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ACTINIDE PROPERTIES AND METHODS OF PRODUCTION

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ACTINIDE PROPERTIES AND METHODS OF PRODUCTION

by

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ABSTRACT

The actinide series is composed of fifteen elements with atomic numbers 89 (actinium) to 103 (lawrencium). The principal naturally occurring actinides are ^{232}Th, ^{231}Pa, ^{235}U, and ^{238}U. Other actinides can be produced by irradiating heavy elements such as uranium with neutrons in a nuclear reactor, by bombarding nuclei with heavy ions in an accelerator, and by exposing heavy elements to very high neutron fluxes generated momentarily in thermonuclear explosions. The actinide elements are separated chemically by a combination of solvent extraction and ion exchange techniques. The nuclides ^{244}Pu through ^{257}Fm are produced by thermal neutron captures in reactors; ^{258}Md and higher nuclides are produced by heavy ion bombardment in accelerators.

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Appendices

I. INTRODUCTION

With the rapid growth of the nuclear power industry and the specialized radioisotope production programs supported by the U. S. Atomic Energy Commission, relatively large amounts of actinide radioisotopes are being produced by nuclear reactors. Smaller amounts are being produced by nuclear accelerators and by thermonuclear explosions. This report surveys the principal methods of actinide production and separation, and outlines their nuclear properties and uses.

The nuclides discussed are of special interest in industrial or scientific fields; have found practical industrial application; are useful for characterizing chemical, physical, and metallurgical properties of elements; or have interesting nuclear properties. The actinides are also important as targets for producing the still heavier "transactinide" and "superactinide" elements by accelerator reactions. These nuclides include all those from ^{237}Np (element 93) through ^{254}Es (element 99) that have half-lives longer than 162 days (^{242}Cm), and also the longest-lived nuclides of elements of higher atomic number than einsteinium.

II. THE ACTINIDE SERIES

The actinide series is composed of fifteen elements with the atomic numbers 89 (actinium) to 103 (lawrencium); of these elements, only actinium, thorium, protactinium, and uranium occur naturally to any significant extent. The relationship of the actinide series to the similar lanthanide series is shown in the periodic table (Figure II-1). Also shown are predicted positions for a "transactinide series" from elements 104 (rutherfordium*) to the postulated element 121, a hypothetical "superactinide series" from element 122 to 153, and a further hypothetical series from element 154 to 168.

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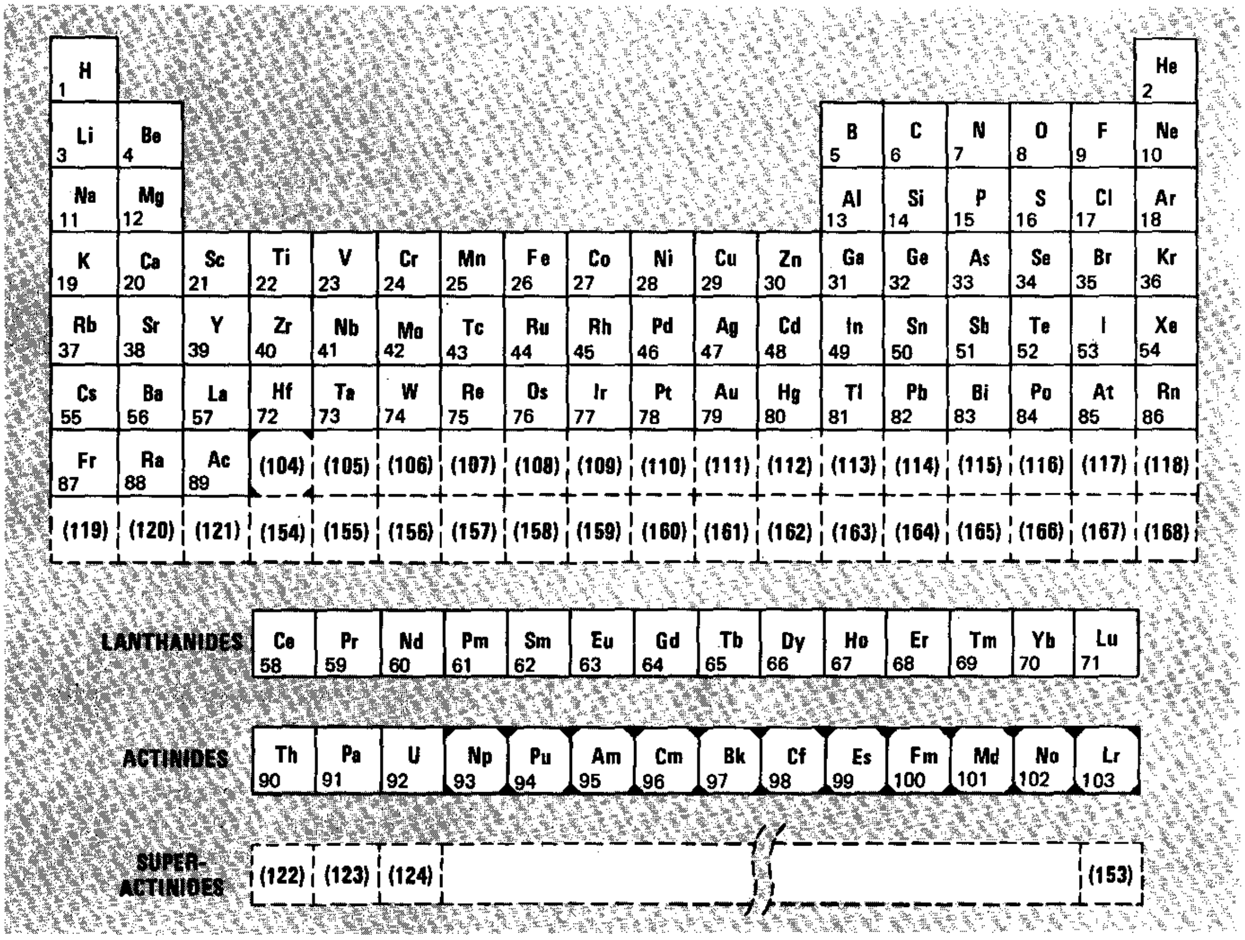
* Suggested name, not approved by The Commission on the Nomenclature of Inorganic Chemistry of the International Union of Pure and Applied Chemistry.

Calculated electron shells for these elements are shown in Figure II-2; some measured and predicted properties are listed in Table II-1. Nuclear properties of the longer-lived isotopes of these elements are listed in Table II-2.

Chemical properties of the actinides are similar to those of the lanthanides, which are the rare earth series extending from lanthanum (element 57) to lutetium (element 71). Separation of the actinides from each other is difficult; a combination of solvent extraction, ion exchange, and chromatography techniques must be used (discussed beginning on page 35).

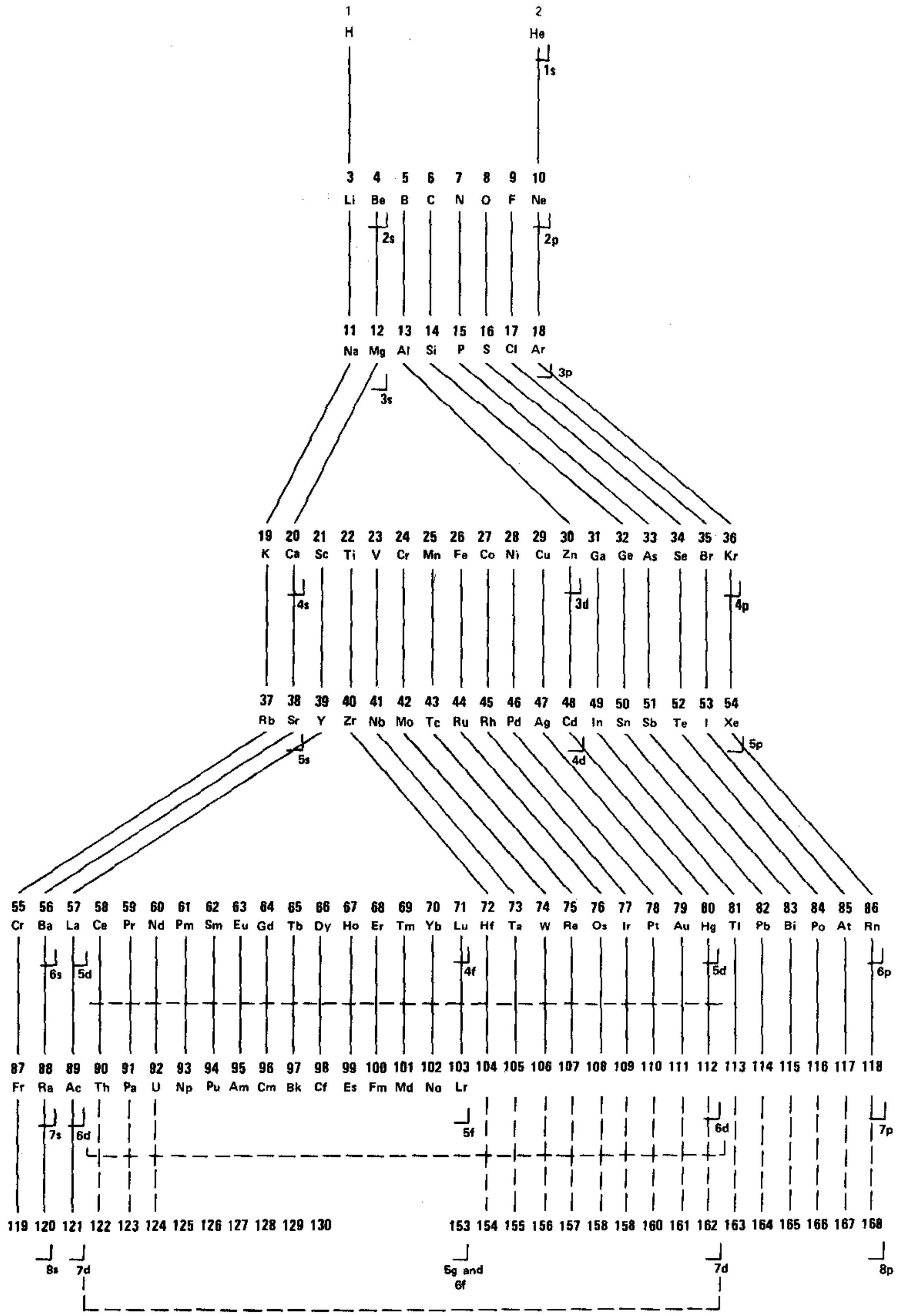


H 1																	He 2
Li 3	Be 4											B 5	C 6	N 7	O 8	F 9	Ne 10
Na 11	Mg 12											Al 13	Si 14	P 15	S 16	Cl 17	Ar 18
K 19	Ca 20	Sc 21	Ti 22	V 23	Cr 24	Mn 25	Fe 26	Co 27	Ni 28	Cu 29	Zn 30	Ga 31	Ge 32	As 33	Se 34	Br 35	Kr 36
Rb 37	Sr 38	Y 39	Zr 40	Nb 41	Mo 42	Tc 43	Ru 44	Rh 45	Pd 46	Ag 47	Cd 48	In 49	Sn 50	Sb 51	Te 52	I 53	Xe 54
Cs 55	Ba 56	La 57	Hf 72	Ta 73	W 74	Re 75	Os 76	Ir 77	Pt 78	Au 79	Hg 80	Tl 81	Pb 82	Bi 83	Po 84	At 85	Rn 86
Fr 87	Ra 88	Ac 89	(104)	(105)	(106)	(107)	(108)	(109)	(110)	(111)	(112)	(113)	(114)	(115)	(116)	(117)	(118)
(119)	(120)	(121)	(154)	(155)	(156)	(157)	(158)	(159)	(160)	(161)	(162)	(163)	(164)	(165)	(166)	(167)	(168)

LANTHANIDES	Ce 58	Pr 59	Nd 60	Pm 61	Sm 62	Eu 63	Gd 64	Tb 65	Dy 66	Ho 67	Er 68	Tm 69	Yb 70	Lu 71
ACTINIDES	Th 90	Pa 91	U 92	Np 93	Pu 94	Am 95	Cm 96	Bk 97	Cf 98	Es 99	Fm 100	Md 101	No 102	Lr 103
SUPER-ACTINIDES	(122)	(123)	(124)											(153)

ISOTOPES AND RADIATION TECHNOLOGY, Vol. 7, No. 3, Spring 1970

FIGURE II-1. Conventional Form of Periodic Table Showing Predicted Locations of New Elements[14]



ISOTOPES AND RADIATION TECHNOLOGY, Vol. 7, No. 3, Spring 1970

FIGURE II-2. Modified Form of Periodic Table Showing Known and Predicted Electron Shells[14]

TABLE II-1

Some Predicted and Measured Properties of the Actinide, Transactinide, and Superactinide Elements[14]

Atomic Weight	Atomic Number	Element	Symbol	Electronic Configuration	Most Stable Oxidation State	Melting Point, °C	Density, g/cm^3 at 25°C	Ionic Radius, Å
	89	Actinium	Ac	$6d^1 7s^2$	3	1100		1.11
	90	Thorium	Th	$6d^2 7s^2$	4	1750	11.7	0.99
	91	Protactinium	Pa	$5f^2 6d^1 7s^2$	5	<1873	15.4	0.96 (4)
	92	Uranium	U	$5f^3 6d^1 7s^2$	6	1132	19.0	0.93 (4)
	93	Neptunium	Np	$5f^4 6d^1 7s^2$	5	637	20.5	0.92 (4)
	94	Plutonium	Pu	$5f^6 7s^2$	4	640	19.7	0.90
	95	Americium	Am	$5f^7 7s^2$	3	995	13.6	0.99
	96	Curium	Cm	$5f^7 6d^1 7s^2$	3	1340	7	0.98
	97	Berkelium	Bk	$5f^8 6d^1 7s^2$ $5f^9 7s^2$	3	986		
	98	Californium	Cf	$5f^{10} 7s^2$	3			0.96
	99	Einsteinium	Es	$5f^{11} 7s^2$	3			
	100	Fermium	Fm	$5f^{12} 7s^2$	3			
	101	Mendelevium	Md	$5f^{13} 7s^2$				
	102	Nobelium	No	$5f^{14} 7s^2$				
257	103	Lawrencium	Lw	$5f^{14} 6d^1 7s^2$				
272	104[a]	Rutherfordium	Rf	$6d^2 7s^2$	4	2100	18	0.78
	105	Hahnium	Ha	$6d^3 7s^2$				
	106			$6d^4 7s^2$				
	107			$6d^5 7s^2$				
	108			$6d^6 7s^2$				
	109			$6d^7 7s^2$				
	110			$6d^8 7s^2$				
	111			$6d^9 7s^2$				
	112			$6d^{10} 7s^2$				
297	113			$7s^2 7p^1$	1	430	16	1.48
298	114			$7s^2 7p^2$	2	70	14	1.31
	115			$7s^2 7p^3$				
	116			$7s^2 7p^4$				
311	117			$7s^2 7p^5$		450		2.3
314	118			$7s^2 7p^6$		-15		
315	119			$7s^2 7p^6 8s^1$		15	3	1.85
316	120			$7s^2 7p^6 8s^2$		680	7	1.6
	121			$8p^1 8s^2$				
	122			$5g^0 6f^0 7d^1 8s^2$				
	123			$5g^0 6f^1 7d^1 8p^1 8s^2$				
	124			$5g^0 6f^3 7d^0 8p^1 8s^2$				
	125			$5g^1 6f^3 7d^0 8p^1 8s^2$				
	126			$5g^2 6f^2 7d^1 8p^1 8s^2$ $5g^2 6f^3 8p^1 8s^2$				
	127			$5g^3 6f^3 7d^1 8s^2$				
	128			$5g^4 6f^3 7d^1 8s^2$				
	129			$5g^5 6f^2 7d^2 8s^2$				
	130			$5g^6 6f^2 7d^2 8s^2$				
	131			$5g^7 6f^1 7d^3 8s^2$				
	132			$5g^8 6f^1 7d^3 8s^2$				

---

[a] All properties for element 104 and higher are predicted.

TABLE II-2

Nuclear Properties of Longer-Lived Isotopes of Atomic Numbers 92-101[1,2]

	Half-Life		Principal Mode of Decay[a]	Approximate Energies of Major Radiations, Mev				Neutron Thermal Cross Sections, barns[c]	
Nuclide	Total	Spontaneous Fission		α	β^-	γ	e^-[b]	σ_c	σ_f
^{232}U	72 yr	8x10^{13} yr	α	5.32		0.058	0.050	78	72
^{233}U	1.59x10^{5} yr		α	4.82		0.042	0.023	47	531
^{234}U	2.47x10^{5} yr	2x10^{16} yr	α	4.77		0.053		94	<5 mb
^{235}U	7.1x10^{8} yr	1.9x10^{17} yr	α	4.16-4.60		0.19		98	580
^{236}U	2.42x10^{7} yr	2x10^{16} yr	α	4.49		0.05	0.032	5.5	
^{238}U	4.51x10^{9} yr	6.5x10^{15} yr	α	4.20		0.048	0.030	2.73	<0.5 mb
^{235}Np	410 days		ε	4.37-5.10		0.026			
^{236}Np	>5x10^{3} yr		β^-	-		-			2800
^{237}Np	2.14x10^{6} yr	>10^{18} yr	α	4.52-4.87		0.029-0.24	0.009	169	0.018
^{236}Pu	2.85 yr	3.5x10^{9} yr	α	5.77		0.047	0.028		150
^{238}Pu	87.4 yr	4.9x10^{10} yr	α	5.50		0.044	0.024	540	17
^{239}Pu	24,390 yr	5.5x10^{15} yr	α	5.16		0.052	0.008	271	742
^{240}Pu	6600 yr	1.34x10^{11} yr	α	5.17		0.045	0.026	280	<0.08
^{241}Pu	14.3 yr		β^-	4.90	0.021	0.149		364	1005
^{242}Pu	3.87x10^{5} yr	7.1x10^{10} yr	α	4.90		0.04		20	<0.2
^{244}Pu	8.3x10^{7} yr	2.5x10^{10} yr	α	4.59				1.8	
^{241}Am	433 yr	2x10^{14} yr	α	5.49		0.060	0.022	710	3
^{242m}Am	152 yr	9.5x10^{11} yr	IT	5.21		0.042-0.206	0.028	1500	6500
^{243}Am	7370 yr		α	5.28		0.075	0.011	100	<0.07
^{242}Cm	163 days	7.2x10^{6} yr	α	6.11		0.044	0.022	25	<5
^{243}Cm	32 yr		α	5.78		0.043	0.022	13	1
^{245}Cm	8300 yr	1.31x10^{7} yr	α	5.36		0.173		270	2000
^{246}Cm	4710 yr	1.7x10^{7} yr	α	5.39				9	
^{247}Cm	1.6x10^{7} yr		α					180	108
^{248}Cm	3.52x10^{5} yr	4.6x10^{6} yr	α	5.08				7	
^{250}Cm	1.1x10^{4} yr	1.9x10^{4} yr	SF						
^{247}Bk	1400 yr		α	5.52		0.084			
^{248}Bk	>9 yr		α						
^{249}Bk	311 days		β^-	5.41	0.125	0.33		800	6
^{248}Cf	350 days	>1.5x10^{4} yr	α	6.27					
^{249}Cf	352 yr	1.5x10^{9} yr	α	5.81		0.388		270	1650
^{250}Cf	13.1 yr	1.7x10^{4} yr	α	6.03		0.043	0.023	1500	<350
^{251}Cf	900 yr		α	5.67		0.18		3000	1500
^{252}Cf	2.65 yr	85 yr	α	6.12		0.043	0.022	20	
^{254}Es	276 days	7x10^{5} yr	α	6.44		0.063	0.011	<40	3060
^{257}Fm	80 days	100 yr	α	6.52		0.062	0.037		
^{258}Md	54 days		α	6.73					
^{255}No	3 min		α	8.08					
^{256}Lw	35 sec		α	8.4					
^{261}Rf	1 min		α	8.2					
^{261}Ha	0.1-3 sec		α	8.4					

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a. α, β^-, γ: alpha, negatron, gamma emission energy in Mev.
IT: isomeric transition - decay from an excited metastable state to a lower state.

b. e^-: conversion electron emission energy in Mev.

c. σ_c, σ_f: thermal neutron capture and fission cross sections in barns.

III. PRODUCTION OF ACTINIDE ELEMENTS

The principal naturally occurring actinides are ^{227}Ac, ^{232}Th, ^{231}Pa, ^{235}U, and ^{238}U. Other actinides can be produced by irradiating heavy elements such as uranium with neutrons in a nuclear reactor, by bombarding nuclei with heavy ions in an accelerator, and by exposing heavy elements to the very high neutron fluxes generated momentarily in thermonuclear explosions.

The neutron irradiation routes have the advantage of lower costs, primarily because the uncharged neutron needs no energy to penetrate the repulsion forces of the target nucleus.

Reactor irradiation of uranium is a well developed technology. Large quantities of relatively stable actinides through ^{244}Cm will soon be available as byproducts of nuclear power reactor operation. Heavier nuclides through ^{257}Fm are being produced in smaller quantities by special irradiations in AEC reactors.

The technologies of charged particle irradiation in an accelerator and neutron irradiation in a thermonuclear explosion are less developed than that of neutron irradiation in a reactor; however, these two processes may be competitive with reactor neutron irradiation for producing some heavier nuclides. Furthermore, it is theoretically possible, although undemonstrated, that thermonuclear explosions may yield nuclides heavier than ^{257}Fm. Moreover, since not all desired nuclides can be made by successive neutron capture, heavy-ion bombardment in an accelerator is the only practical route to elements from fermium (element 100) through hahnium (element 105) and to neutron-deficient isotopes of the lower actinides.

Table III-1 compares the cost of making ^{252}Cf by each of these three methods:

- The High-Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL) was chosen to illustrate a high-flux reactor because production and cost information has been published in unclassified literature.[23] The operating thermal flux in the HFIR flux-trap region is about 2×10^{15} n/(cm^2-sec) with a target loading of 300 g of ^{242}Pu. Assuming continuous operation of the reactor for a year, an annual operating budget of \$4 million, and a 20-year writeoff of the initial capitalization, the cost per mole of neutrons produced per cm^2 is \$45 million.

TABLE III-1

Cost Comparison for Synthesis of ^{252}Cf

	High Flux Isotope Reactor (HFIR)	Nominal Heavy Ion Accelerator	Thermonuclear Explosion
Flux	2×10^{15} n/(cm^2-sec)	1×10^{16} ions/sec[a]	6×10^{23} n/10^{-7} sec
Irradiation Time	1 yr	1 yr	10^{-7} sec
Exposure	6×10^{22} n/cm^2	3×10^{23} ions	6×10^{23} n
Duty Factor, %	100	100	-
Initial Capitalization, $\$10^6$	15	50	80
Annual Depreciation, $\$10^6$	0.75 (20 yr)	2.5 (20 yr)	8 (10 yr)
Annual Operating Cost, $\$10^6$	4	5	3
Cost per mole of Particles, $\$10^6$	45 (n)	15 (ions)	9
Amount of ^{252}Cf Produced	8 mg	0.1 mg[b]	1-5g[c]
Unit Cost of ^{252}Cf, \$/μg	6-10	6×10^4	1-4

---

[a] Beam area is approximately 0.1 cm^2.

