yields from the analysis of the four target slugs are given in the following table and compared graphically in Figure 6.

High Flux Yields of Transcurium Nuclides in $^{242}PuO_2$ Target Slugs at Discharge from Reactor

	Weight, g per g of ^{242}Pu irradiated			
	Slug P-3	Slug P-4	Slug P-5	Slug P-6
Flux[a]:	3.8×10^{22}	5.2×10^{22}	7.3×10^{22}	9.3×10^{22}
Nuclide				
^{249}Bk	3.6×10^{-7}	1.1×10^{-6}	2.1×10^{-6}	3.5×10^{-6}
^{250}Cf	3.0×10^{-7}	8.6×10^{-7}	2.3×10^{-6}	4.1×10^{-6}
^{251}Cf	6.8×10^{-8}	2.1×10^{-7}	5.0×10^{-7}	8.0×10^{-7}
^{252}Cf	7.1×10^{-7}	3.0×10^{-6}	1.1×10^{-5}	2.0×10^{-5}
^{253}Cf	8.6×10^{-9}	5.0×10^{-8}	1.1×10^{-7}	3.3×10^{-7}
^{254}Cf	$< 4 \times 10^{-11}$	$< 4 \times 10^{-12}$	1.6×10^{-9}	5.3×10^{-9}
^{254}Es	2.7×10^{-11}	7.4×10^{-11}	1.4×10^{-10}	4.1×10^{-10}
^{255}Es	-	-	2×10^{-11}	8×10^{-11}
^{244}Cm	0.276	0.374	0.417	0.430

[a] Integrated neutron flux, n/cm^2

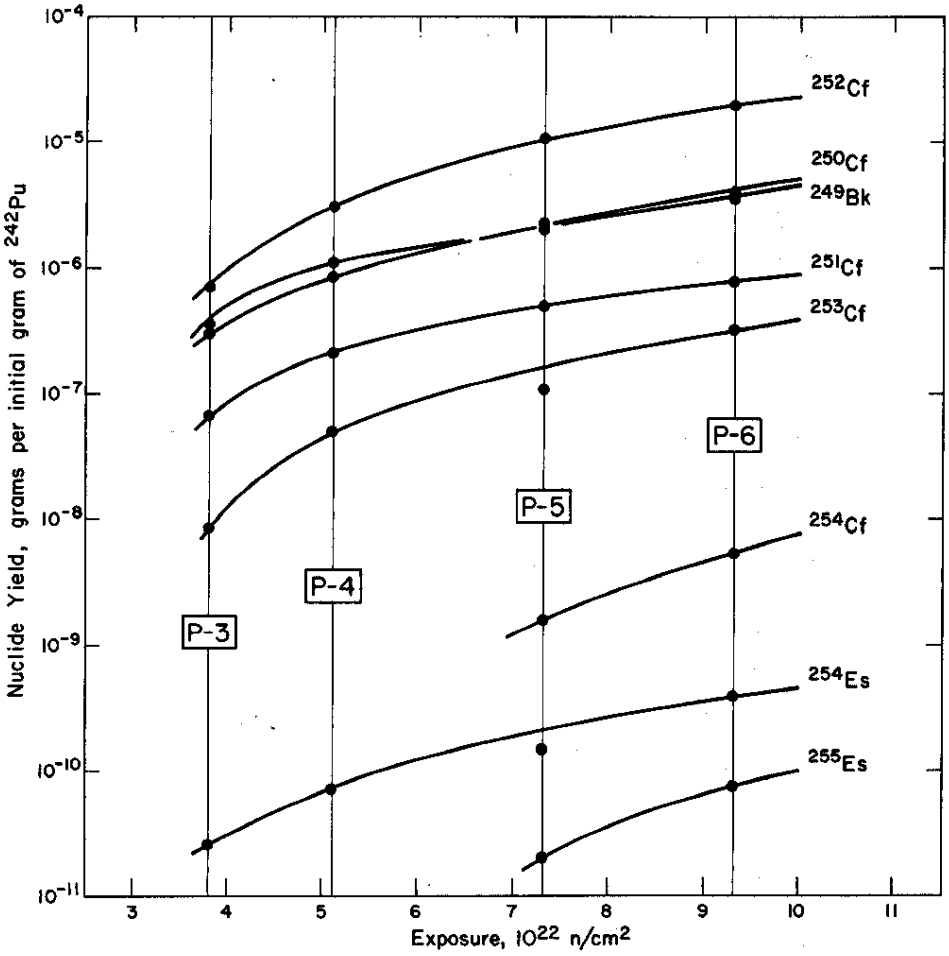

FIG. 6 TRANSCURIUM ACTINIDE YIELDS FROM HIGH FLUX IRRADIATION OF $^{242}PuO_2$

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