[b] Assumes 1 particle in 1000 will form compound nucleus, and 1 in 1000 will escape fission.

[c] See Table III-12. Cost based upon recovering 30-150 g of ^{252}Cf from 30 explosions over a 10-year period.

- For comparison with the high-flux neutron reactor route, costs were estimated for a Heavy-Ion Linear Accelerator priced at about three times the HFIR initial capitalization, and producing an ion beam of 10^{16} particles/sec. The accelerator beam area is about 0.1 cm^2, which — unlike the reactor — represents the total area available for reactions. With the same depreciation time and equipment utilization used for HFIR, the annual cost per mole of particles produced is about 1/3 that of HFIR. In the case of the accelerator, however, only an estimated one particle in 10^6 is captured to form a compound nucleus that escapes subsequent fission. Because of this, the accelerator heavy-ion irradiation is not competitive with neutron irradiation for synthesizing those nuclides that are accessible by reactor irradiation. However, accelerators do provide the only route now available to elements beyond fermium and to neutron-poor actinides.

- In the case of a thermonuclear explosion, approximately 1 mole of neutrons is produced in less than a microsecond. If a heavy element target such as ^{238}U is included in the shot, very heavy uranium isotopes such as ^{257}U are produced by rapid, successive addition of neutrons; these uranium isotopes then undergo almost immediate beta decay to form nuclides from ^{257}Fm downward. With "Gasbuggy"[67] costs as a guide, the $9 million cost per mole of neutrons produced is competitive with neutron costs in high-flux thermal reactors, and the high efficiency of conversion to the higher nuclides such as ^{252}Cf makes this technique attractive. However, processing large volumes of rock and recovering the isotopes would require large, expensive plants.

The sales prices of a few heavy isotopes, as described by Oak Ridge National Laboratory and Amersham/Searle, are given in Appendix A, page 68.

A. PRODUCTION OF ACTINIDES BY REACTOR IRRADIATION

1. Power Reactors

Commercial power reactors are expected to become the primary source of plutonium, neptunium, americium, and curium isotopes within the next few years. The principal types of power reactors predicted for installation through 1980 are the Boiling Water (BWR) and Pressurized Water (PWR) reactors operating with primarily thermal neutron fluxes of about 2 x 10^{13} n/(cm^2-sec), and the Liquid Metal Fast Breeder reactors (LMFBR) operating with primarily fast (unmoderated) neutrons. Typical examples of these reactors are listed in Table III-2.

Using these reactor types and the AEC's prediction that 150 x 10^9 watts of nuclear generating capacity will be installed by 1980, a Richland study group[17] calculated the amount of fuel to be discharged from the reactors and their plutonium recycle capability; these results are listed in Table III-3.

Of the cases listed in Table III-3, two are detailed in Table III-4. The first has all generating capacity satisfied by light water reactors (PWR's) without plutonium or uranium recycle; in the second, the generating capacity is satisfied by PWR's with uranium and limited plutonium recycle plus fast breeder reactor additions beginning in 1980. For these two cases, the annual availability of ^{236}U, ^{237}Np, ^{238}Pu, ^{241}Am, ^{243}Am, and ^{244}Cm was calculated. No nuclides above ^{244}Cm will be obtained from power reactors.

Table III-5 lists the production of high-mass nuclides obtained by irradiating power reactor americium (50/50 mixture of ^{241}Am and ^{243}Am) at a flux of 1.75 x 10^{14} n/(cm^2-sec).

TABLE III-2

Typical Power Reactors

Reactor Type	Electrical Power, MW	Manufacturer	Name	Location	Owner
Boiling Water (BWR)	1065	General Electric	Brown's Ferry	Decatur, Ala.	TVA
Pressurized Water (PWR)	1060	Westinghouse	Diablo Canyon	San Luis Obispo, Calif.	Pacific Gas and Electric
Liquid Metal Fast Breeder (LMFBR)	1000	Westinghouse	1000 MW(e) Study	—	—

TABLE III-3

Power Reactor Capacity[17]

Calendar Year	Installed Capacity, 10^9 electrical watts			Annual Fuel Discharge, metric tons/year[a]		Plutonium Recycle Capacity Installed, 10^9 electrical watts		
	Case A	Case B		Case A	Case B	Case B		
	PWR	PWR	FBR	PWR	LWR-U/Pu & FBR	PWR 4 Pu[b]	PWR 10 Pu[b]	FBR
1970	12	12	0	100	100	0	0	0
1975	70	70	0	1830	1850	0	5	0
1980	150	148	2	4280	4270	8	13	2
1985	296	268	28	10700	10100	14	13	28
1990	500	390	110	20700	17700	16	13	110

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a. PWR - Light Water Reactors includes Boiling Water Reactors and Pressurized Water Reactors
FBR - Fast Breeder Reactor includes Light Metal Fast Breeder Reactors (LMFBR's)

Case A: Light Water Reactors (PWR) fueled with slightly enriched uranium without plutonium or uranium (^{236}U) recycle. Both cases assume 150 x 10^9 electrical watts installed by 1980.

Case B: Light Water Reactors with 50% ^{236}U recycle, limited plutonium recycle, and maximum Fast Breeder Reactor (FBR) additions beginning in 1980. PWR-U/Pu and FBR.

b. 4 Pu denotes reactors operating on Pu recycle for 4 years.
10 Pu denotes reactors operating on Pu recycle for 10 years.

TABLE III-4

Power Reactor Nuclide Availability[17]

Calendar Year	Cumulative Pu Stockpile, metric tons/yr		Annual Availability of ^{236}U, metric tons/yr		Annual Availability of ^{237}Np, kg/yr		Total Annual ^{238}Pu Availability from All Sources, kg/yr		Annual Availability of ^{241}Am in Am Mixture, kg/yr	
	Case A	Case B	Case A	Case B	Case A	Case B	Case A	Case B	Case A	Case B
	LWR	LWR-U/Pu, FBR	LWR	LWR-U/Pu, FBR	LWR	LWR-U/Pu, FBR	LWR	LWR-U/Pu, FBR	LWR	LWR-U/Pu, FBR
1970	2	2	0.15	0.15	15	15	6	6	1	1
1975	25	8	3.6	3.7	376	358	78	75	38	39
1980	130	2	11.0	11.8	1220	1180	515	500	122	225
1985	383	15	24.6	29.2	2580	2720	675	791	237	568
1990	907	1	46.4	57.2	4520	4970	1370	1690	377	695

Calendar Year	Annual Availability of ^{243}Am in Am Mixture, kg/yr		Total Annual Availability of ^{244}Cm from Both Spent Fuel and Am Target Irradiation, kg/yr		Annual Availability of ^{241}Am from Processing Pu Stockpile, kg/yr	
	Case A	Case B	Case A	Case B	Case A	Case B
	LWR	LWR-U/Pu, FBR	LWR	LWR-U/Pu, FBR	LWR	LWR-U/Pu, FBR
1970	2	2	1	1	10	10
1975	50	53	4	4	118	119
1980	167	384	44	74	823	806
1985	312	913	117	428	2560	2510
1990	462	645	203	603	6290	6210

TABLE III-5

Production of High Mass Nuclides by Irradiation of Power Reactor Americium at a Flux of 1.75 x 10^{14} n/(cm^2-sec)[a]

Fluence, 10^{22} n/cm^2	Days	Atoms Produced per Single "Average Atom" of ^{241}Am and ^{243}Am Irradiated						
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{242m}Am
0							1.000	
0.2	132	0.841^{-1}[a]	0.182^{-1}	0.278^{-2}	0.643^{-3}	0.702^{-1}	0.356	0.685^{-2}
0.4	265	0.150	0.540^{-1}	0.132^{-1}	0.437^{-2}	0.906^{-1}	0.124	0.245^{-2}
0.6	397	0.152	0.693^{-1}	0.246^{-1}	0.939^{-2}	0.960^{-1}	0.426^{-1}	0.849^{-3}
0.8	529	0.122	0.640^{-1}	0.307^{-1}	0.127^{-1}	0.978^{-1}	0.147^{-1}	0.293^{-3}
1.0	661	0.870^{-1}	0.497^{-1}	0.309^{-1}	0.137^{-1}	0.989^{-1}	0.514^{-2}	0.102^{-3}
1.2	794	0.575^{-1}	0.348^{-1}	0.272^{-1}	0.129^{-1}	0.993^{-1}	0.190^{-2}	0.377^{-4}
1.4	926	0.362^{-1}	0.228^{-1}	0.224^{-1}	0.112^{-1}	0.985^{-1}	0.784^{-3}	0.154^{-4}
1.6	1058	0.221^{-1}	0.143^{-1}	0.181^{-1}	0.940^{-2}	0.965^{-1}	0.385^{-3}	0.741^{-5}
1.8	1190	0.132^{-1}	0.872^{-2}	0.148^{-1}	0.781^{-2}	0.935^{-1}	0.229^{-3}	0.435^{-5}
2.0	1323	0.771^{-2}	0.519^{-2}	0.125^{-1}	0.663^{-2}	0.897^{-1}	0.161^{-3}	0.300^{-5}

Fluence, 10^{22} n/cm^2	Atoms Produced per Single "Average Atom" of ^{241}Am and ^{243}Am Irradiated							
	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm	^{247}Cm	^{248}Cm
0	1.000							
0.2	0.833	0.304	0.349^{-2}	0.189	0.175^{-2}	0.488^{-3}	0.196^{-5}	0.598^{-7}
0.4	0.687	0.277	0.509^{-2}	0.341	0.305^{-2}	0.188^{-2}	0.149^{-4}	0.984^{-6}
0.6	0.561	0.191	0.433^{-2}	0.459	0.399^{-2}	0.388^{-2}	0.440^{-4}	0.455^{-5}
0.8	0.457	0.119	0.300^{-2}	0.549	0.468^{-2}	0.631^{-2}	0.902^{-4}	0.129^{-4}
1.0	0.371	0.701^{-1}	0.189^{-2}	0.616	0.518^{-2}	0.905^{-2}	0.152^{-3}	0.279^{-4}
1.2	0.301	0.403^{-1}	0.112^{-2}	0.664	0.554^{-2}	0.120^{-1}	0.228^{-3}	0.515^{-4}
1.4	0.244	0.229^{-1}	0.651^{-3}	0.696	0.578^{-2}	0.151^{-1}	0.316^{-3}	0.849^{-4}
1.6	0.199	0.129^{-1}	0.371^{-3}	0.716	0.594^{-2}	0.182^{-1}	0.412^{-3}	0.129^{-3}
1.8	0.162	0.727^{-2}	0.210^{-3}	0.727	0.602^{-2}	0.214^{-1}	0.514^{-3}	0.185^{-3}
2.0	0.132	0.409^{-2}	0.119^{-3}	0.730	0.605^{-2}	0.246^{-1}	0.621^{-3}	0.253^{-3}

---

[a] Exponents are powers of ten by which the number is to be multiplied.

2. High-Flux Reactors

Since PWR's and LFMBR's are not capable of large-scale nuclide production above ^{244}Cm, high-flux AEC reactors are now the major route to the heavier nuclides through ^{257}Fm.

The yield in the production of higher actinides can be improved by carrying out the irradiation of plutonium in two stages. Most of the ^{239}Pu is burned out during a preliminary irradiation in a Savannah River production reactor at an average flux of about 10^{14} n/(cm^2-sec) (Table III-6). The first irradiation is stopped at an exposure of 0.3×10^{22} n/(cm^2-sec) (dashed line, Figure III-1) when not enough fissile ^{239}Pu remains to cause heat removal problems during the second irradiation at about a tenfold higher flux.

Production of some of the longer-lived isotopes of the heavier elements in the actinide series using ^{242}Pu as a target is shown in Figures III-2 and III-3. For this second stage, flux levels up to 6×10^{15} n/(cm -sec) are available in high-flux lattices at Savannah River, and up to 3×10^{15} n/(cm^2-sec) in the HFIR at ORNL. In Table III-7 exposures up to 2×10^{23} n/cm^2 at an average thermal neutron flux of 3×10^{15} n/(cm^2-sec) are given. Production of ^{257}Fm and ^{259}Md are order-of-magnitude estimates.

A maximum flux level of 6.1×10^{15} n/(cm^2-sec) was achieved at SRP during a high-flux demonstration in 1965, with an initial target loading of 0.52 kg of ^{242}Pu.[22] The lattice volume of 230 ft^3 contained 107 fuel elements and 7 target elements, all surrounded by a D_2O reflector. A schematic of the lattice is shown in Figure III-4. Similar lattices have since been used for ^{252}Cf production at SRP.

The HFIR, shown in Figure III-5, was designed to achieve a thermal neutron flux of 3×10^{15} n/(cm^2-sec) in the flux trap region with a target loading of 0.25 kg of actinides at a maximum power of 100 MW. The reactor has been used almost exclusively for producing berkelium, californium, einsteinium, and fermium.

The Advanced Test Reactor located at the National Reactor Testing Station at Idaho Falls, Idaho, also develops a maximum thermal flux in excess of 10^{15} n/(cm^2-sec) in the target area, but this reactor is used primarily for materials testing rather than heavy element production.

TABLE III-6

Composition of Plutonium After Exposure of ^{239}Pu to 3 x 10^{21} n/cm^2

	Composition, %
^{239}Pu	22.5
^{240}Pu	51.0
^{241}Pu	15.0
^{242}Pu	11.5

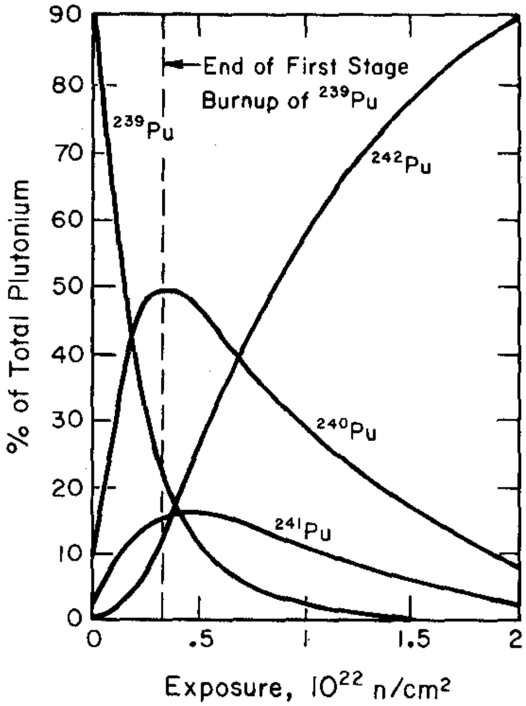

FIGURE III-1. Yield of Plutonium Isotopes from Irradiation of ^{239}Pu

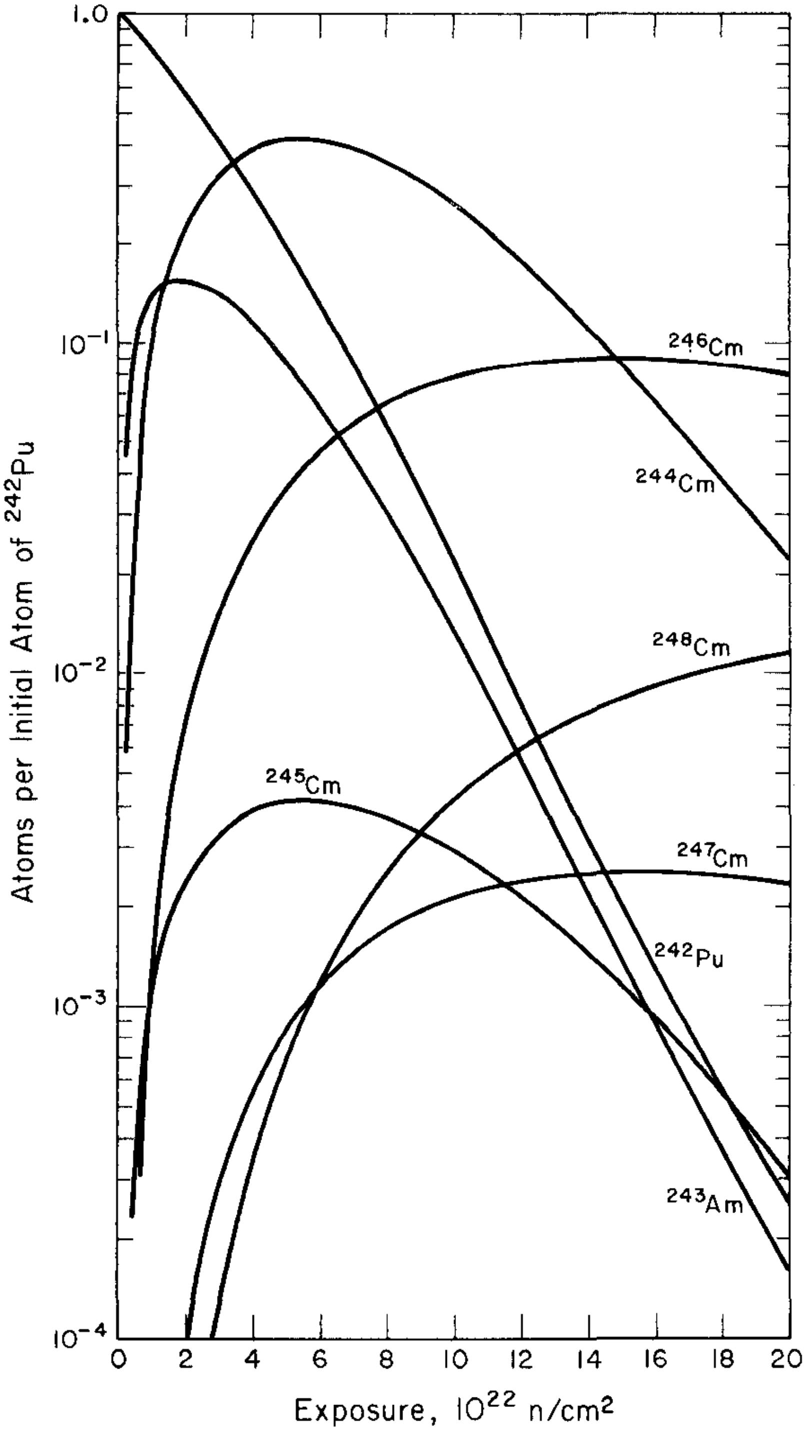

FIGURE III-2. Production of Americium and Curium from Irradiation of ^{242}Pu

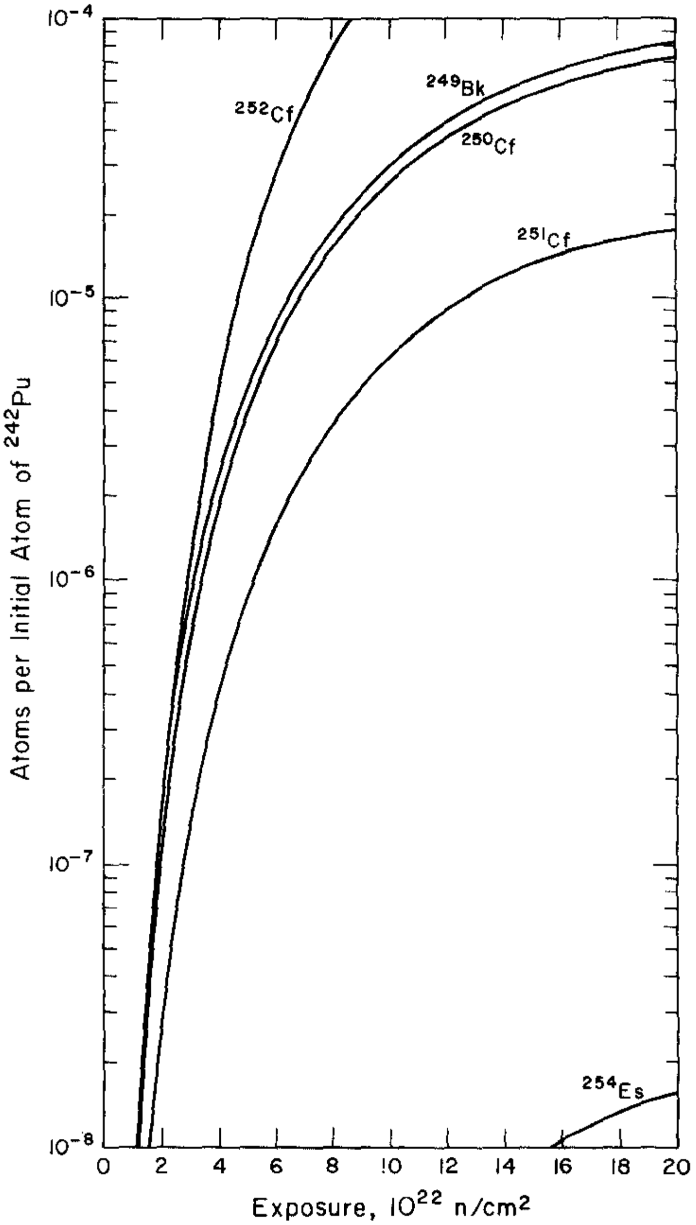

FIGURE III-3. Production of Californium, Berkelium, and Einsteinium from Irradiation of ^{242}Pu

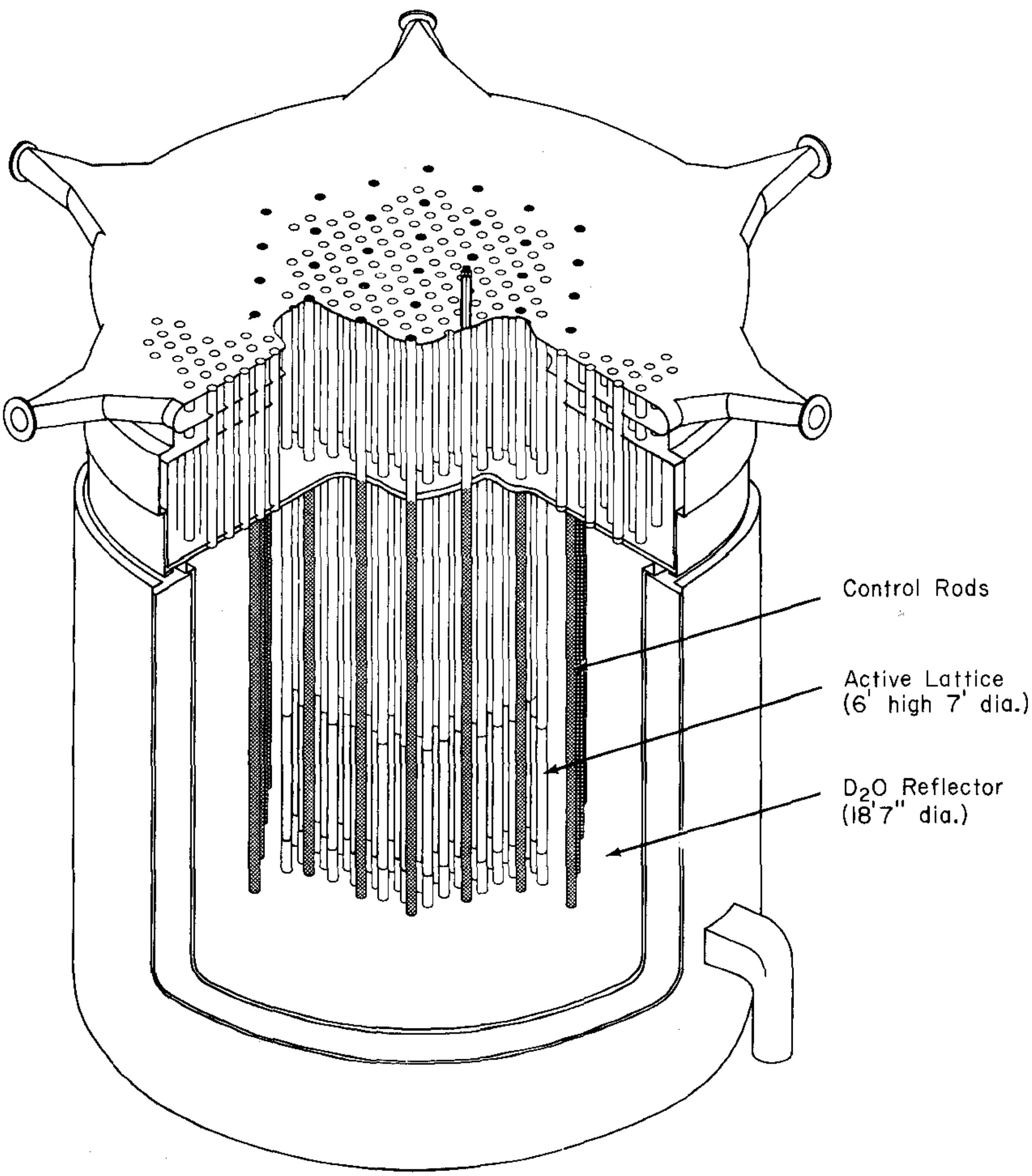

FIGURE III-4. High-Flux Demonstration Lattice in a Savannah River Reactor[22]

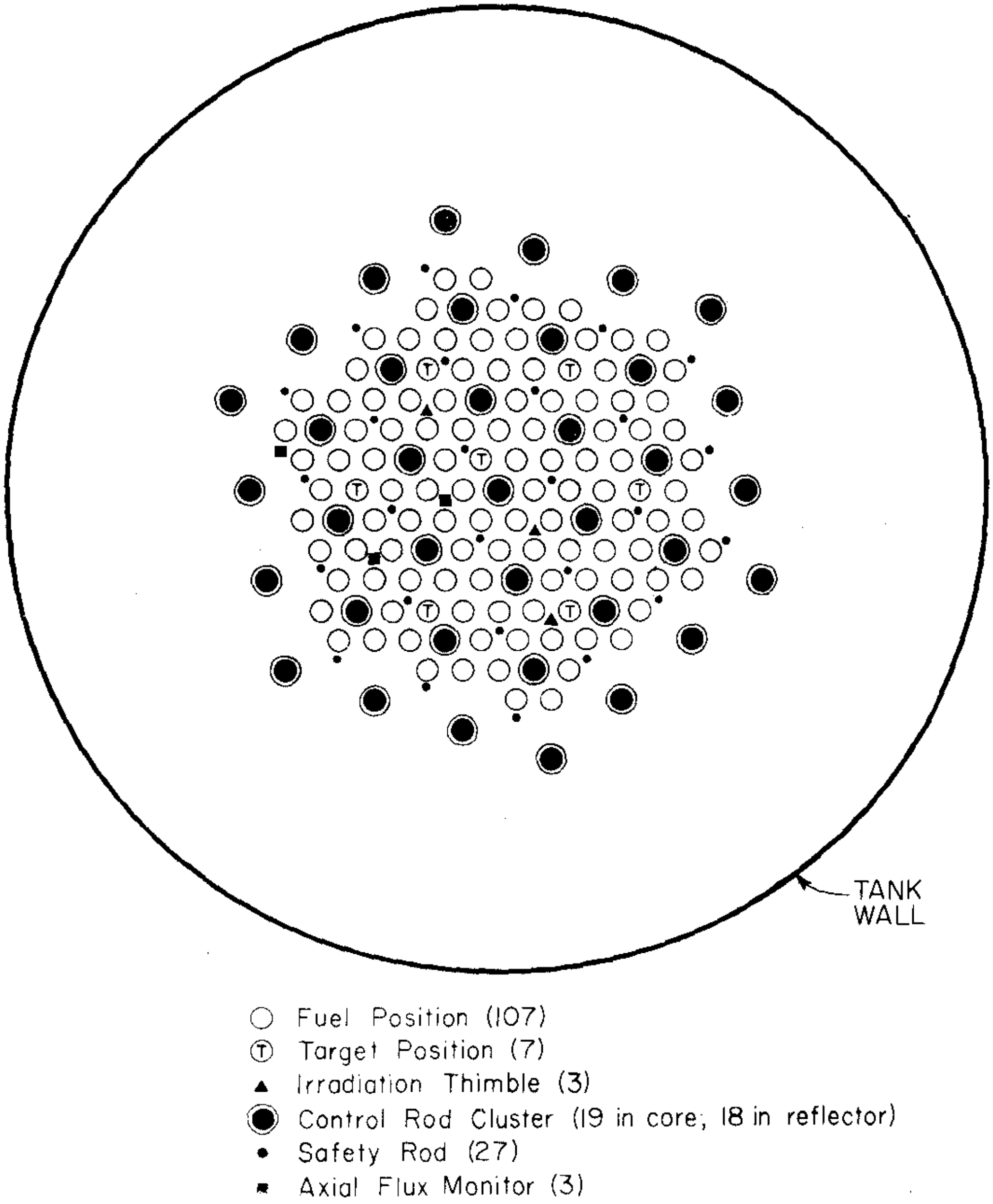

FIGURE III-4. (Continued)

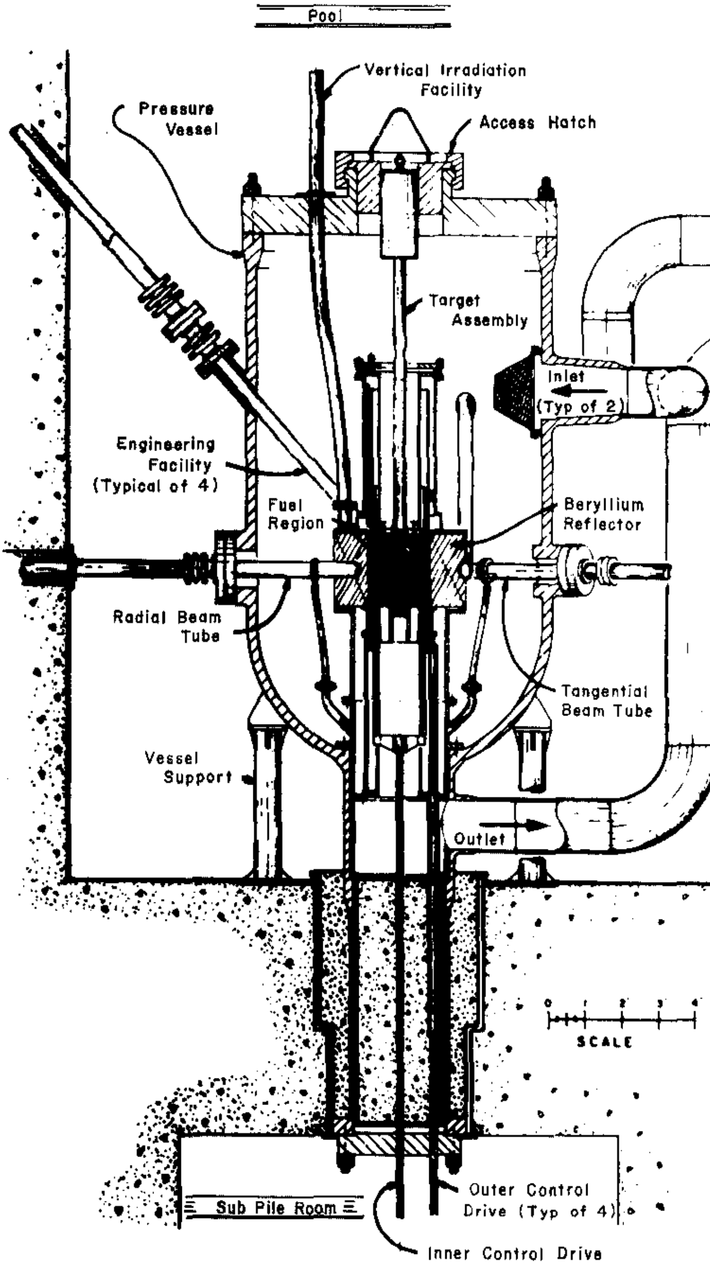

FIGURE III-5. Oak Ridge National Laboratory High-Flux Isotope Reactor

J. A. Swartout, et al. "The Oak Ridge High Flux Isotope Reactor." *Proc. U. N. Conf. Peaceful Uses At. Energy, 3rd, Geneva 7,* 360 (1965).

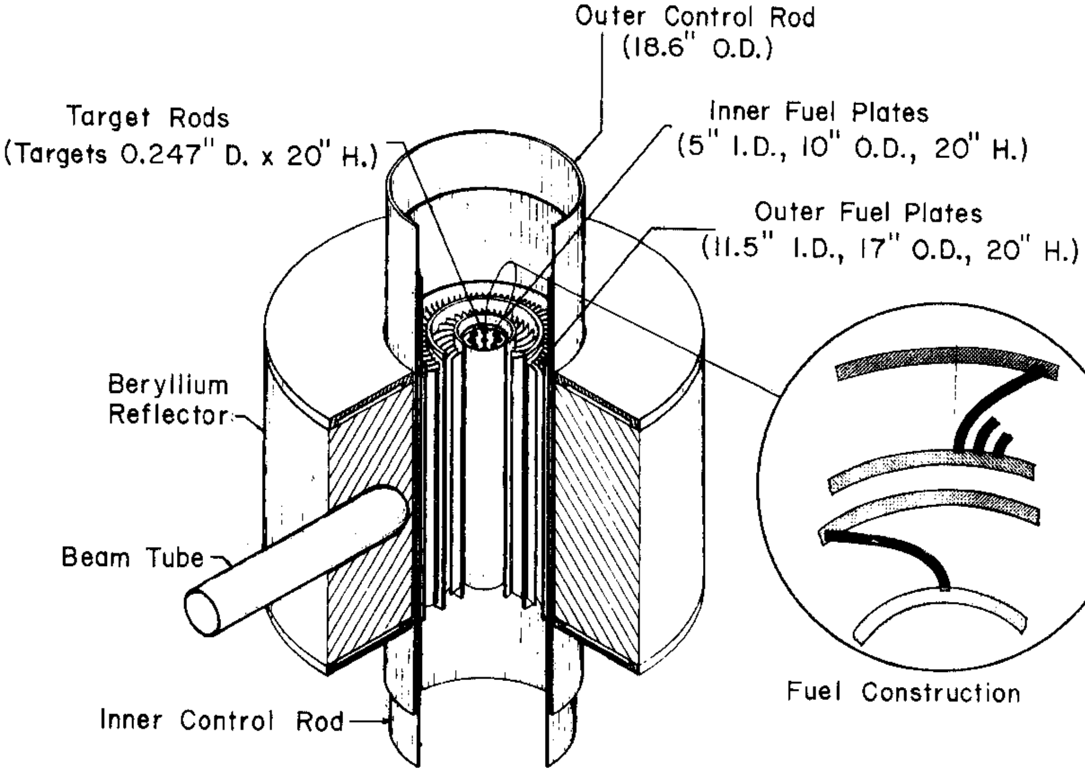

FIGURE III-5. (Continued)

C. E. Winter. "The High Flux Isotope Reactor." *Nucl. Sci. Eng.* *17*, 443 (1963).

TABLE III-7

Production of High Mass Nuclides by Irradiation of ^{242}Pu at a Flux of 3 x 10^{15} n/(cm^2-sec)

Fluence, 10^{22} n/cm^2	Days	Atoms Produced per Atom Irradiated									
		^{242}Pu	^{244}Pu	^{241}Am	^{242m}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm
0	0	1.00	0	0	0	0	0	0	0	0	0
2	77	0.566	0.173^{-2}[a]	0.547^{-7}	0.106^{-8}	0.154	0.868^{-7}	0.133^{-8}	0.226	0.254^{-2}	0.660^{-2}[a]
4	154	0.290	0.284^{-2}	0.121^{-6}	0.237^{-8}	0.113	0.432^{-6}	0.746^{-8}	0.395	0.400^{-2}	0.255^{-1}
6	232	0.131	0.349^{-2}	0.139^{-6}	0.274^{-8}	0.646^{-1}	0.770^{-6}	0.138^{-7}	0.421	0.421^{-2}	0.472^{-1}
8	309	0.542^{-1}	0.384^{-2}	0.124^{-6}	0.243^{-8}	0.313^{-1}	0.935^{-6}	0.170^{-7}	0.358	0.374^{-2}	0.661^{-1}
10	386	0.214^{-1}	0.400^{-2}	0.941^{-7}	0.185^{-8}	0.135^{-1}	0.920^{-6}	0.170^{-7}	0.265	0.296^{-2}	0.797^{-1}
12	463	0.834^{-2}	0.405^{-2}	0.644^{-7}	0.127^{-8}	0.549^{-2}	0.789^{-6}	0.146^{-7}	0.178	0.213^{-2}	0.874^{-1}
14	540	0.330^{-2}	0.402^{-2}	0.410^{-7}	0.808^{-9}	0.219^{-2}	0.614^{-6}	0.115^{-7}	0.111	0.142^{-2}	0.901^{-1}
16	617	0.134^{-2}	0.397^{-2}	0.247^{-7}	0.488^{-9}	0.883^{-3}	0.445^{-6}	0.834^{-8}	0.665^{-1}	0.887^{-3}	0.890^{-1}
18	694	0.572^{-3}	0.388^{-2}	0.144^{-7}	0.285^{-9}	0.367^{-3}	0.306^{-6}	0.575^{-8}	0.385^{-1}	0.530^{-3}	0.855^{-1}
20	772	0.255^{-3}	0.378^{-2}	0.821^{-8}	0.162^{-9}	0.159^{-3}	0.202^{-6}	0.380^{-8}	0.218^{-1}	0.307^{-3}	0.806^{-1}

Fluence, 10^{22} n/cm^2	Days	Atoms Produced per Atom Irradiated									
		^{247}Cm	^{248}Cm	^{249}Bk	^{249}Cf	^{250}Cf	^{251}Cf	^{252}Cf	^{254}Es	^{257}Fm[b]	^{259}Md[c]
0	0	0	0	0	0	0	0	0	0	0	0
2	77	0.100^{-3}	0.270^{-4}	0.164^{-6}	0.805^{-9}	0.125^{-6}	0.288^{-7}	0.142^{-6}	0.108^{-11}	0.1^{-16}	0.1^{-20}
4	154	0.554^{-3}	0.346^{-3}	0.232^{-5}	0.123^{-7}	0.194^{-5}	0.459^{-6}	0.495^{-5}	0.636^{-10}	0.6^{-15}	0.6^{-19}
6	232	0.116^{-2}	0.120^{-2}	0.826^{-5}	0.450^{-7}	0.710^{-5}	0.169^{-5}	0.280^{-4}	0.429^{-9}	0.4^{-14}	0.4^{-18}
8	309	0.172^{-2}	0.254^{-2}	0.178^{-4}	0.981^{-7}	0.155^{-4}	0.371^{-5}	0.815^{-4}	0.136^{-8}	0.1^{-13}	0.1^{-17}
10	386	0.215^{-2}	0.420^{-2}	0.297^{-4}	0.165^{-6}	0.261^{-4}	0.625^{-5}	0.169^{-3}	0.295^{-8}	0.3^{-13}	0.3^{-17}
12	463	0.242^{-2}	0.598^{-2}	0.426^{-4}	0.237^{-6}	0.375^{-4}	0.901^{-5}	0.284^{-3}	0.513^{-8}	0.5^{-13}	0.5^{-17}
14	540	0.253^{-2}	0.769^{-2}	0.550^{-4}	0.307^{-6}	0.486^{-4}	0.117^{-4}	0.418^{-3}	0.772^{-8}	0.8^{-13}	0.8^{-17}
16	617	0.254^{-2}	0.920^{-2}	0.660^{-4}	0.369^{-6}	0.585^{-4}	0.141^{-4}	0.557^{-3}	0.105^{-7}	0.11^{-12}	0.11^{-16}
18	694	0.246^{-2}	0.105^{-1}	0.751^{-4}	0.421^{-6}	0.668^{-4}	0.160^{-4}	0.692^{-3}	0.132^{-7}	0.13^{-12}	0.13^{-16}
20	772	0.233^{-2}	0.114^{-1}	0.822^{-4}	0.461^{-6}	0.731^{-4}	0.176^{-4}	0.814^{-3}	0.156^{-7}	0.16^{-12}	0.16^{-16}

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[a] Exponents are powers of ten by which number is to be multiplied.
[b] Estimated by assuming 1 x 10^{-5} atoms ^{257}Fm produced per atom ^{254}Es produced.
[c] Estimated by assuming 1 x 10^{-4} atoms ^{259}Md produced per atom ^{257}Fm produced.

B. PRODUCTION OF ACTINIDES BY ACCELERATORS

Production of actinides by accelerator-induced reactions has been an essential feature of the discovery of most of the synthetic actinide elements from neptunium (element 93) in 1940 to hahnium (element 105) in 1970.* The 60-in. cyclotron and the Heavy-Ion Linear Accelerator (HILAC) at the Ernest Orlando Lawrence Livermore Laboratory of the University of California have played the most significant roles. Some of the reactions leading to the discovery and identification of these elements are listed in Table III-8.

Other American high-energy accelerators useful for actinide synthesis are listed in Table III-9. These accelerators are also expected to find use in the production of superheavy elements by bombarding actinide targets with ions of calcium, argon, and similar projectiles. Some costs for existing installations are shown in Table III-9. Projected costs for a typical proton synchrotron, a Heavy-Ion Linear Accelerator, and the Intense Neutron Generator (ING) proposed by the Canadians are shown in Table III-10. The "typical proton synchrotron" is based on an approximate average of the accelerators listed in Table III-9. Costs for the Heavy-Ion Linear Accelerator are based simply on constructing a machine capable of accelerating a doubly charged ion beam of 10 milliamps. The HILAC at the University of California ion beam current is about 10 microamps, and is currently being modified to accelerate ions as heavy as uranium to 8.5 MeV per nucleon. Data for the Intense Neutron Generator were taken from AECL-2760.[60] For this comparison, it was assumed that the beam cross section for both the synchrotron and the Heavy-Ion Linear Accelerator is 0.1 cm^2 and that all of the neutrons produced by the ING would be available for target irradiation; all machines were compared at 100% annual operation.

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* Einsteinium, fermium, and ^{252}Cf were discovered in the debris of the 1952 thermonuclear test shot "Mike" at Eniwetok Atoll.

TABLE III-8

Some Accelerator Reactions That Produce Transuranium Elements

Element	Atomic Number	Reaction	Location	Date Reaction Observed	Accelerator
Np	93	$^{238}\mathrm{U}(n,\gamma)^{239}\mathrm{U} \xrightarrow[24\ \mathrm{m}]{\beta^-} {}^{239}\mathrm{Np}$	Univ. of Calif.	1940	60-inch Cyclotron
		$^{235}\mathrm{U}(2n,\gamma)^{237}\mathrm{U} \xrightarrow[6.8\ \mathrm{d}]{\beta^-} {}^{237}\mathrm{Np}$	Univ. of Calif.	1942	60-inch Cyclotron
Pu	94	$^{238}\mathrm{U}(d,2n)^{238}\mathrm{Np} \xrightarrow[2.1\ \mathrm{d}]{\beta^-} {}^{238}\mathrm{Pu}$	Univ. of Calif.	1940	60-inch Cyclotron
		$^{238}\mathrm{U}(n,\gamma)^{239}\mathrm{U} \xrightarrow[24\ \mathrm{m}]{\beta^-} {}^{239}\mathrm{Np} \xrightarrow[2.4\ \mathrm{d}]{\beta^-} {}^{239}\mathrm{Pu}$	Univ. of Calif.	1941	60-inch Cyclotron
Am	95	$^{241}\mathrm{Am}(n,\gamma)^{242}\mathrm{Am}$	Argonne	1945	60-inch Cyclotron
		$^{241}\mathrm{Am}(n,\gamma)^{242m}\mathrm{Am}$	Univ. of Calif.	1950	60-inch Cyclotron
Cm	96	$^{239}\mathrm{Pu}(\alpha,n)^{242}\mathrm{Cm}$	Univ. of Calif.	1944	60-inch Cyclotron
Bk	97	$^{241}\mathrm{Am}(\alpha,2n)^{243}\mathrm{Bk}$	Univ. of Calif.	1949	60-inch Cyclotron
Cf	98	$^{242}\mathrm{Cm}(\alpha,n)^{245}\mathrm{Cf}$	Univ. of Calif.	1950	60-inch Cyclotron
Es	99	$^{252}\mathrm{Cf}(n,\gamma)^{253}\mathrm{Cf} \xrightarrow[17.6\ \mathrm{d}]{\beta^-} {}^{253}\mathrm{Es}$	Univ. of Calif.	1954	60-inch Cyclotron
Fm	100	$^{252}\mathrm{Cf}(^{13}\mathrm{C},4p,4n)^{257}\mathrm{Fm}$	Univ. of Calif.	1964	HILAC
Md	101	$^{253}\mathrm{Es}(\alpha,2n)^{255}\mathrm{Md}$	Univ. of Calif.	1958	60-inch Cyclotron
		$^{253}\mathrm{Es}(\alpha,n)^{256}\mathrm{Md}$	Univ. of Calif.	1955	60-inch Cyclotron
		$^{252}\mathrm{Cf}(^{13}\mathrm{C},3p,5n)^{257}\mathrm{Md}$	Univ. of Calif.	1964	HILAC
		$^{238}\mathrm{U}(^{19}\mathrm{F},5n)^{252}\mathrm{Md}$	USSR - Dubna	1965	10-GeV Proton Synchrotron
No	102	$^{246}\mathrm{Cm}(^{12}\mathrm{C},4n)^{254}\mathrm{No}$	Univ. of Calif.	1958	HILAC
		$^{252}\mathrm{Cf}(^{13}\mathrm{C},2p,8n)^{255}\mathrm{No}$	Univ. of Calif.	1961	HILAC
		$^{238}\mathrm{U}(^{22}\mathrm{Ne},4n)^{256}\mathrm{No}$	USSR - Dubna	1963	10-GeV Proton Synchrotron
		$^{242}\mathrm{Pu}(^{18}\mathrm{O},4n)^{256}\mathrm{No}$	USSR - Dubna	1963	10-GeV Proton Synchrotron
Lw	103	$^{243}\mathrm{Am}(^{18}\mathrm{O},5n)^{256}\mathrm{Lw}$	USSR - Dubna	1965	
		$^{250}\mathrm{Cf}(^{11}\mathrm{B},4n)^{257}\mathrm{Lw}$	Univ. of Calif.	1961	HILAC
		$^{251}\mathrm{Cf}(^{11}\mathrm{B},5n)^{257}\mathrm{Lw}$	Univ. of Calif.	1961	HILAC
		$^{252}\mathrm{Cf}(^{11}\mathrm{B},6n)^{257}\mathrm{Lw}$	Univ. of Calif.	1961	HILAC
		$^{250}\mathrm{Cf}(^{10}\mathrm{B},3n)^{257}\mathrm{Lw}$	Univ. of Calif.	1961	HILAC
		$^{251}\mathrm{Cf}(^{10}\mathrm{B},4n)^{257}\mathrm{Lw}$	Univ. of Calif.	1961	HILAC
		$^{252}\mathrm{Cf}(^{10}\mathrm{B},5n)^{257}\mathrm{Lw}$	Univ. of Calif.	1961	HILAC
Rf	104	$^{249}\mathrm{Cf}(^{12}\mathrm{C},4n)^{257}\mathrm{Rf}$	Univ. of Calif.	1969	HILAC
		$^{249}\mathrm{Cf}(^{13}\mathrm{C},3n)^{259}\mathrm{Rf}$	Univ. of Calif.	1969	HILAC
Ha	105	$^{249}\mathrm{Cf}(^{15}\mathrm{N},4n)^{260}\mathrm{Ha}$	Univ. of Calif.	1970	HILAC

TABLE III-9

USA High-Energy Accelerators[58,59]

Location	Name	Year Started Up	Peak Energy, GeV	Operating Energy, GeV	Operating Flux, particles/sec	Pulse/sec	Facility Cost, $\$10^6$	Annual Operating Cost, $\$10^6$	Annual Research Cost, $\$10^6$
Proton Synchrotrons ($E > 1$ GeV)[a]									
Argonne	Zero Gradient Synchrotron	1963	12.7	12.7	5×10^{11}	0.5	50	-	-
Brookhaven	Cosmotron[b]	1952	2.96	2.85	2.5×10^{11}	0.2-0.5	10.5	-	-
Brookhaven	Alternating Gradient Synchrotron	1960	33	28	10^{12}	0.4	30.65	-	-
Lawrence	Bevatron	1954	6.2	6.2	2.2×10^{12}	0.2	30	5	10
Princeton	Princeton, Pennsylvania Accelerator	1963	3	3	6×10^{11}	18.5	30	4.7	-
Weston	National Accelerator[c] Synchrotron	1974	200	200	1.5×10^{13}	0.25	240	-	-
Electron Synchrotrons ($E > 10^9$ eV)									
Cambridge	Cambridge Electron Accelerator	1962	6.28	6	10^{12}	60	25	3.5	-
Caltech	Caltech Electron Synchrotron	1952	1.53	1.5	2×10^9	2	1.6[d]	1.6	0.6
Cornell	Electron Synchrotron	1964	2.1	2.1	5×10^{10}	30	0.3[d]	0.25	0.35
Cornell	Electron Synchrotron	1967	10	10	10^{11}	60	11.5	1	1.2
Electron Linear Accelerators ($E > 10^9$ eV)									
Stanford	Mark III Electron Linear Accelerator	1950/52	1.2	1	1.5×10^{13}	60	7	0.8	1.6
Stanford	Stanford Linear Accelerator Center	1966	20.16	12	10^5	360	114	11.3	10
Proton Linear Accelerators ($E > 500$ MeV)									
Los Alamos	Los Alamos Meson Physics Facility[c]	1972	800	800	6×10^{15}	120	55	-	-

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a. GeV = 10^9 eV.
b. Retired 12/66.
c. Under construction.
d. Accelerator cost only.

TABLE III-10

Projected Annual Costs for Accelerators

	Typical Proton Synchrotron	Heavy-Ion Linear Accelerator	Proposed Intense Neutron Generator
Beam Intensity	5×10^{12} protons/sec	1×10^{16} ions/sec	1×10^{19} n/sec[a]
Capital Cost, $\$10^6$	30	50	128
Operating Cost, $\$10^6$/yr	5	5	16
Cost/Mole Particles Accelerated, $	2.5×10^{10}	15×10^6	4×10^4

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[a] Neutrons generated by irradiating a Pb-Bi target with a 1-GeV proton beam.

C. PRODUCTION OF ACTINIDES BY THERMONUCLEAR EXPLOSIONS

Another route to the actinides is through thermonuclear explosions. Heavier actinides are formed by nearly simultaneous addition of many neutrons to uranium or other heavy element nuclei forming nuclides as high as ^{257}U. These decay rapidly by beta emission to beta-stable isotopes as heavy as ^{257}Fm.

On July 16, 1969, a target of 8.8 g of ^{232}Th and 17.8 g of ^{238}U was exposed to a neutron fluence of 45 moles of n/cm^2 (2.7 x 10^{25} n/cm^2) from an 1800-ft-deep underground thermonuclear explosion ("Hutch"). An estimated 40 mg of ^{250}Cm, 10 mg of ^{252}Cf, and 0.25 mg of ^{257}Fm were produced. ^{250}Cm cannot be made by sequential neutron capture in a reactor because of the 64-min half-life of ^{249}Cm. No isotopes of mendelevium, nobelium, lawrencium, rutherfordium, or hahnium were found; nor were closed neutron shell nuclides such as ^{46}Ar, ^{35}P, ^{34}Si, or ^{62}Fe detected.

Calculated yields from a target containing actinium, thorium, protactinium, and uranium, exposed to neutron fluences ranging from 10 to 50 moles/cm^2, are shown in Figure III-6. For comparison, the explosion exposure of 45 moles of n/cm^2 developed in less than a microsecond would require a high-flux reactor operating continuously at a flux of 3 x 10^{15} n/(cm^2-sec) nearly 300 years to develop.

Costs for preparing a site and detonating a thermonuclear explosion are listed in Table III-11. These costs are based on project "Gasbuggy," detonated on December 10, 1967, and are estimated at $2.6 million for a 26-kiloton device. The test was made to evaluate the feasibility of stimulating natural gas flow from underground natural storage by thermonuclear explosions. The costs do not reflect any research or experimental work, purchase of land, or geological studies. The explosive cost is the AEC stated price.

Table III-12 estimates the unit cost of producing ^{252}Cf from a series of 30 explosions over a period of 10 years assuming production of 10 grams per shot and 50% recovery of the ^{252}Cf. Although still highly problematical because of the need for developing optimum shot conditions and nuclide recovery techniques, the $1 to $4 per microgram of ^{252}Cf estimate is competitive with the current AEC selling price of $10 per microgram.

Current facilities at Ernest Orlando Lawrence Livermore Laboratory can process up to 500 kg of shot-melted rock over a period of 10 weeks, using the process shown in Figure III-7. This is equivalent to only 3 tons per year. Larger rock processing facilities would have to be constructed before production of heavy nuclides by thermonuclear explosions would be practical. The process consists of an acid leach of the dried and crushed rock using HF and HCl, liquid-liquid extraction of the actinides into di(2-ethylhexyl) pyrophosphoric acid, hydrolysis of the pyrophosphoric acid, back extraction of the actinides into an HCl-Fe(III) solution, and concentration of the aqueous solution by evaporation.

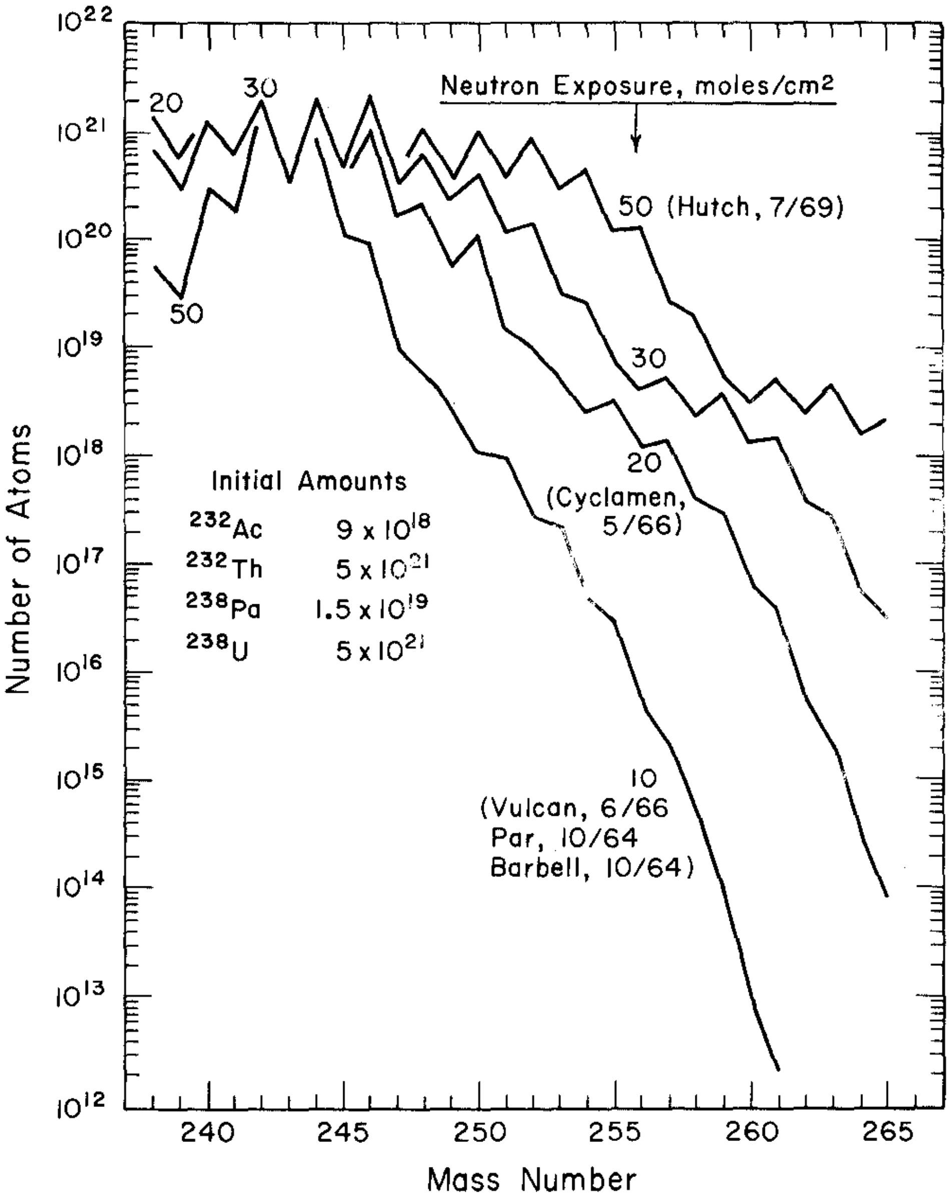


FIGURE III-6. Calculated Mass-Yield Curve for a ^{238}U, ^{238}Pa, ^{232}Th, and ^{232}Ac Combination Target in an Underground Nuclear Explosion[65]

TABLE III-11

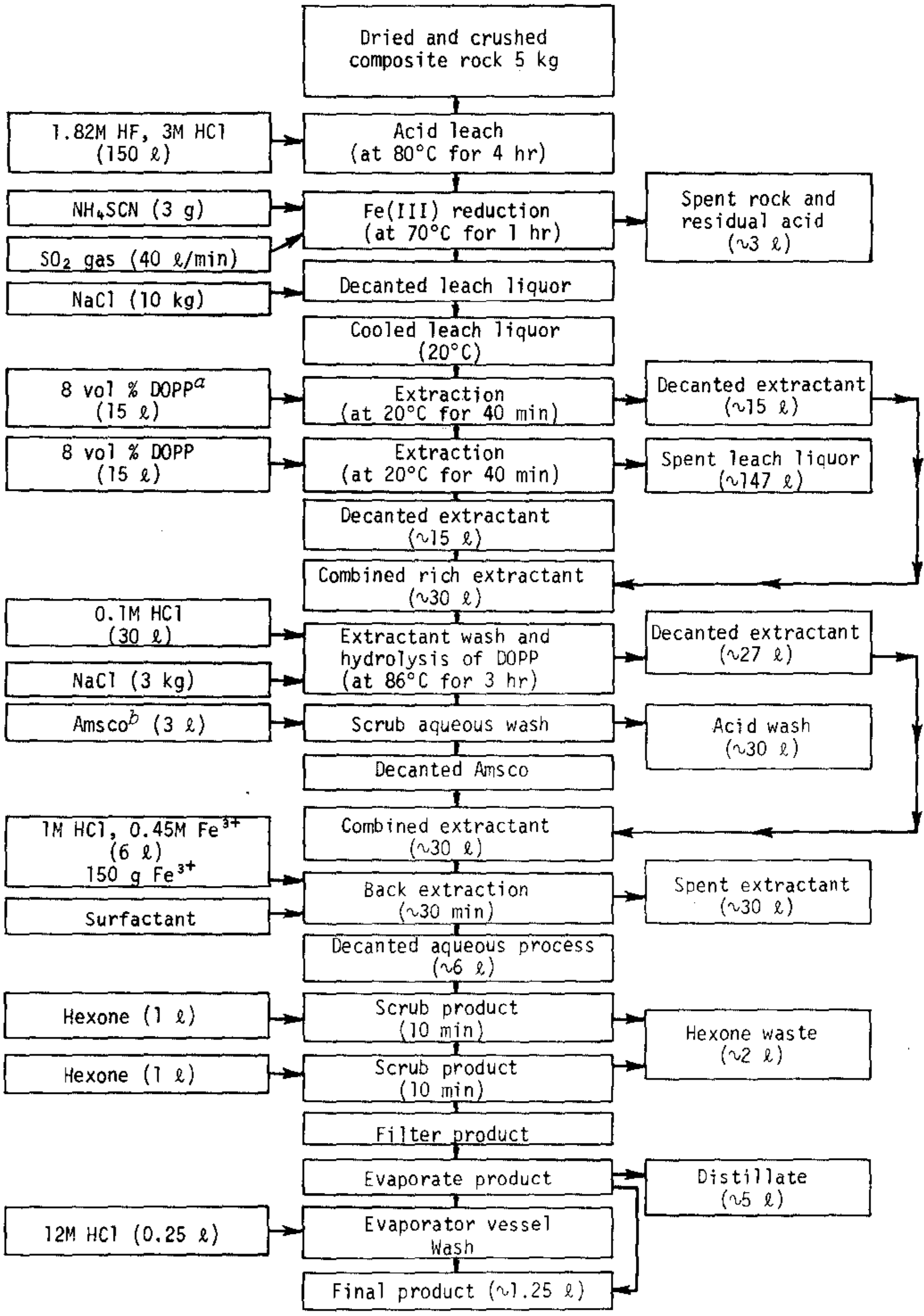
Costs of Project "Gasbuggy" for a 26-Kiloton Device[67]

	Thousands of Dollars
Field Construction and Support	1,484
Logistical Support and Maintenance	215
Operational Support	23
Safety	326
Security	59
Nuclear Explosive	395
Administration	60
Total	2,562

TABLE III-12

Estimated ^{252}Cf Costs from Thermonuclear Explosions
(10-year program producing 150 g of ^{252}Cf)

30 Explosions at $500,000 Each	$ 15,000,000
Site Preparation and Monitoring	15,000,000
Mining and Ore Processing Equipment	10,000,000
Actinide Separations and Processing Plant	10,000,000
Mining and Separations Costs	60,000,000
	$110,000,000
^{252}Cf Production (10 g/shot, 10-50% recovery)	30-150 g
^{252}Cf Unit Cost	$1-$4/μg



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a. DOPP - di(2-ethylhexyl) pyrophosphoric acid
b. Amsco - solvent similar to kerosene

FIGURE III-7. Flow Diagram of Lawrence Livermore Laboratory Rock Processing Plant[66]

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IV. SEPARATION OF ACTINIDES

The actinide elements are separated chemically by a combination of solvent extraction and ion exchange techniques; however, a number of other techniques, including fluoride-volatility and pyrochemical processes, are being developed. Mixed-actinide separations have been required primarily in connection with the reprocessing of irradiated fuel from nuclear power plants. Existing fuel reprocessing plants include the AEC installations at the Idaho National Reactor Testing Station, Savannah River, Richland, and the commercial Nuclear Fuel Service plant at West Valley, N. Y. All of these plants fundamentally use the Purex solvent extraction process. All U. S. commercial plants in existence, under construction, or being planned are listed in Table IV-1.

The primary purpose of these fuel reprocessing plants is to separate plutonium, uranium, and fission products. Separations plants for the higher actinides are being planned as additions

TABLE IV-1

U. S. Commercial Plants for Reprocessing Nuclear Reactor Fuels

Location	Annual Capacity, metric tons	Startup Date	Process
Nuclear Fuel Services Plant West Valley, N. Y.	300	1966	Purex process. Fuel pieces are cut and dissolved. Solution is decontaminated and separated into Pu and U streams by solvent extraction using pulsed columns. Pu is purified by ion exchange, concentrated, and shipped. U is purified by second solvent extraction cycle.
General Electric Midwest Fuel Recovery Plant Morris, Ill.	300	1972	Aquaflor process. Head end is similar to Purex; Pu, Np, and U are partitioned by anion exchange contactors; U is calcined to UO_3, fluorinated to UF_6, filtered, and condensed.
Allied Nuclear Fuel Reprocessing Plant Barnwell, S. C.	1800	1973	Modified Purex process using centrifugal contactors and pulse columns for solvent extraction.
Atlantic Richfield ARCO Plant Leeds, S. C.[a]	2200	1976	Modified Purex process adapted to recover Np.
High Temperature Gas Reactors at Peach Bottom Delta, Penn. and Fort St. Vrain, Col.	-	Study	Graphite fuel elements. Fuel elements are crushed and burned in fluidized bed of alumina; products are leached with HNO_3; Th and U are separated and decontaminated by Thorex solvent extraction process.
Light Metal Fast Breeder Reactors	-	Study	Modified Purex; or Volatility process. Fuel is declad mechanically and converted to UF_6 and PuF_6 in fluorinator. Fuel is decontaminated by fractional condensation, U and Pu are separated by thermal decomposition of PuF_6 and purified by fluorination and distillation; or Pyrochemical process: Fuel is declad by dissolving with liquid zinc. U is precipitated from molten $CaCl_2$-CaF_2 salt with liquid Cu-Mg alloy. Pu is extracted from the Cu-Mg alloy to a Zn-Mg alloy and distilled to separate Pu. U is removed from equipment by alloying with Fe.

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[a] Cancelled.

to the fuel reprocessing plants. Specific facilities for higher actinide separations exist at Oak Ridge based on a tertiary amine extraction (Tramex) process and are under construction at Savannah River based on a pressurized ion exchange process.

A. SOLVENT EXTRACTION SEPARATION OF PLUTONIUM AND URANIUM

The Savannah River Purex process uses tributyl phosphate (TBP) in kerosene to strip plutonium from uranium in dilute nitric acid solution. The organic and aqueous phases are contacted in multistage mixer-settlers or centrifugal contactors. The process, described in Figure IV-1, consists of several steps:

- The irradiated, cooled slugs are charged to a dissolver where the aluminum jackets are removed with a solution of NaOH and $NaNO_3$, and the uranium is dissolved in nitric acid.
- The solution is clarified with gelatin and partially decontaminated of fission products by contact with a precipitate of manganese dioxide in a "head-end" treatment step.
- The clarified uranium solution is concentrated; then sodium nitrite is added to reduce plutonium to the tetravalent state to facilitate extraction into the organic phase.
- In the first solvent extraction cycle, the solution is contacted with TBP-kerosene to extract the plutonium and uranium into the organic phase; the organic phase is scrubbed to remove fission products; and the plutonium and uranium are stripped back into separate aqueous streams. The direction of plutonium transfer is controlled by valence adjustment; the direction of uranium is controlled by uranium and nitric acid concentration.
- The plutonium and uranium streams are further decontaminated from fission products by separately repeating the extraction-scrub-strip process in second solvent extraction cycles.
- The plutonium is concentrated by ion exchange and precipitated with hydrofluoric acid, and the dried precipitate is reduced with calcium to plutonium metal.
- The purified aqueous uranium nitrate stream is concentrated by evaporation, passed through silica gel for additional decontamination and removal of suspended solids, dehydrated, and calcined to the oxide.
- The aqueous stream from the first cycle solvent extraction contains rare earths as well as fission products. Separation of americium, curium, and heavier actinides is accomplished by ion exchange (page 38).

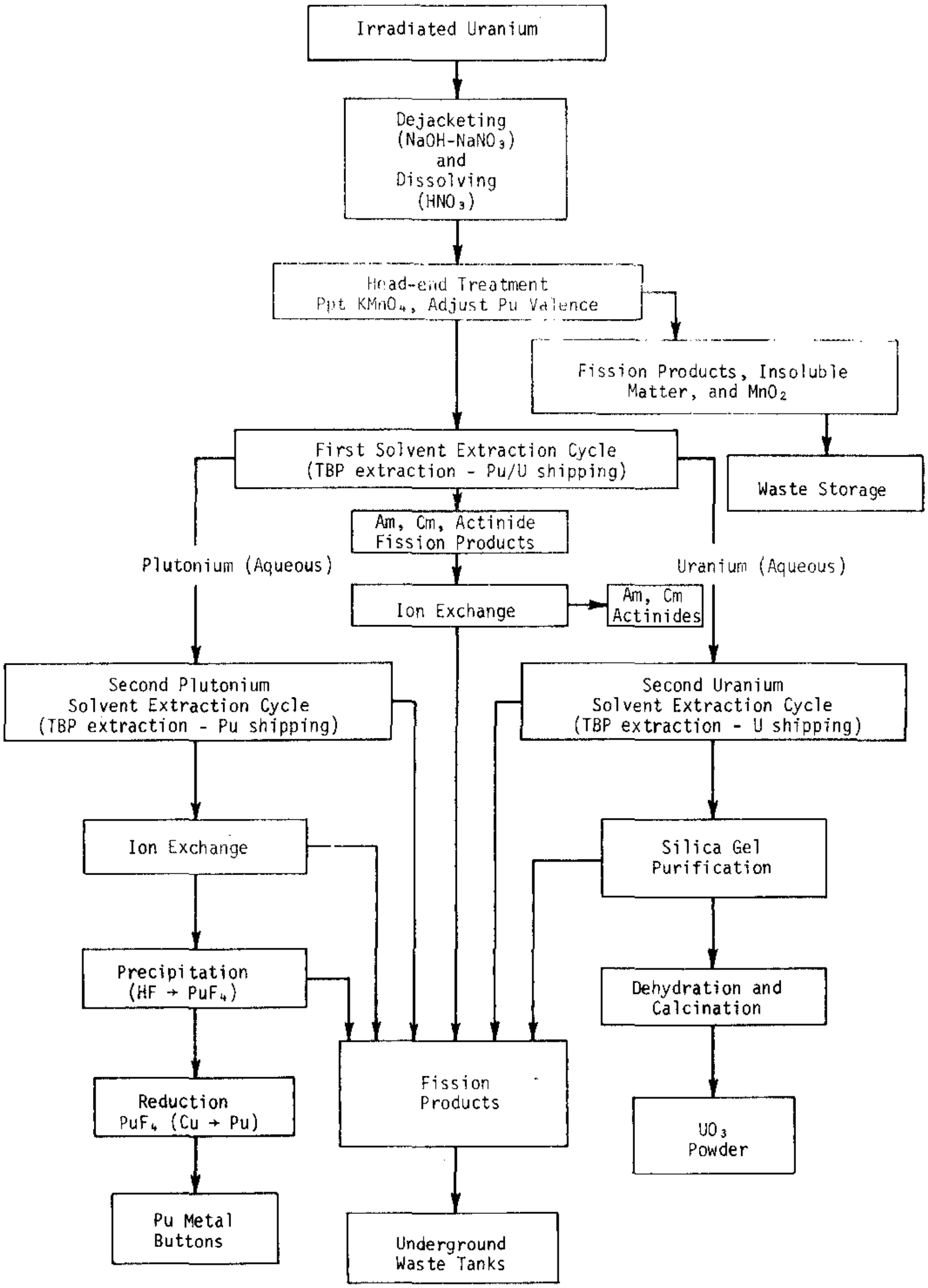

FIGURE IV-1. Uranium Fuel Processing by Purex Process at the Savannah River Plant

B. SOLVENT EXTRACTION SEPARATION OF TRANSPLUTONIUM ELEMENTS

Americium, curium, and rare earths are recovered at Savannah River by the process outlined in Figure IV-2. In general, aluminum jacket removal, dissolution of the actinides, and clarification of the product solution is similar to that used in the Purex process (page 36).

In the transplutonium recovery process, the aqueous stream from the first solvent extraction cycle contained the actinides and was concentrated by evaporation; a lanthanum carrier and $NaHSO_4$ were added; the sulfate ion concentration and pH were adjusted; and the double sulfates of the actinides were precipitated by digestion. The sulfates were converted to hydroxides, dissolved in nitric acid, and shipped to ORNL for processing by the Tramex ion exchange process.

In later transplutonium recovery programs, the actinides were extracted from the aqueous stream of the first solvent extraction cycle with TBP and then were stripped with dilute nitric acid. This aqueous stream was further purified, and the actinides were separated by ion exchange, using either ORNL's Tramex or SRP's pressurized ion exchange processes.

In the ion exchange purification, all of the elements in the tripositive oxidation state are reacted with an organic polymer that has exchangeable cations such as "Dowex" 50W-X8* (copolymer of 8% cross-linked divinylbenzene in polystyrene with sulfonic acid groups). The individual actinides are sequentially eluted by a solution of diethylenetriaminepentaacetic acid (DTPA), which forms complex ions with the actinides. The elution sequence, which depends on a balance between adherence to the resin and the stability of the complex ion formed with the eluting agent, may be correlated with ionic radius and atomic number.

Actinide ions in the III, IV, and V oxidation states can be adsorbed by other cation exchange resins and can be eluted by other anions such as chloride, nitrate, citrate, lactate, α-hydroxyisobutyrate, or ethylenediaminetetraacetate.

The reverse procedure can also be used with anion exchange resins where the actinides are present as complex ions. Resins such as "Dowex"-1 (copolymer of styrene and divinylbenzene with quaternary ammonium groups) have been used.

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* Registered trademark of Dow Chemical Co.

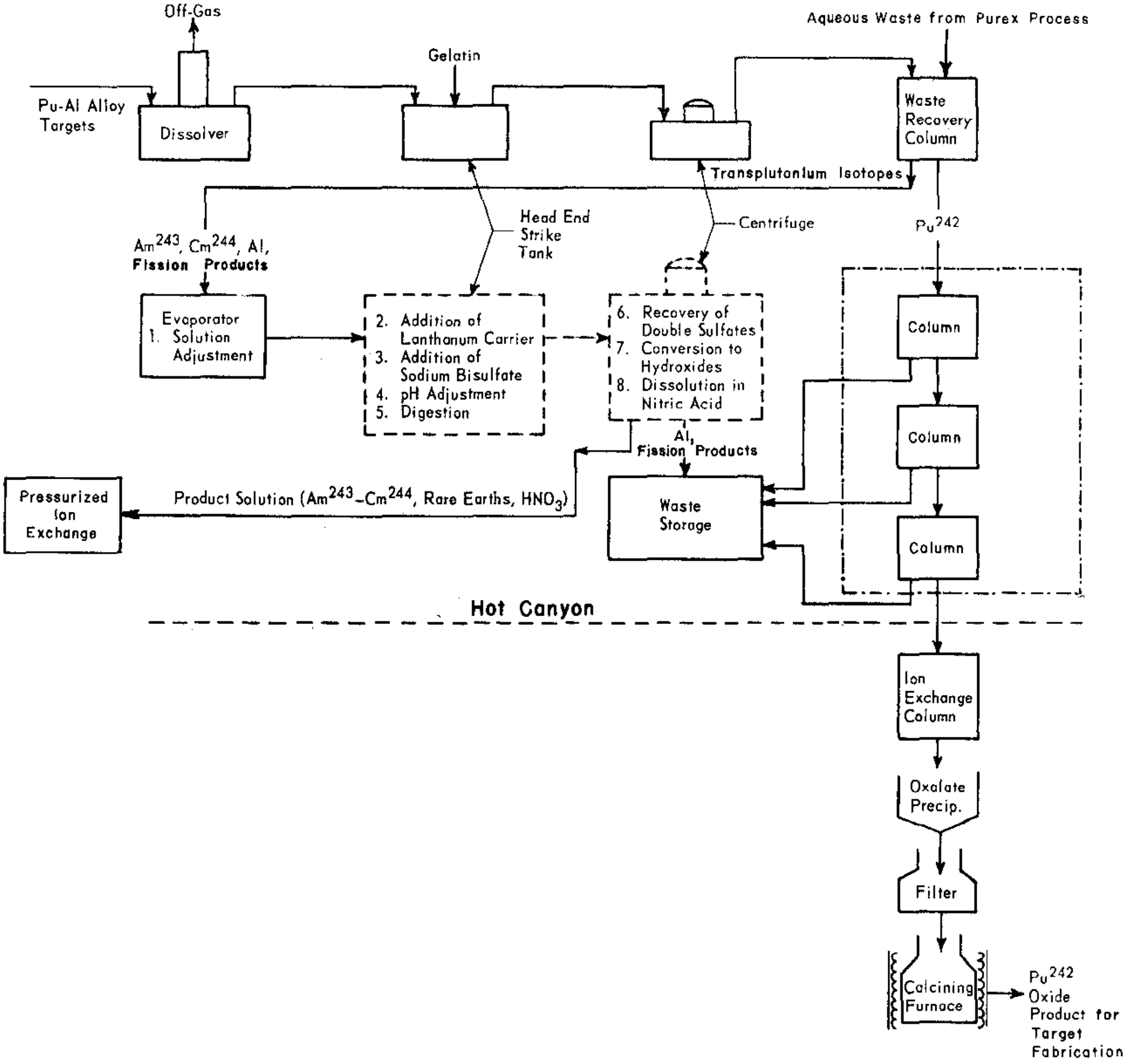


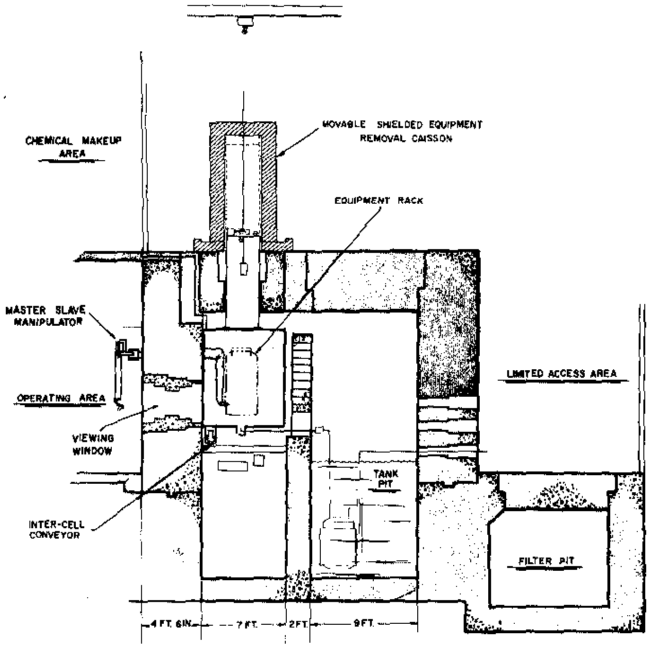
FIGURE IV-2. Flowsheet of Savannah River Plant Transplutonium Process

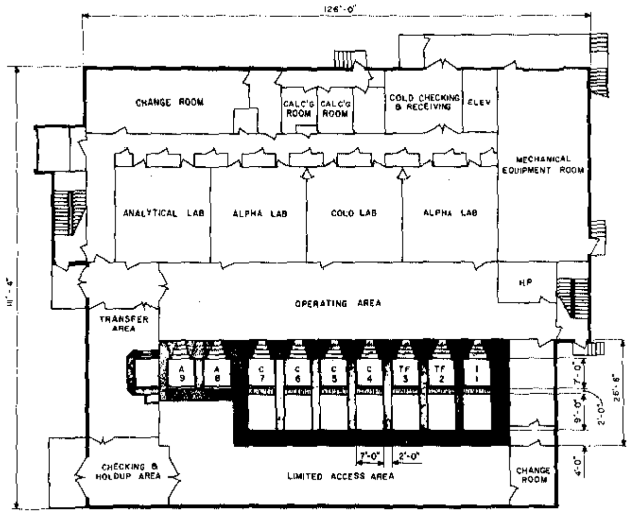
C. TRANSURANIUM PROCESSING PLANT AT OAK RIDGE NATIONAL LABORATORY

Currently, much of the chemical processing of transplutonium isotopes is done in the Transuranium Processing Plant (TRU) at ORNL. This facility, completed in 1965, contains nine shielded process cells and supporting laboratories, as illustrated in Figure IV-3. Four cells contain chemical processing equipment for dissolution, solvent extraction, and precipitation; three cells contain equipment for the remote preparation and inspection of irradiated targets; and two cells are used for remote analytical operations. The Tramex process, outlined in Figure IV-4, consists of:

- Dissolution of aluminum target jackets with NaOH and $NaNO_3$, followed by dissolution of the actinide oxides in HCl.
- Extraction of plutonium with di(2-ethylhexyl) phosphoric acid (HDEHP) in diethylbenzene.
- Recovery of transplutonium elements by extraction with HDEHP, after neutralizing the acid and stripping transplutonium elements from HDEHP with HCl.
- Partition of americium and curium from transcurium isotopes with LiCl-based anion exchange.
- Decontamination of americium and curium by repeated oxalate precipitation, followed by calcination and fabrication into targets for further irradiation.
- Purification of berkelium, californium, einsteinium, and fermium by precipitation with NH_4OH and separation with high-pressure ion exchange using α-hydroxyisobutyrate elutriant.
- Separation of berkelium, californium, einsteinium, and fermium by high-pressure ion exchange chromatography.

This cross section of a typical cell at the TRU facility shows the cubicle containing an equipment rack, the tank pit behind and below the cubicle, the shielding window, manipulators, and the movable equipment transfer case.





The two-story TRU facility is divided into a laboratory wing and a process area which includes the shielded cell bank. Remote target fabrication is performed in cells 1, 2 and 3. Chemical separations processes are done in cells 4-7, while 8 and 9 are for analytical chemistry work.

FIGURE IV-3. Oak Ridge National Laboratory Transuranium Processing Plant

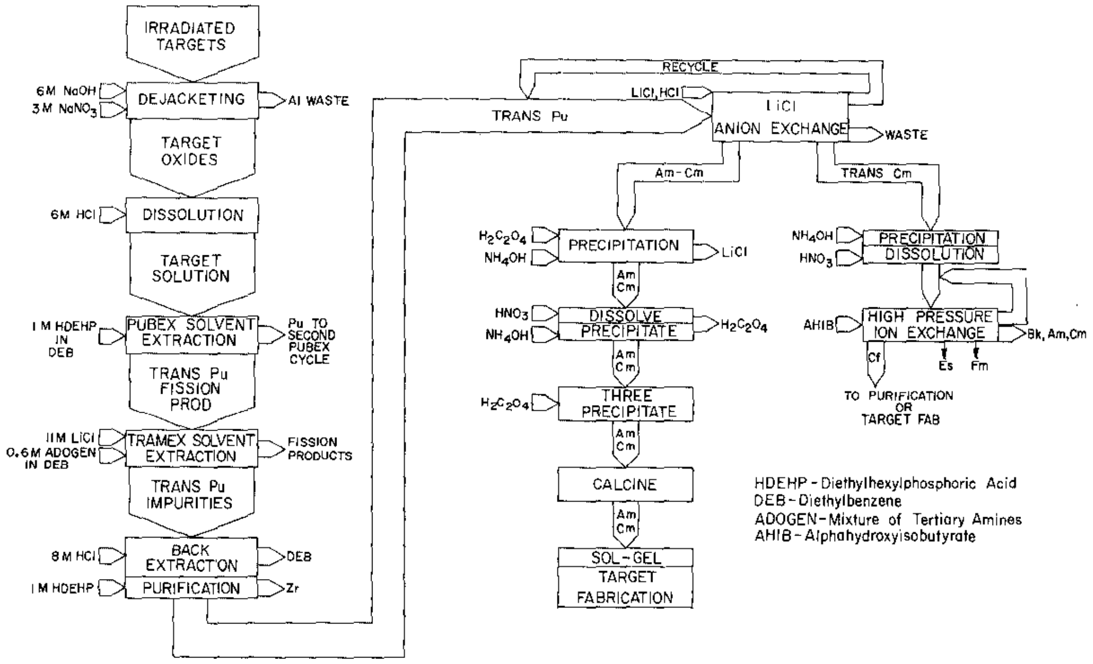
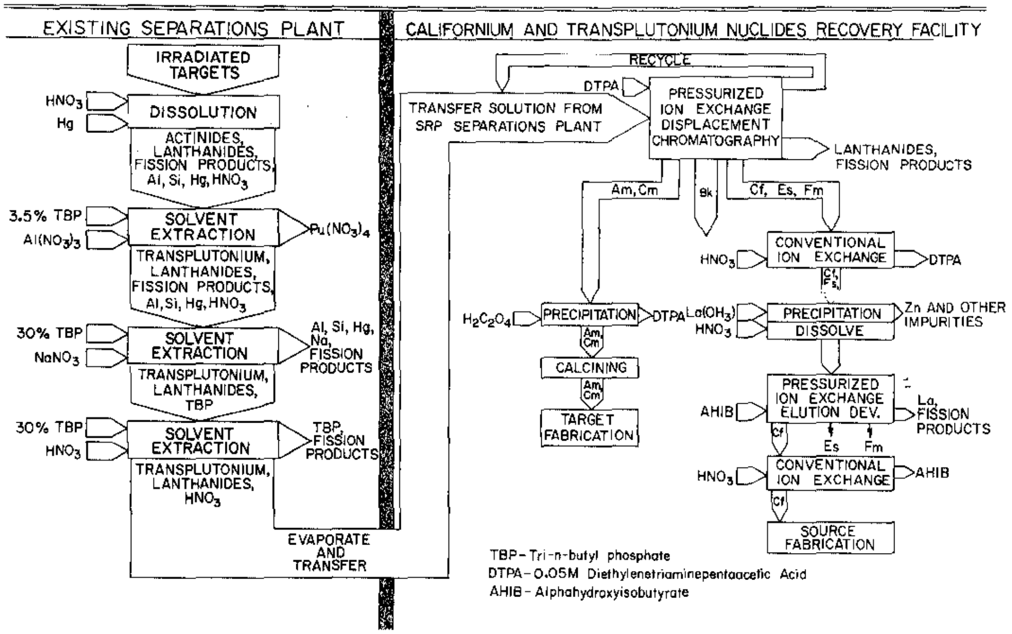


FIGURE IV-4. Tramex Process Used at Transuranium Processing Plant

D. MULTIPURPOSE PROCESSING FACILITY AT SAVANNAH RIVER PLANT

The Savannah River Plant is constructing a Multipurpose Processing Facility (MPPF) to process transplutonium elements on a production scale. The facility is being installed in five of the "hot" process canyon modules in the separations plant, as shown in Figure IV-5.



TBP- Tri-n-butyl phosphate
DTPA- 0.05M Diethylenetriaminepentaacetic Acid
AHIB- Alphahydroxyisobutyrate

FIGURE IV-5 Savannah River Process for Transplutonium Separations

Target slugs and target housing tubes from plutonium irradiation programs will be dissolved and processed through the Purex TBP extraction process in present plant equipment to separate plutonium from transplutonium isotopes and lanthanides, as shown in Figure IV-I. Target slugs of americium and curium, however, will be dissolved in the MPPF dissolver. The aqueous actinide-lanthanide solution will be transferred to the new MPPF area where americium-curium fractions, berkelium-californium fractions and other transcalifornium elements will be separated from the' lanthanides and decontaminated by pressurized ion exchange chromatography.

Americium and curium oxalates will be precipitated from the ion exchange product fractions by addition of oxalic acid. The americium-curium oxalates will be calcined to oxides and fabricated into targets for irradiation to make transcurium isotopes.

Similarly, the transcurium elements will be purified by sorption onto cation exchange resin and removal of DTPA with a wash of dilute nitric acid. Further concentration and separation will be accomplished by hydroxide precipitation with lanthanum hydroxide as a carrier. The hydroxides will be dissolved with dilute nitric acid and fed to a second pressurized ion exchange system to separate californium, einsteinium, and fermium with ammonium alphahydroxyisobutyrate (AHIB). The AHIB will then be removed by cation exchange.

Interchangeable modular equipment racks designed for specific process steps will make the MPPF a versatile new separations plant. Sets of racks will provide for the following exclusive annual capacities:

- ^{252}Cf up to 20 g/yr and the associated curium-americium target material up to 25 kg/yr; or
- Am-Cm mixtures up to 23 kg/yr from plutonium burning; or
- ^{244}Cm up to 10 kg/yr; or
- ^{238}Pu (<30 ppb ^{236}Pu) up to 10 kg/yr from irradiation of ^{241}Am; or
- Fission product ^{147}Pm up to 6.5 kg/yr; or
- Fission product palladium and rhodium up to 10 kg/yr; or
- Fission product ^{137}Cs up to 20 MCi and ^{90}Sr up to 5 MCi/yr; or
- Fission product fractions up to a total of 1 kg/yr of dysprosium, holmium, thulium, terbium, and europium.

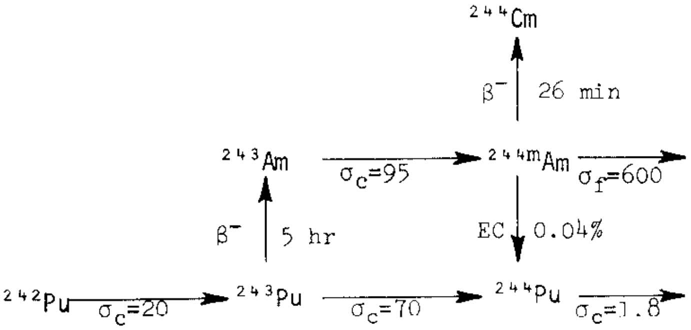
Berkelium, einsteinium, and fermium will be separated in a two-step pressurized ion exchange method. In the first step, four primary separations will be made: americium, curium, berkelium, and californium-einsteinium-fermium. In the second step, berkelium will be further purified, and californium will be separated from einsteinium-fermium. A process for final separation between einsteinium and ferium requires further development.

V. PRINCIPAL METHODS OF MAKING ISOTOPICALLY PURE ACTINIDES

This section is primarily concerned with the production of nuclides from ^{244}Pu through ^{257}Fm by thermal neutron captures in reactors, and with production of ^{258}Md and higher nuclides by heavy-ion bombardment in accelerators. Production by thermonuclear explosions is indicated for a few nuclides.

A. PLUTONIUM-244 (8.3×10^7 yr)

^{244}Pu is chiefly produced by reactor irradiation of plutonium. The best target isotope is ^{242}Pu, for which the production sequence is shown below. (The projected route from ^{242}Pu to ^{104}Rf is given in Appendix B.) Direct neutron capture is the primary source of ^{244}Pu, although a very small fraction is formed by electron capture (EC) in ^{244m}Am.



The measured isotopic composition of ^{239}Pu irradiated to an exposure of 1.4×10^{22} n/sec is shown in Table V-1: a calculated extension of ^{242}Pu irradiated to an exposure of 2×10^{23} n/sec is shown in Table III-7. Irradiation of ^{239}Pu yields primarily ^{242}Pu and ^{244}Pu.

TABLE V-1

Isotopic Composition of Irradiated ^{239}Pu

Exposure	^{242}Pu Abundance, atom %	^{244}Pu Abundance, atom %	^{244}Pu/^{242}Pu
0.4×10^{22}	34.1	0.0018	0.53×10^{-4}
1.1×10^{22}	96.3	0.037	3.84×10^{-4}
1.4×10^{22}	98.8	0.052	5.26×10^{-4}

Isotopically pure samples of ^{244}Pu can be obtained by irradiation of ^{243}Am to form ^{244}Pu via electron capture in ^{244m}Am, followed by chemical separation of the americium, plutonium, and curium isotopes. Americium from power reactors, however, will be about a 50/50 mixture of ^{241}Am/^{243}Am, producing an isotopic mixture of plutonium isotopes due to formation of ^{242}Cm from ^{243}Am followed by alpha decay to ^{238}Pu. Smaller amounts of ^{239}Pu and ^{240}Pu will also be formed by the alpha decay of ^{243}Cm and ^{244}Cm.

^{244}Pu is also available in good yield from thermonuclear explosions by successive neutron addition to ^{238}U to form ^{244}U, followed by beta decay to ^{244}Pu. From Figure III-6, if a mixed target contains 5 x 10^{21} atoms of ^{238}U, the production of ^{244}Pu is of the order of 2 x 10^{21} atoms. Recovery of the product depends on efficiently processing large volumes of salt or rock.

Plutonium is separated from irradiated uranium through solvent extraction and ion exchange processes as discussed on page 36. Individual plutonium nuclides can be separated by either electromagnetic or gas centrifuge methods. The shielded calutron facility at ORNL is available for separating small amounts of isotopes. A typical single pass in this facility, for example, separated 7.6 mg of plutonium containing 76.8% ^{244}Pu from 30.2 g of plutonium containing about 0.6% ^{244}Pu. The distribution of other plutonium isotopes is shown in Table V-2. For ^{244}Pu, the feed contained 0.568% or 0.171 gram; the product contained 76.80% or 0.0058 gram. The remaining 0.1657 gram is collected as feed for a second pass.

TABLE V-2

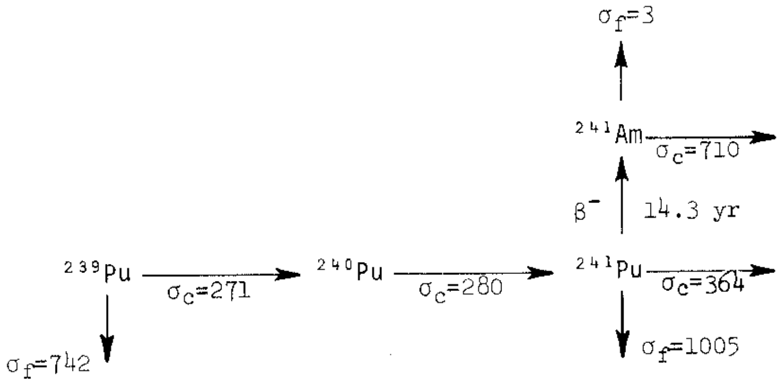
Separation of Plutonium Isotopes in an ORNL Electromagnetic Calutron

	Feed	Product
Total weight, g	30.2	0.0076
^{238}Pu, %	0.002	0.0003
^{239}Pu, %	0.0045	0.0029
^{240}Pu, %	1.7525	0.159
^{241}Pu, %	0.1031	0.020
^{242}Pu, %	97.57	23.02
^{244}Pu, %	0.568	76.80

^{244}Pu is of primary interest as a heavy, neutron-rich accelerator target for the production of superheavy elements. It is also useful for studying the physical and chemical properties of plutonium because it has the longest half-life of any of the plutonium isotopes.

B. AMERICIUM-241 (433 yr)

^{241}Am is formed by beta decay of ^{241}Pu, which is produced by the irradiation of ^{239}Pu, as shown in Figure III-1. Typical composition after an exposure of 3×10^{21} n/cm^2 is shown in Table III-6.



Large quantities of ^{241}Am and ^{243}Am are currently being produced by power reactors in an approximately 50/50 mixture of these two nuclides. Production of ^{241}Am from this source is expected to reach over 100 kg/year in 1977 and to increase to nearly 600 kg/year in 1983, as shown in Table V-3. Annual availability of ^{241}Am and ^{243}Am from various reactor combinations and from reworking the plutonium stockpile is shown in Table III-4.

TABLE V-3

Estimated Annual Production of Am From Power Reactors

Fiscal Year	^{241}Am, kg	^{243}Am, kg
1977	115	60
1978	180	145
1979	250	245
1980	285	295
1981	370	380
1982	475	485
1983	565	590

Americium can be separated and purified by Purex solvent extraction and ion exchange facilities available at Savannah River, Richland, Oak Ridge National Laboratory, and Rocky Flats. At Rocky Flats, americium is available as a ^{241}Pu beta-decay product from the normal plutonium rework stream. ^{241}Pu beta decays to ^{241}Am with a 14.3 yr half-life. It is convenient to set aside a sample of plutonium with a high ^{241}Pu content and periodically "milk" it by chemically separating the americium and plutonium, and recovering the plutonium for additional aging. For example, as shown in Figure V-3, a 10-g sample of ^{241}Pu will produce about 4 g of ^{241}Am after about 10 years of decay.

Some estimated capacities and recovery costs quoted are listed in Table V-4.

TABLE V-4

Americium Capability at Rocky Flats[29]

Work Schedule	kg/yr	Estimated Americium Cost[a]
5 days, 3 shifts	3.6	$140/gram
7 days, 3 shifts	5.0	-

[a] Plutonium recovery costs are not charged to the americium product. If all plutonium rich in americium were sent to recovery for chemical separation, the costs for plutonium processing would then be charged to americium. These costs are estimated at $500/g Am. The variation is expected to be between $140 and $700.

Figure V-1 shows a block diagram of the Rocky Flats ion exchange process for the recovery of ^{241}Am. After ion exchange purification, americium is precipitated as the oxalate, calcined to the oxide, and reduced with lanthanum at 1500°C. Americium metal is distilled off at 1600°C. If the oxide is 95.5% pure, the metal will be about 99.8% pure. This process recovers about 93% of the available americium.

^{241}Am has important research and industrial applications. The 60-keV gamma ray that accompanies its alpha decay is used as a standard in nuclear spectroscopy for calibrating the energy scale and geometry of counting equipment. Industrial uses include measurement of fluid density, as a thickness gage, fuel gage, and distance sensing device. Other uses are eliminating static, detecting smoke, radiography, oil and water well logging, X-ray fluorescence, and scanning thyroid gland functioning. The isotope can also be used as a low specific power source producing 0.1 w/g. The specific alpha activity is 7.0×10^{12} dpm/g (equivalent to 3.2 Ci/g). The 5.49-Mev alpha particle is useful for initiating α,n reactions to produce neutrons via a ^{241}Am-Be source.

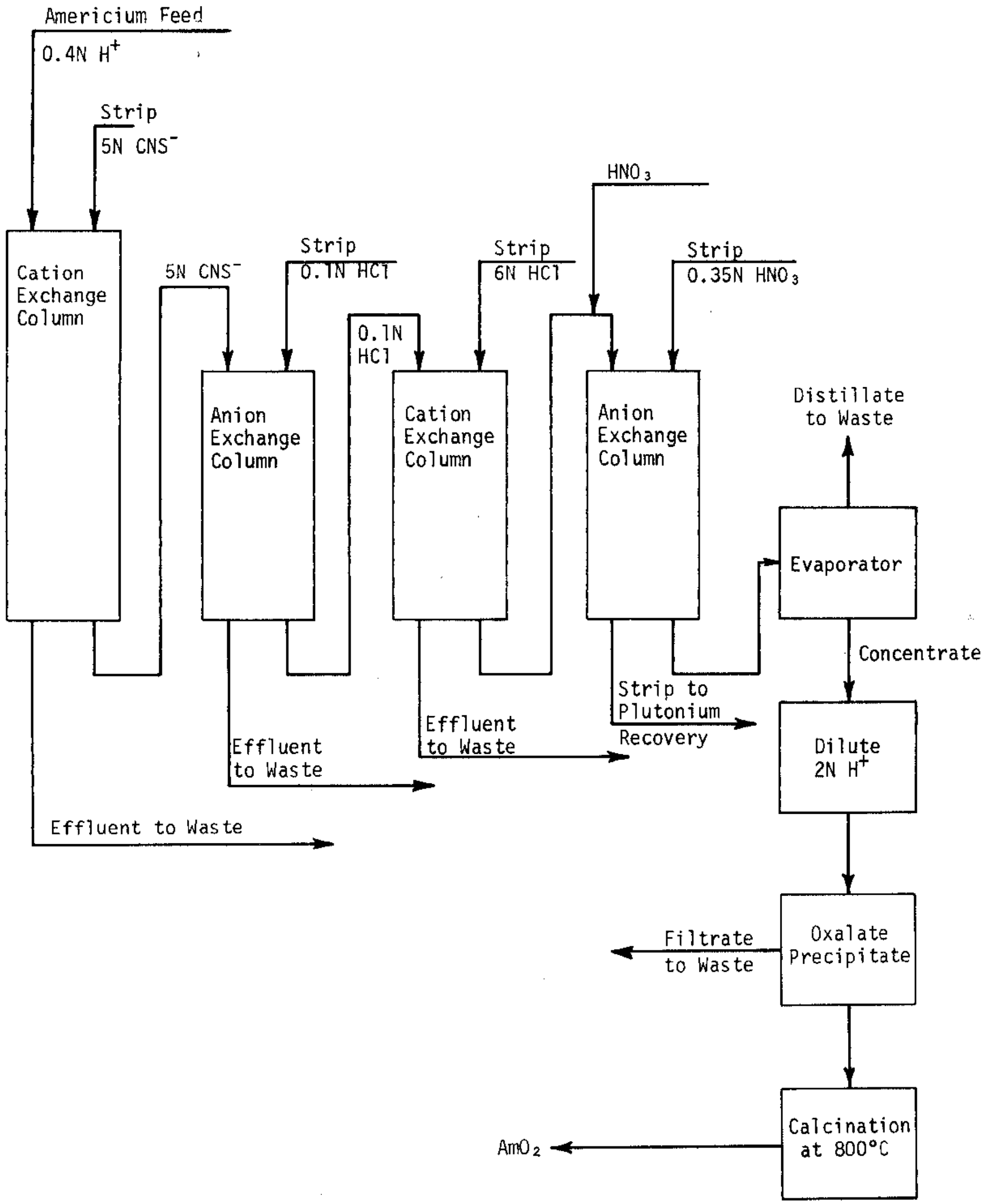
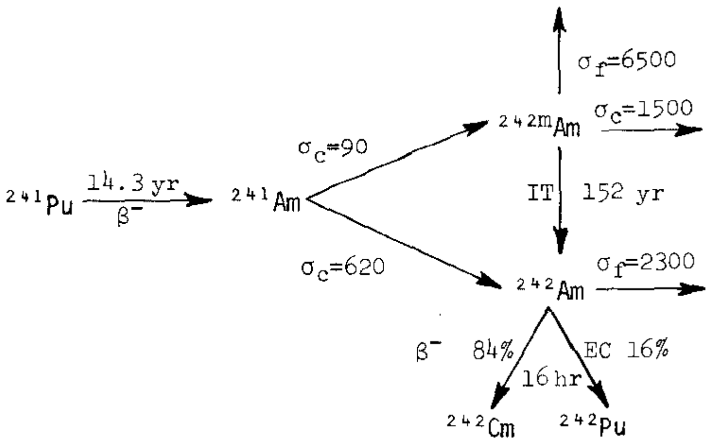


FIGURE V-1. Rocky Flats Americium Recovery Process

C. AMERICIUM-242m (152 yr)

Irradiation of ^{241}Am by thermal neutrons produces both ^{242m}Am and short-lived ^{242}Am. Only about one-eighth of the neutron captures form the desired ^{242m}Am isotope.



Large-scale production of ^{242m}Am is possible by the irradiation of mixtures of ^{241}Am and ^{243}Am from spent power reactor fuel. Figure V-2 shows that irradiation of a 50/50 mixture at a thermal flux of 1.75 x 10^{14} n/(cm^2-sec) yields useful amounts of ^{242m}Am together with larger amounts of ^{242}Cm and ^{244}Cm. The yield of ^{242m}Am is a maximum at an exposure of only 0.5 x 10^{21} n/cm^2; at this point, the americium mixture contains:

	Atoms	Atom, %
^{241}Am	0.777	44.4
^{242m}Am	0.0134	0.77
^{243}Am	0.958	54.8

The ^{242m}Am content can be raised to ∿1.7 atom % by irradiating pure ^{241}Am obtained by "milking" a plutonium stockpile initially containing only ^{241}Pu.

^{242m}Am can be separated from irradiated ^{241}Am only by electromagnetic techniques. No volatile americium compound is known that is suitable for gaseous diffusion or gas centrifuge methods. For small-scale electromagnetic separations, the calutron facilities at ORNL can be used. A single calutron has a feed capacity of about 1 kg/yr for americium containing 1.5% ^{242m}Am. Of the ^{242m}Am

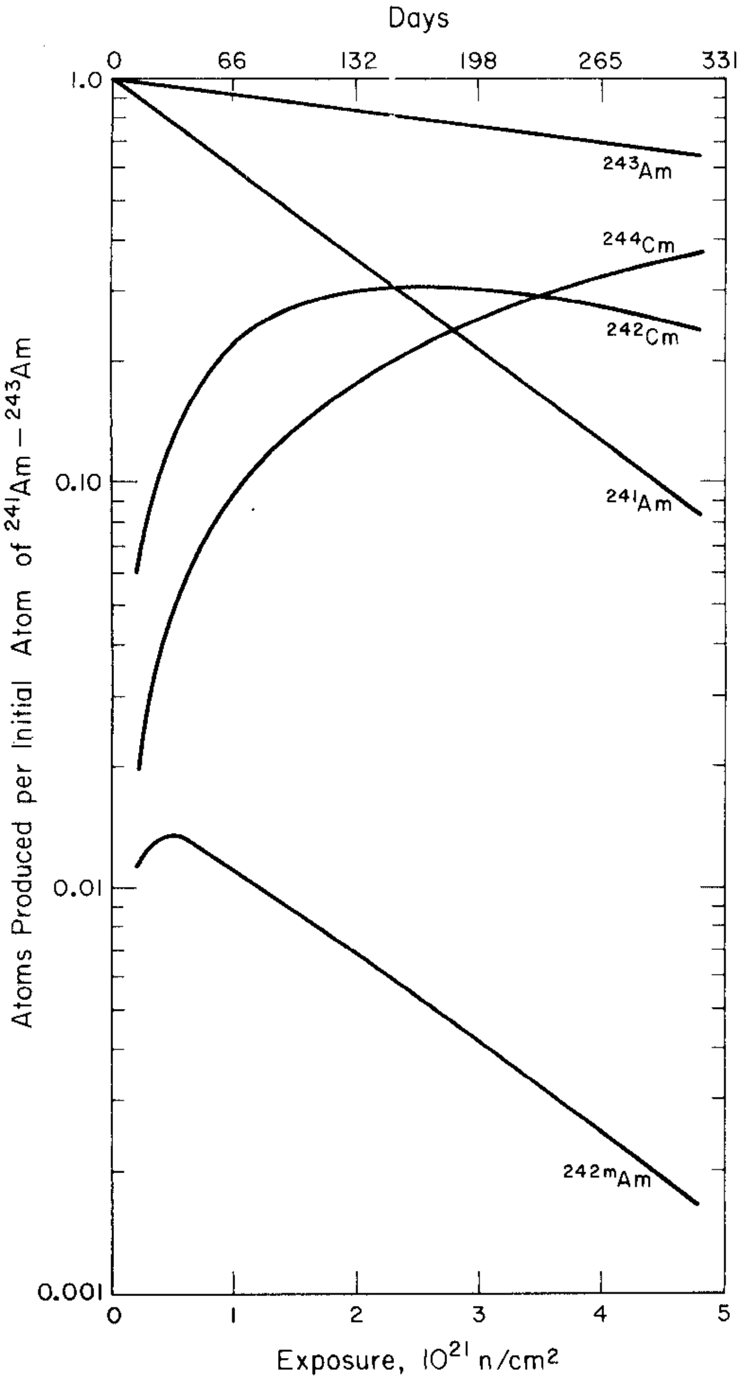


FIGURE V-2. Americium and Curium Produced from a Thermal Neutron Power Reactor initially present in the feed, about 15% is collected at an isotopic purity of 35% in a single pass. Of the remaining feed material, about 95% is recovered at its original isotopic purity. A second pass on the 35% fraction will produce product containing 95% ^{242m}Am. The throughput for the second pass is about 50% higher, or 1.5 kg/yr.

If one assumes a capitalization of $100,000 for added shielding to permit a calutron to handle radioactive materials, an additional $50,000 to modify the chemical facilities to process more americium, a 20-year writeoff, and a $100,000 average operating cost per calutron, the average annual cost is about $107,500. Distributing this cost over the first 2.25 grams of ^{242m}Am separated, the cost per gram of ^{242m}Am separated (35% purity) is about $50,000.

^{242m}Am is one of the few fissionable nuclides containing odd numbers of protons and neutrons and having a long half-life. It has the highest known cross section for thermal neutron fission (6000 barns), a high neutron yield in thermal fissions (3.24 neutrons/fission), and a low thermal neutron capture-to-fission ratio of 0.3.

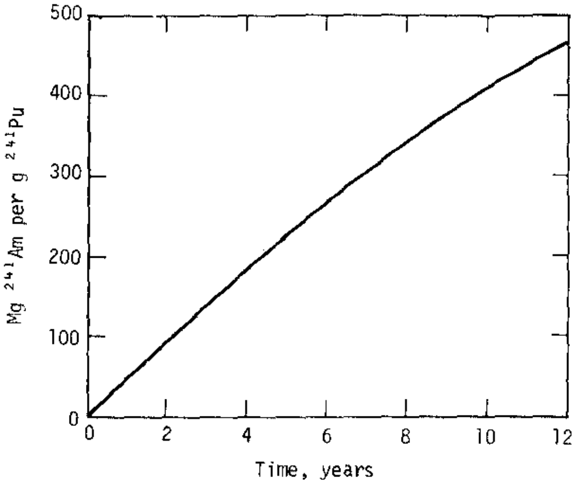


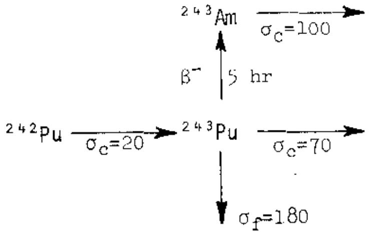
FIGURE V-3. ^{241}Am Produced in Plutonium Contains 1 g of 13-yr ^{241}Pu

D. AMERICIUM-243 (7370 yr)

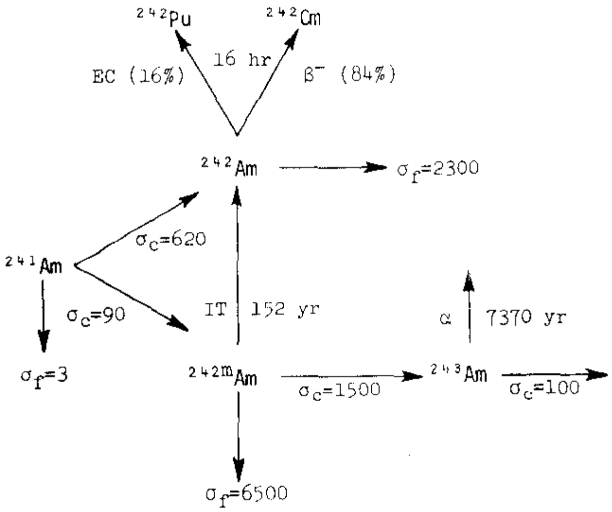
This nuclide is the longest-lived americium isotope, with a half-life about 17 times longer than that of ^{241}Am. When recovery facilities become available, the principal source of ^{243}Am will be spent fuel from power reactors. Projected production of ^{243}Am is shown in Table V-3, assuming that recovery starts in 1977.

The nuclide is formed by irradiation of ^{242}Pu as described in the ^{242}Pu section (Table III-7) and by irradiation of ^{241}Am as shown in the ^{242m}Am section. The plutonium-burning programs at Savannah River will produce nearly isotopically pure ^{243}Am by irradiating ^{242}Pu. Table III-7 shows ^{243}Am production is a maximum after exposure of ^{242}Pu to 1.7 x 10^{22} n/cm^2 at an average thermal flux of 3 x 10^{15} n/(cm^2-sec). About 0.16 atom of ^{243}Am are produced per atom of ^{242}Pu irradiated.

The principal reaction obtained by irradiating ^{242}Pu is:



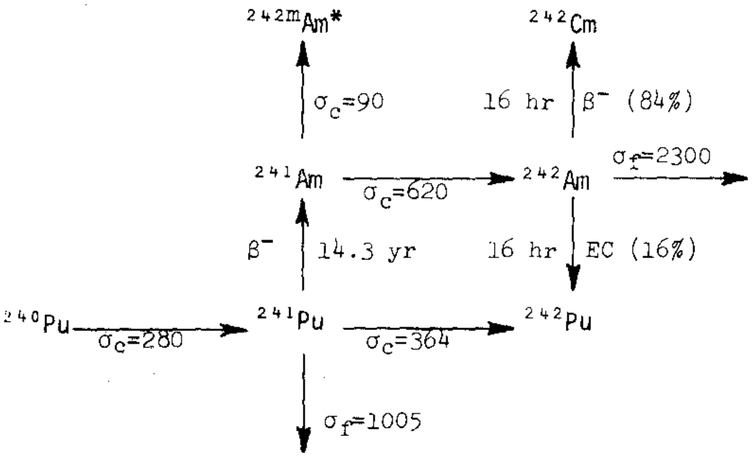
Some ^{243}Am is also formed when ^{241}Am is irradiated with thermal neutrons by the sequence shown below. Although power reactors will also make large amounts of ^{243}Am, it will always be mixed with ^{241}Am, as shown in Table V-3.



^{243}Am is separated as described under ^{241}Am. The relatively low specific alpha activity of ^{243}Am facilitates chemical studies of americium.

E. CURIUM-242 (163 days)

Production of the nuclide is by extended thermal neutron irradiation of ^{241}Am or of ^{240}Pu. Data on irradiation of a 50/50 mixture of ^{241}Am/^{243}Am at an average thermal flux of 1.75×10^{14} n/cm^2 is shown in Table III-5 and illustrated in Figure V-3. For this target, optimum exposure occurs in 172 days, producing about 0.31 atom ^{242}Cm per atom of ^{241}Am-^{243}Am irradiated. The thermal neutron capture sequence is:



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* See previous section on ^{243}Am production.

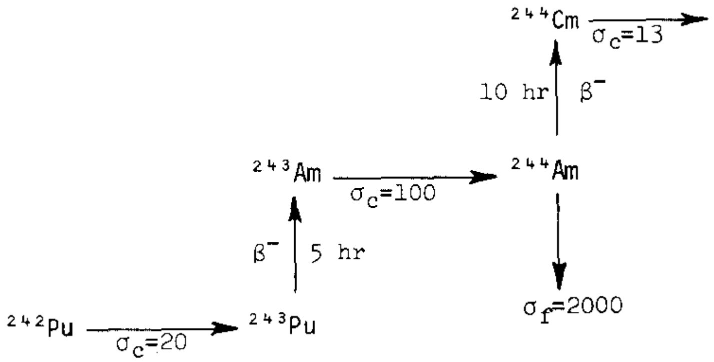
The solvent extraction and ion exchange methods are described under ^{244}Cm. ^{242}Cm is particularly difficult to process because of its intense alpha radiation. New separations methods will be required to separate this material directly rather than allowing it to alpha decay to ^{238}Pu with a 163-day half-life.

^{242}Cm is potentially one of the most important isotopic heat sources because of its high power output (120 w/g), minor shielding requirements (principally Pu L-X-rays), and moderate half-life. It is useful as an extraterrestrial power source, because ^{242}Cm and its daughter ^{238}Pu are man-made, so their existence as contaminants need not interfere with analytical studies to determine lunar (or other) compositions. Chemical analysis using alpha scattering techniques from ^{242}Cm were used in recent lunar explorations, with alpha particles from ^{254}Es serving as an energy standard.

^{242}Cm is compared to other heat sources in Appendix C. The relatively high specific power of ^{242}Cm (120 w/g) is exceeded only by the specific power of ^{254}Cf (11,000 w/g).

F. CURIUM-244 (18.1 yr)

The principal method of production is by thermal reactor neutron irradiation of plutonium, the later stages of which are shown below. The resulting mixture consists primarily of ^{242}Pu, ^{243}Am, and ^{244}Cm, because adjacent isotopes either decay rapidly or have high cross sections.



Production via irradiation of ^{242}Pu is described in Table III-7 for a high-flux reactor operating at 3 x 10^{15} n/(cm^2-sec). Optimum production is achieved in about 5.4 x 10^{22} n/cm (208 days) at a concentration of 0.425 atom ^{244}Cm per atom ^{241}Pu irradiated.

Irradiation of ^{241}Am and ^{243}Am from power reactor fuel residues is described in Table III-5. Optimum production is achieved at an exposure of about 2 x 10^{22} n/cm^2 (1323 days). At this exposure, the concentration of ^{244}Cm is 0.73 atom per atom of ^{241}Am and ^{243}Am irradiated. ^{244}Cm is expected to be available in tonnage quantities in spent power reactor fuels by 1990 or earlier.

Curium has been separated by the ORNL Tramex process, which is diagrammed in Figures IV-3 and IV-4 and discussed in Section IV(C).

The process has been modified at SRL with the introduction of pressurized ion exchange, described in Section IV(D). CmO_2 produced by this process is reduced to metal by heating with magnesium and zinc at 900°C for 2 hours. The MgF_2 and $MgCl_2$ are removed by dissolution in water, and the residual magnesium and zinc are vacuum distilled at 750°C.

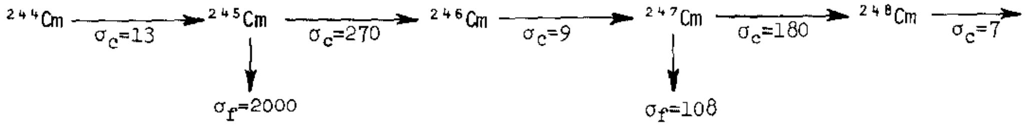
^{244}Cm is important as a long-lived compact power source because of its moderate heat output (2.7 w/g), minor shielding requirements (principally Cm L-X-rays), and long half-life of 18 years.

G. CURIUM-248 (3.52 x 10^5 yr)

^{248}Cm is made by irradiating ^{238}U and higher isotopes by thermal neutrons in a reactor. Production of ^{248}Cm is shown in Table III-7 and in Figure III-2, for irradiation of ^{242}Pu in a high flux of 3 x 10^{15} n/(cm^2-sec). Production at an exposure of 2 x 10^{23} (772 days) is 0.011 atom per initial atom of ^{242}Pu.

Irradiation of a 50/50 mixture of ^{241}Am and ^{243}Am at a flux of 1.75 x 10^{14} n/(cm^2-sec) is shown in Table III-5. At an exposure of 5 x 10^{21} n/cm^2 (331 days) the atom concentration is 0.18 x 10^{-5}. Production of this isotope in relatively low flux power reactors is not attractive.

The neutron capture route from ^{244}Cm is inefficient because of the high fission cross section of ^{245}Cm and the low capture cross sections of ^{244}Cm and ^{246}Cm.



The nuclide is also available as the daughter product of ^{252}Cf alpha decay. The californium fraction consists of the isotopes ^{249}Cf through ^{252}Cf; however, most of the alpha activity is due to ^{252}Cf and ^{250}Cf, so the daughter products are principally ^{248}Cm and ^{246}Cm.

Chemical separation of curium isotopes is described under the section on ^{244}Cm. ^{248}Cm presents no special processing problems because its specific alpha activity is low.

^{248}Cm is useful for chemical studies of curium because its long half-life minimizes analytical problems caused by alpha irradiation.

H. BERKELIUM-247 (1400 yr)

The nuclide is prepared through cyclotron-induced reactions using alpha particles with a target of mixed curium isotopes, since it cannot be prepared through the usual neutron addition to ^{238}U. In the neutron addition sequence, ^{247}Cm is beta stable, and the addition proceeds to form ^{248}Cm. Furthermore, ^{251}Es is also not on the neutron addition route, and its alpha decay is only 0.53%; finally, the electron capture route through ^{247}Cf is also not on the neutron irradiation path. Typical cyclotron reactions are:

$$^{244}\mathrm{Cm}(\alpha,n)^{247}\mathrm{Cf} \xrightarrow[2.4\ \mathrm{hr}]{\mathrm{EC}} {}^{247}\mathrm{Bk}$$

$$^{244}\mathrm{Cm}(\alpha,p)^{247}\mathrm{Bk}$$

$$^{245}\mathrm{Cm}(\alpha,2n)^{257}\mathrm{Cf} \xrightarrow[2.4\ \mathrm{hr}]{\mathrm{EC}} {}^{247}\mathrm{Bk}$$

$$^{245}\mathrm{Cm}(\alpha;p,n)^{247}\mathrm{Bk}$$

$$^{246}\mathrm{Cm}(\alpha,3n)^{247}\mathrm{Cf} \xrightarrow[2.4\ \mathrm{hr}]{\mathrm{EC}} {}^{247}\mathrm{Bk}$$

$$^{246}\mathrm{Cm}(\alpha;p,2n)^{247}\mathrm{Bk}$$

The target curium isotopes are made by successive neutron irradiation to plutonium and would be available from the Savannah River ^{252}Cf production programs.

The recovery and purification of berkelium isotopes is described under ^{249}Bk.

^{247}Bk is useful for chemical and physical studies of the element because it is the longest lived of the berkelium isotopes.

I. BERKELIUM-249 (311 days)

^{249}Bk is produced by neutron irradiation of plutonium, americium, or curium in high flux thermal reactors. Because curium isotopes 242 through 248 are beta stable, a beta decay path exists only at ^{249}Cm. This route from ^{248}Cm is shown below.

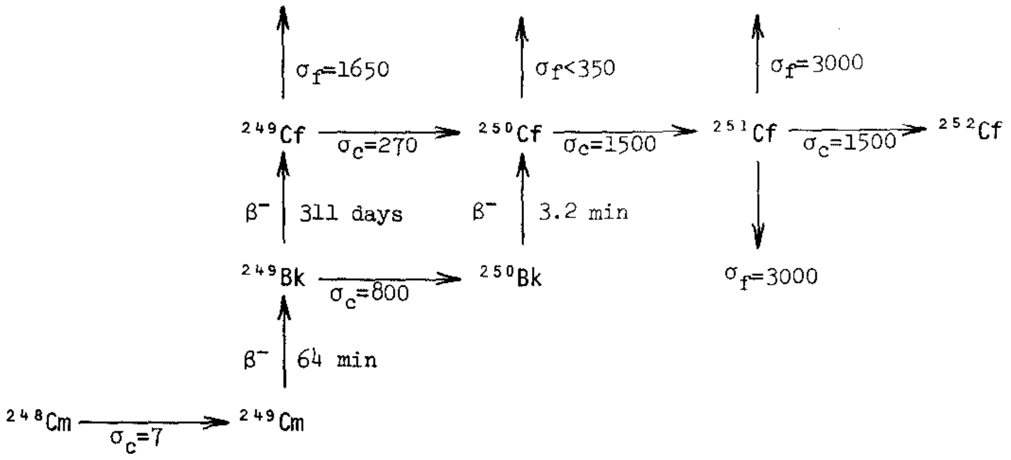
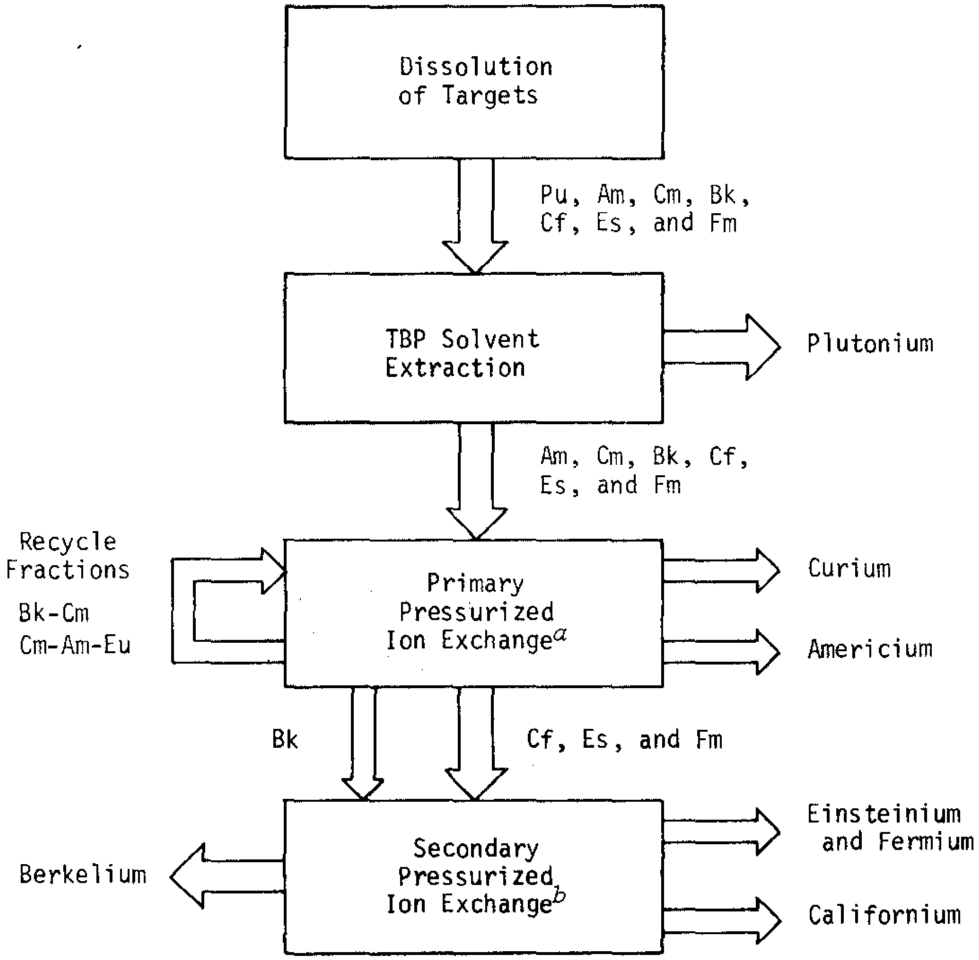


Table III-7 shows that as ^{242}Pu is irradiated at a flux of 3×10^{15} n/(cm^2-sec), the berkelium content increases steadily. At an exposure of 2×10^{23} n/cm^2 (772 days), the ^{249}Bk concentration is 8.2×10^{-5} atoms per atom of ^{242}Pu irradiated. Withdrawal of ^{249}Bk from the Savannah River ^{252}Cf program (at some loss in ^{252}Cf production) is likely to be the best source of berkelium for the next few years.

Berkelium, einsteinium, and fermium can be separated in the ORNL Transuranium Processing Plant (TRU) and in the Multipurpose Processing Facility (MPPF) under construction at SRP. SRP's process is outlined in Figure V-4 and is described in Section IV(D); it is a part of the process designed primarily for the recovery of ^{252}Cf.

Because ^{249}Bk can be made in much larger quantities than ^{247}Bk, it is useful for chemical and metallurgical studies of berkelium. Its 311-day half-life, although much shorter than that of ^{247}Bk (1400 yr), is satisfactory for some studies.



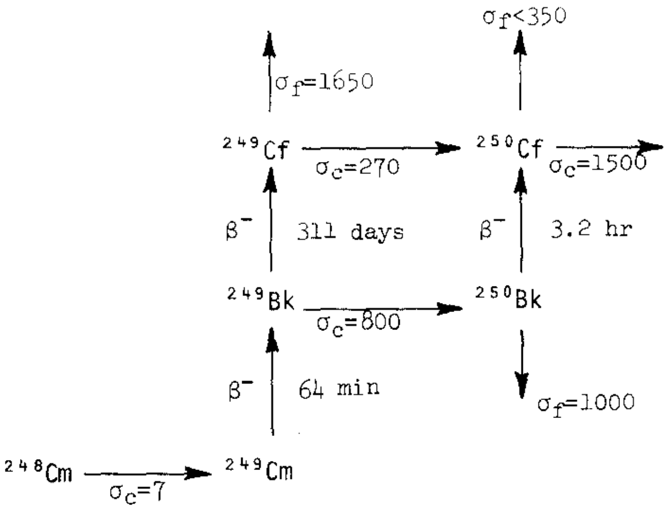
[a] Displacement development chromatography with diethylenetriaminepentaacetic acid

[b] Elution development chromatography with ammonium α-hydroxyisobutyrate

FIGURE V-4. Outline of Savannah River Plant Process for Recovering Transplutonium Nuclides

J. CALIFORNIUM-249 (352 yr)

The nuclide is produced by extended neutron irradiation of plutonium and higher actinides, as shown in Table III-7. The principal route from ^{248}Cm is:



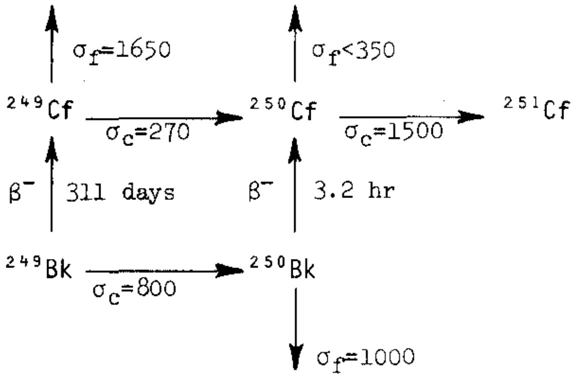
Irradiation of ^{242}Pu to an exposure of 2 x 10^{23} n/cm^2 (772 days) will produce 4.6 x 10^{-7} atom per atom of ^{242}Pu irradiated. ^{249}Cf made by neutron irradiation of ^{248}Cm will always contain some ^{250}Cf. To obtain isotopically pure ^{249}Cf, a chemical separation of curium, berkelium, and californium made after almost all of the ^{250}Bk has decayed to ^{250}Cf will isolate the ^{249}Bk fraction.

^{249}Cf is obtained by making a second chemical separation after the ^{249}Bk has beta-decayed with a 314-day half-life.

Californium isotopes are separated from americium, curium, berkelium, etc., as outlined in the ^{249}Bk section. Because of its long half-life and the absence of spontaneous fission neutrons, ^{249}Cf is useful for the study of chemical and physical properties of californium.

K. CALIFORNIUM-250 (13.1 yr)

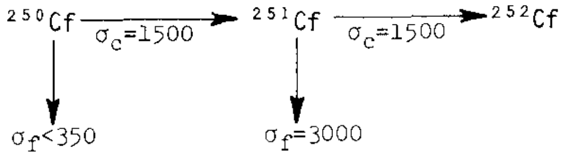
^{250}Cf is prepared by extended neutron irradiation of plutonium and higher actinides. Table III-7 shows that exposure of ^{242}Pu to 2 x 10^{23} n/cm^2 at a flux of 3 x 10^{15} n/(cm^2-sec) produces 7.3 x 10^{-5} atoms per atom of ^{242}Pu exposed. Isotopically pure ^{250}Cf can be chemically separated from the parent ^{254}Es.



The isotope emits 1.2 x 10^{10} n/(sec-gram), and delivers power at 4 w/g. The nuclide could satisfy a need for a long-lived source with a neutron activity comparable to ^{210}Po-Be (138 days at 1.1 x 10^{10} n/(sec-g) or to ^{124}Sb-Be (60 days at 2.8 x 10^{10} n/(sec-g).

L. CALIFORNIUM-251 (900 yr)

The nuclide is prepared from continued neutron irradiation of ^{250}Cf as shown in Table III-7 and Figure III-3.



Irradiation of ^{242}Pu at a flux of 3 x 10^{15} n/(cm^2-sec) to an exposure of 2 x 10^{23} n/cm^2 (772 days) produces 1.8 x 10^{-5} atoms of ^{251}Cf per atom of ^{242}Pu irradiated.

^{251}Cf has the longest half-life of the californium isotopes (900 yr), so it is the primary candidate for chemical and physical characterizations of the elements. Calutron isotopic separations are required to obtain it in pure form.

M. CALIFORNIUM-252 (2.65 yr)

This isotope is one of the most useful actinide nuclides because it emits large numbers of neutrons by spontaneous fission [2.3 x 10^{12} n/(sec-g)], and has a reasonable half-life (2.65 yr).

^{252}Cf is prepared by prolonged irradiation of plutonium, followed by recovery and refabrication of the americium and curium intermediates into targets for high-flux irradiation. Overall, 8.6 x 10^{-4} atoms of ^{252}Cf are produced per atom of ^{242}Pu irradiated to an exposure of 2 x 10^{23} n/(cm^2-sec). The high-flux ^{252}Cf programs at Savannah River and at Oak Ridge are directed toward an annual production of multigram quantities of ^{252}Cf.

Composition of the californium fraction after an exposure of ^{242}Pu to 2 x 10^{23} n/cm^2 is shown in Table V-5. Production of californium and other isotopes is shown in Table III-7 and Figure III-3.

As discussed earlier, ^{252}Cf can also be produced in accelerators and in underground nuclear explosions, but these methods are not currently used to make californium.

^{252}Cf is finding increasing use in medicine; in industry; in mineral, hydrology, petroleum, and space programs; in civil engineering; in agriculture; and in research. Expanded evaluation is continuing under the AEC loan program.

TABLE V-5

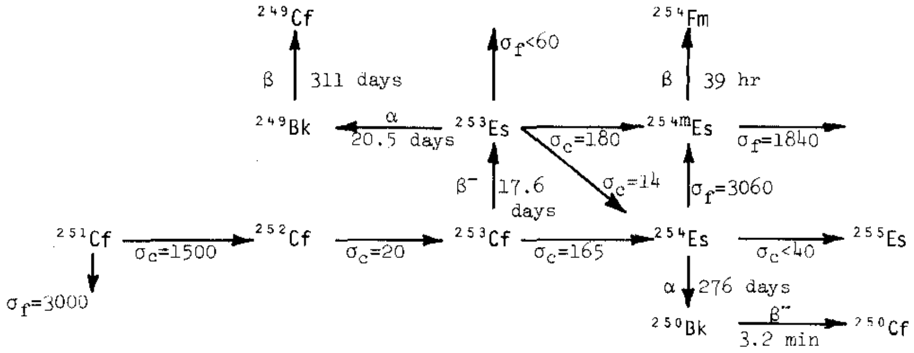
Distribution of Californium Isotopes
After Irradiating ^{242}Pu to an Exposure of 2 x 10^{23} n/cm^2

	Atom %
^{249}Cf	0.05
^{250}Cf	7.83
^{251}Cf	1.88
^{252}Cf	89.32
^{253}Cf	0.84
^{254}Cf	0.08

N. EINSTEINIUM-254 (276 days)

Continued neutron irradiation of californium yields ^{254}Es as shown in Table III-7 and Figure III-3.

Chemically pure samples of einsteinium are obtained by permitting a californium fraction rich in ^{253}Cf to decay by the 18-day beta-decay route, and performing a chemical separation. Neutron irradiation of ^{253}Es produces two isomers - ^{254m}Es, which decays by beta emission with a 39-hour half-life to ^{254}Fm, and

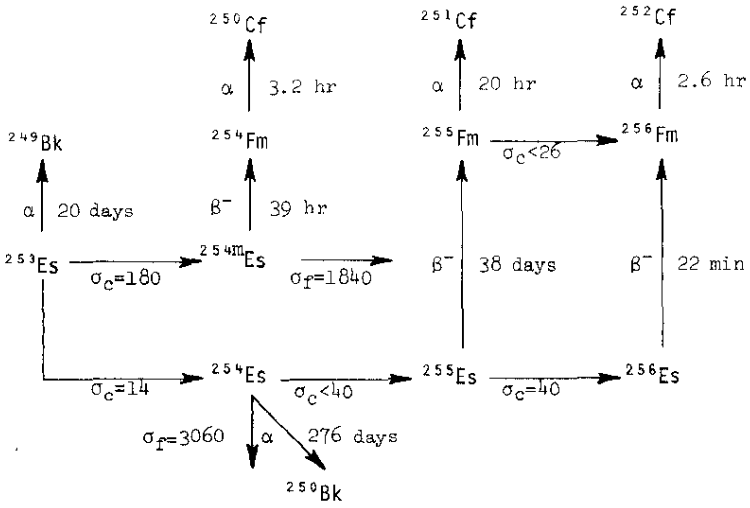


^{254}Es, which decays by alpha emission with a 276-day half-life to ^{250}Bk. Thus, the irradiation of ^{253}Es produces products that are chemically separable.

Because of the absence of ^{254}Es in terrestrial areas, the isotope finds use as a reference energy marker for activation analysis. It was used for this purpose in Surveyor V's chemical analysis of the lunar surface. Because it is the longest-lived isotope of einsteinium, ^{254}Es is the prime candidate for chemical studies of this element.

O. FERMIUM-257 (80 days)

^{257}Fm appears to be the heaviest nuclide that can be prepared by slow neutron irradiation of heavy actinides in a reactor. The capture sequence illustrated below starts with irradiation of ^{253}Es. Irradiation of ^{242}Pu produces about 2×10^{-13} atoms ^{257}Fm per atom ^{242}Pu initially irradiated. Production data are shown in Table III-7.



Processing of berkelium, californium, einsteinium, and fermium have been described under ^{249}Bk. In the process used at SRL, four successive elutions with ammonium alphahydroxyisobutyrate are required to separate einsteinium from fermium held on columns of "Dowex"-50. Other sites have used elution with dilute nitric and hydrochloric acid from actinide mixtures held by di-(2-ethylhexyl)-orthophosphoric acid adsorbed on diatomaceous earth.

^{257}Fm is the longest-lived isotope of fermium and, therefore, is the principal candidate for chemical studies of the element.

P. MENDELEVIUM-258 (54 days)

Synthesis of nuclides by neutron irradiation above atomic number 100 appears unlikely because of the beta-stability of fermium, at least as far as ^{258}Fm. In addition, the half life of ^{258}Fm appears to be of the order of 400 μs, effectively mocking the neutron addition route. Mendelevium isotopes, for example, have been made by irradiation of uranium and transplutonium isotopes with alpha particles and heavier ions. Typical reactions are:

$${}^{253}_{99}\mathrm{Es} + {}^{4}_{2}\mathrm{He} \rightarrow {}^{255}_{101}\mathrm{Md} + 2n \quad (28\ \mathrm{min})$$

$${}^{253}_{99}\mathrm{Es} + {}^{4}_{2}\mathrm{He} \rightarrow {}^{256}_{101}\mathrm{Md} + n \quad (78\ \mathrm{min})$$

$${}^{238}_{92}\mathrm{U} + {}^{22}_{10}\mathrm{Ne} \rightarrow {}^{256}_{101}\mathrm{Md} + 3n + p$$

$${}^{252}_{98}\mathrm{Cf} + {}^{13}_{6}\mathrm{C} \rightarrow {}^{257}_{101}\mathrm{Md} + 5n + 3p \quad (\sim 5\ \mathrm{hr})$$

Mendelevium may be separated by ion exchange chromatography similar to that described under ^{249}Bk. Typically, it would be adsorbed on a "Dowex"-50 x 12 cation resin column from a 0.05 M HCl solution and eluted with ammonium alphahydroxyisobutyrate or with ammonium lactate. Mendelevium can be separated from fermium by sequential elution.

^{258}Md can be used for experimental tracer chemistry.

Q. NOBELIUM

Production of nobelium, like mendelevium, requires accelerator bombardment of transplutonium targets. Typical reactions using transplutonium targets are:

Isotope	Reaction Used	Half-Life, sec
^{251}No	^{244}Cm + ^{12}C → ^{251}No + 5n	0.8 ±0.3
^{252}No	^{244}Cm + ^{12}C → ^{252}No + 4n	2.3 ±0.3
	^{244}Cm + ^{13}C → ^{252}No + 5n	∿2.5
	^{239}Pu + ^{18}O → ^{252}No + 5n	
^{253}No	^{244}Cm + ^{13}C → ^{253}No + 4n	105 ±20
	^{246}Cm + ^{12}C → ^{253}No + 5n	∿100
	^{242}Pu + ^{16}O → ^{253}No + 5n	95 ±10
	^{239}Pu + ^{18}O → ^{253}No + 4n	
^{254}No	^{246}Cm + ^{12}C → ^{254}No + 4n	55 ±5
	^{246}Cm + ^{13}C → ^{254}No + 5n	∿50
	^{244}Cm + ^{13}C → ^{254}No + 3n	∿50
	^{243}Am + ^{15}N → ^{254}No + 4n	
	^{238}U + ^{22}Ne → ^{254}No + 6n	50 ±10
	^{242}Pu + ^{16}O → ^{254}No + 4n	75 ±15
^{255}No	^{246}Cm + ^{13}C → ^{255}No + 4n	185 ±20
	^{248}Cm + ^{12}C → ^{255}No + 5n	∿180
	^{238}U + ^{22}Ne → ^{255}No + 5n	∿120
	^{242}Pu + ^{18}O → ^{255}No + 5n	180 ±10
^{256}No	^{248}Cm + ^{12}C → ^{256}No + 4n	2.7 ±0.5
	^{248}Cm + ^{13}C → ^{256}No + 5n	3.2 ±0.2
	^{246}Cm + ^{13}C → ^{256}No + 3n	∿3
	^{238}U + ^{22}Ne → ^{256}No + 4n	6 ±2
	^{242}Pu + ^{18}O → ^{256}No + 4n	9 ±3
^{257}No	^{248}Cm + ^{13}C → ^{257}No + 4n	23 ±2
	^{248}Cm + ^{12}C → ^{257}No + 3n	∿20

Appendix D gives the energy levels and decay schemes for some of the principal isotopes from ^{241}Am to ^{255}No.

R. LAWRENCIUM

Lawrencium, the last member of the actinide series, has so far found use only for experimental tracer chemistry. Typical accelerator reactions that produce lawrencium are:

$$^{243}\mathrm{Am} + {}^{18}_{8}\mathrm{O} \rightarrow {}^{256}\mathrm{Lw} + 5n$$

$$^{248}\mathrm{Cm} + {}^{15}\mathrm{N} \rightarrow {}^{259}\mathrm{Lw} + 4n$$

$$^{249}\mathrm{Cf} + {}^{12}\mathrm{C} \rightarrow {}^{258}\mathrm{Lw} + p,2n$$

$$^{249}\mathrm{Cf} + {}^{13}\mathrm{C} \rightarrow {}^{258}\mathrm{Lw} + p,3n$$

$$\left.\begin{matrix} ^{250}\mathrm{Cf} \\ ^{251}\mathrm{Cf} \\ ^{252}\mathrm{Cf} \end{matrix}\right\} + {}^{11}_{5}\mathrm{B} \rightarrow {}^{257}\mathrm{Lw} + \left\{\begin{matrix} 4n \\ 5n \\ 6n \end{matrix}\right.$$

$$\left.\begin{matrix} ^{250}\mathrm{Cf} \\ ^{251}\mathrm{Cf} \\ ^{252}\mathrm{Cf} \end{matrix}\right\} + {}^{10}_{5}\mathrm{B} \rightarrow {}^{257}\mathrm{Lw} + \left\{\begin{matrix} 3n \\ 4n \\ 5n \end{matrix}\right.$$

S. RUTHERFORDIUM

Rutherfordium, the first element of the "transactinide" series, behaves chemically like hafnium and zirconium, falling into the group IVB elements in the periodic table. Typical accelerator reactions that yield rutherfordium isotopes are:

$$^{242}\mathrm{Pu} + {}^{22}\mathrm{Ne} \rightarrow {}^{260}\mathrm{Rf} + 4n$$

$$^{246}\mathrm{Cm} + {}^{18}\mathrm{O} \rightarrow {}^{260}\mathrm{Rf} + 4n$$

$$^{248}\mathrm{Cm} + {}^{18}\mathrm{O} \rightarrow {}^{261}\mathrm{Rf} + 5n$$

$$^{249}\mathrm{Cf} + {}^{12}\mathrm{C} \rightarrow {}^{260}\mathrm{Rf} + n$$

$$^{250}\mathrm{Cf} + {}^{13}\mathrm{C} \rightarrow {}^{260}\mathrm{Rf} + 3n$$

T. HAHNIUM

Hahnium can be synthesized by bombarding americium, berkelium, and californium with heavy ions:

$$^{243}\mathrm{Am} + {}^{22}\mathrm{Ne} \rightarrow {}^{261}\mathrm{Ha} + 4n$$

$$^{252}\mathrm{Cf} + {}^{15}\mathrm{N} \rightarrow {}^{261}\mathrm{Ha} + 6n$$

$$^{250}\mathrm{Cf} + {}^{15}\mathrm{N} \rightarrow {}^{261}\mathrm{Ha} + 4n$$

$$^{249}\mathrm{Bk} + {}^{16}\mathrm{O} \rightarrow {}^{261}\mathrm{Ha} + 4n$$

$$^{249}\mathrm{Bk} + {}^{18}\mathrm{O} \rightarrow {}^{262}\mathrm{Ha} + 5n$$

U. SUPERHEAVY ELEMENTS

The shell model of the nucleus developed by M. Goeppert-Mayer and others, provides for regions of unusual stability under specific combinations of neutrons and protons. The magic numbers of protons are generally recognized as 2, 8, 20, 28, 50, 82, and 114. The corresponding numbers of neutrons that form unusually stable combinations with these protons are 2, 8, 20, 28, 50, 126, and 184. Efforts to synthesize an element of 114 protons and 184 neutrons have been made by bombarding targets of transplutonium isotopes with ions of argon and calcium, for example,

$$ {}^{244}_{94}\mathrm{Pu} + {}^{48}_{20}\mathrm{Ca} \rightarrow {}^{288}_{114}\mathrm{X} + 4n $$

$$ {}^{248}_{96}\mathrm{Cm} + {}^{40}_{18}\mathrm{Ar} \rightarrow {}^{282}_{114}\mathrm{X} + 6n $$

$$ \rightarrow {}^{283}_{114}\mathrm{X} + 5n $$

$$ \rightarrow {}^{284}_{114}\mathrm{X} + 4n $$

To date, the efforts have been unsuccessful. Theoretical estimates for some of these elements show spontaneous fission half-lives as high as 10^{16} years and total half-lives of 10^{8} years.

When superheavy elements with atomic numbers in the range Z = 110 to 114 undergo spontaneous fission, they emit about 10 neutrons with kinetic energies from 0.5 to 7 Mev. This relatively large emission of neutrons with each fission event is an indicator of the presence of a superheavy element. Searches made for heavy element ${}^{229}_{110}\mathrm{X}$ (with a spontaneous fission half-life of 10^{11} years and a total half-life of 10^{8} years) in old (5×10^{7} yr) platinum ores containing chemically similar elements, such as osmium, iridium, and rhodium, were inconclusive. Either the concentration of ${}^{294}_{110}\mathrm{X}$ was less than 1 part in 10^{10} relative to platinum, or the total half-life was less than 10^{8} years.

APPENDIX A

SALES PRICES OF SOME ACTINIDES[a]

Isotope	Price		Effective Date	Available From
^{237}Np	\$225/g; \$0.25/mg for quantities <1 g		4-19-69	ORNL
^{238}Pu	\$700/g, 80-89% enriched		8-26-70	ORNL
	\$1250/g, >90% enriched, with <0.3 ppm ^{236}Pu		8-26-70	ORNL
^{241}Am	\$150/g		8-26-70	ORNL
	\$60/μCi			Amersham/Searle
	\$125/10 mCi			Amersham/Searle
	\$35/1 mCi	Americium oxide in 3N HNO (5 ml)	1970-71	Amersham/Searle
	\$45/2 mCi			Amersham/Searle
	\$75/5 mCi			Amersham/Searle
	\$125/10 mCi			Amersham/Searle
^{243}Am	\$100/mg (supplied as oxide in quantities up to 10 mg, plus \$250 technical service charge)		1-1-69	ORNL
^{242}Cm	\$30/1 mCi	In 3N HNO_3 (5 ml)		Amersham/Searle
	\$35/5 mCi			Amersham/Searle
	\$40/10 mCi			Amersham/Searle
^{244}Cm	\$100/mg (supplied as oxide in quantities up to 10 mg, plus \$250 technical service charge)		1-1-69	ORNL
^{252}Cf	\$10/μg (up to 1 μg is available without AEC approval) (Shipping charge \$1600 for 10 μg - 1 mg)		Early '71	SRP

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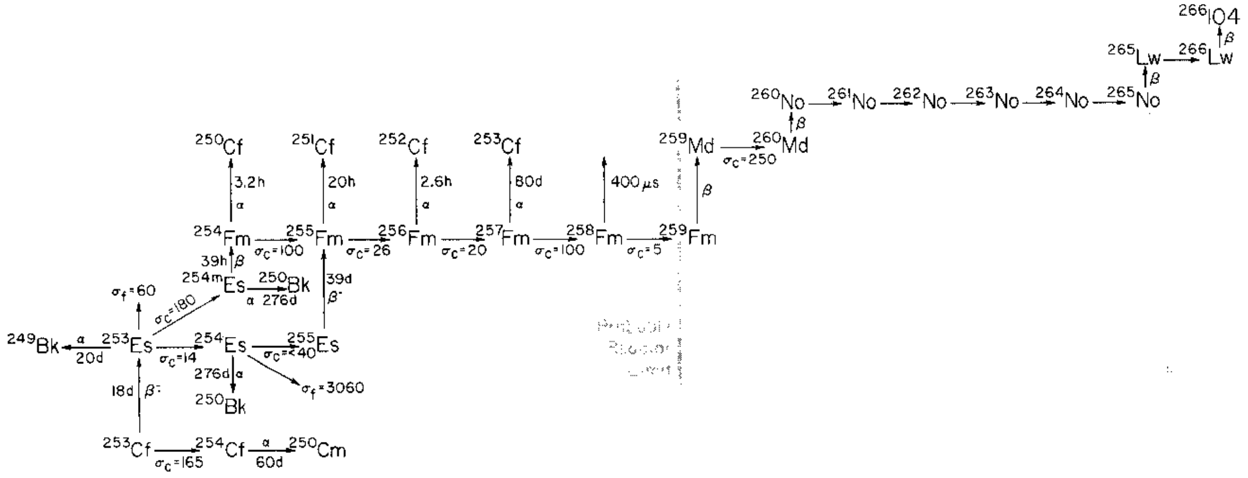
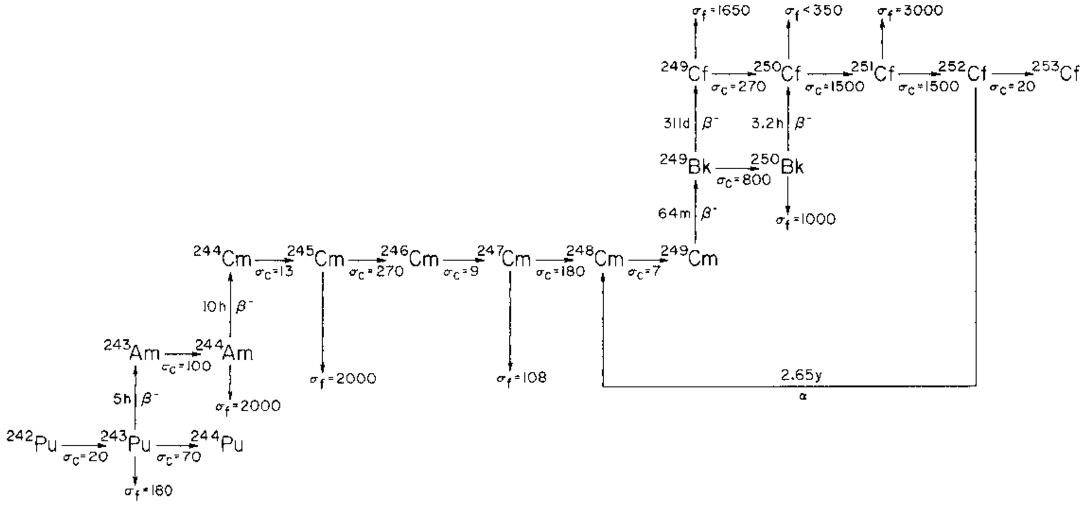
[a] ^{248}Cm, ^{249}Bk, ^{249}Cf, ^{250}Cf, ^{254}Es, and ^{257}Fm are available in "research quantities" on transfer from the ORNL research pool.

APPENDIX B

PRINCIPAL PRODUCTION ROUTES FOR TRANSPLUTONIUM NUCLIDES FOR ^{242}Pu





APPENDIX C

CHARACTERISTICS OF RADIOISOTOPIC HEAT SOURCES

Taken from C. A. Rohrmann, *Radioisotopic Heat Sources*. HW-76323 Supplement, Battelle-Northwest Laboratory, Richland, Wash. (1968).

	^{60}Co	^{90}Sr	^{106}Ru	^{137}Cs	^{144}Ce	^{147}Pm	^{170}Tm	^{210}Po	^{238}Pu	^{242}Cm	^{244}Cm
Specific Power, w/g (100% basis)	17.4	0.96	33.1	0.42	25.6	0.33	13.6	141	0.56	120	2.65
Half-Life, years	5.24	28	1.0	30	0.78	2.6	0.35	0.38	87.4	0.45	18.1
Estimated Isotopic Purity, %	10[a]	55[b]	3.3[b]	35	4.5[b]	95[b]	10[a]	95	80	90	95
Compound Form	Metal	SrTiO$_3$	Metal	CsCl	Ce$_2$O$_3$	Pm$_2$O$_3$[c]	Tm$_2$O$_3$	Metal	PuO$_2$	Cm$_2$O$_3$	Cm$_2$O$_3$
Melting Point of Compound, °C	1480	1910	2310	646	2190	2130	2375	255	2250	1950	1950
Active Isotope in Compound, %	10	23	3.3	28	3.8	82	8.8	95	70	82	86
Specific Power, w/g compound	1.74	0.22	1.1	0.12	1.0	0.27	1.2	134	0.39	98	2.27
Density of Compound, g/cm^3, actual or 90% TD	8.8	4.6	12.4	3.2	6.2	6.6	8.0	9.3	10	9	9
Power Density, w/cm^3 compound	15.8	1.01	13.7	0.38	6.2	1.8[c]	9.6	1210[d]	3.9[d]	882[d]	20.4[d]
Dimension of Capsule for 50 w(t), cm[e]	2.2	4.4	2.3	6.1	2.8	3.9	2.5	2.0	3.8	2.0	2.4
Availability	Avail.	Avail.	Poten. Avail.	Avail.	Avail.	Avail.	Avail.	Avail.	Avail.[f]	Poten. Avail.	Avail.[f]
Type of Radiation (Major)[g]	γβ	β*x*	γβ*x*	βγ*x*	βγ*x*	β	β	α	α	α*n*	α*n*
Shielding Required[h] [MeV of significant β or γ]	Heavy (9.5) [1.33γ]	Heavy (6) [2.26β]	Heavy (9) [3.35β]	Heavy (4.6) [1.17β 0.67γ]	Heavy (10.2) [2.98β 2.18γ]	Minor (1) [0.23β]	Moderate (2.5) [0.97β]	Minor (1) [0.8γ]	Minor (0.1) [0.04γ]	Minor (0.4) [0.04γ]	Moderate (2) [0.04γ]
Biological Hazard, Ci/m^3	3x10^{-9}	10^{-10}	2x10^{-9}	5x10^{-9}	2x10^{-9}	2x10^{-8}	10^{-8}	7x10^{-11}	7x10^{-13}	4x10^{-11}	3x10^{-12}
MPC, Air, w/m^3	4.6x10^{-11}	6.8x10^{-13}	2x10^{-11}	2.4x10^{-11}	1.6x10^{-11}	7.2x10^{-12}	2.3x10^{-11}	2.2x10^{-12}	2.3x10^{-14}	1.4x10^{-12}	10^{-13}
Continuous Exposure, g/m^3	2.7x10^{-12}	7x10^{-13}	5.9x10^{-13}	5.8x10^{-11}	6.3x10^{-13}	2.2x10^{-11}	1.7x10^{-12}	1.6x10^{-14}	4.1x10^{-14}	1.2x10^{-14}	3.7x10^{-14}
Estimated Future Price, \$/g (Pure)[Present price]	280 [452]	20[i] [29]	120[i]	6.5[i] [10.9]	50[i] [477]	75[i] [186]	136	2800[j]	300[j]	2000[k]	170[i]
Estimated Future Price, \$/w	16	21	5	15.5	2	220	10	20	540	17	64
Specific Activity, Ci/g (100% basis)	1130	142	3394	87	3180	928	6048	4500	17	3320	81
Specific Power, Ci/w	65	148	102	207	124	2788	445	32	30	28	29
Total kwh(t)/initial g for 1-year mission	575 (5)	74 (10)	210 (1)	33 (10)	150 (1)	6 (3)	33 (0.4)	350 (0.4)	47 (10)	310 (0.4)	190 (10)
Minimum Cost, \$/kwh(e) for 1-year mission[l]	15 (5)	6 (10)	23 (1)	4 (10)	11 (1)	350 (3)	125 (0.4)	235 (0.4)	138 (10)	180 (0.4)	22 (10)
Spontaneous Fission Half-Life, years	-	-	-	-	-	-	-	-	4.9x10^{10}	7.2x10^{6}	1.4x10^{7}
Estimated Recovery from Power Reactor Fuels, g/tonne at 25,000 MWD/T	-	360	53	835	89	60 (aged)	-	-	(345 Np)	-	9 (200/Pu Recycle)
Power Reactor Production, kg/1000 MWY(e)	-	16	2.3	36	3.8	2.6 (aged)	-	-	(15 Np)	-	0.4 (9/Pu Recycle)
Availability in 1980, kw(t)[m]	MWS	1200	11,000	1200	8400	53	MWS	MWS	250[m]	-	95 150[n]

---

a. Could be produced at higher concentrations, at least double this value, with higher neutron flux and longer exposures.

b. From U.S. power reactor fuels, 1 year after discharge (one half-life for promethium).

c. Metal may be preferred; m.p. = 1080°C [F. Weigel. *Angew. Chem.* 75, 451 (1963)]; expected power density = 2.3 w/cm^3.

d. Does not include the usual 1:1 void volume allowance for helium pressurization with this and all other encapsulated alpha emitters.

e. Capsule is a right cylinder with diameter = height; ^{210}Po and ^{242}Cm assumed to be diluted to 50 w/cm^3. Capsule wall thickness is 0.3 cm.

f. From spent fuels from civilian power reactors (150,000 MW(e) in 1980), except for ^{60}Co, ^{170}Tm, ^{210}Po which could be produced in megawatt quantities via neutron irradiation of targets.

g. α, alpha; β, beta; γ, gamma; *n*, neutron; *x*, penetrating bremsstrahlung.

h. Except for shielding against neutrons. Number in parentheses indicates approximate inches of lead shielding required for 1 kw source for 10 mR/hr at 1 m. (See ORNL-3576 except for promethium.)

i. From large, civilian, power reactor, spent fuel processing and isotope recovery facility, after 1970 (see BNWL-25).

j. Upward revision of minimum costs.

k. Anticipated price with large scale reactor fueling with plutonium (Pu at \$10/g), and ^{241}Am recycle.

l. At terminal or lowest specific power (w/g); at 5% thermal-to-electrical conversion efficiency. For comparison, power from chemical batteries (specifically flashlight batteries) is calculated to cost \$100/kwh(e).

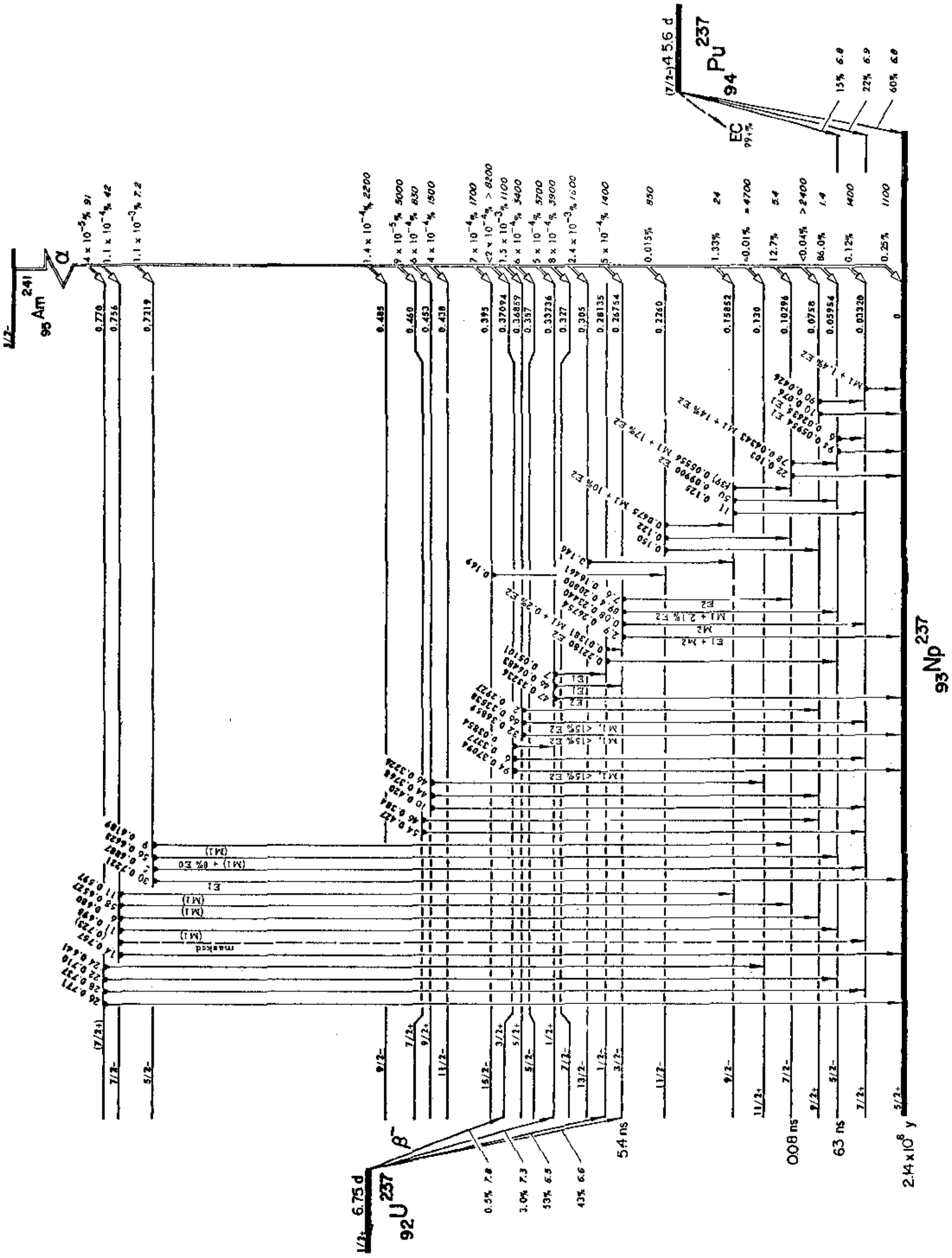
m. From accumulated ^{237}Np from spent fuels from civilian power reactors (550 kg neptunium recoverable in the year 1980).

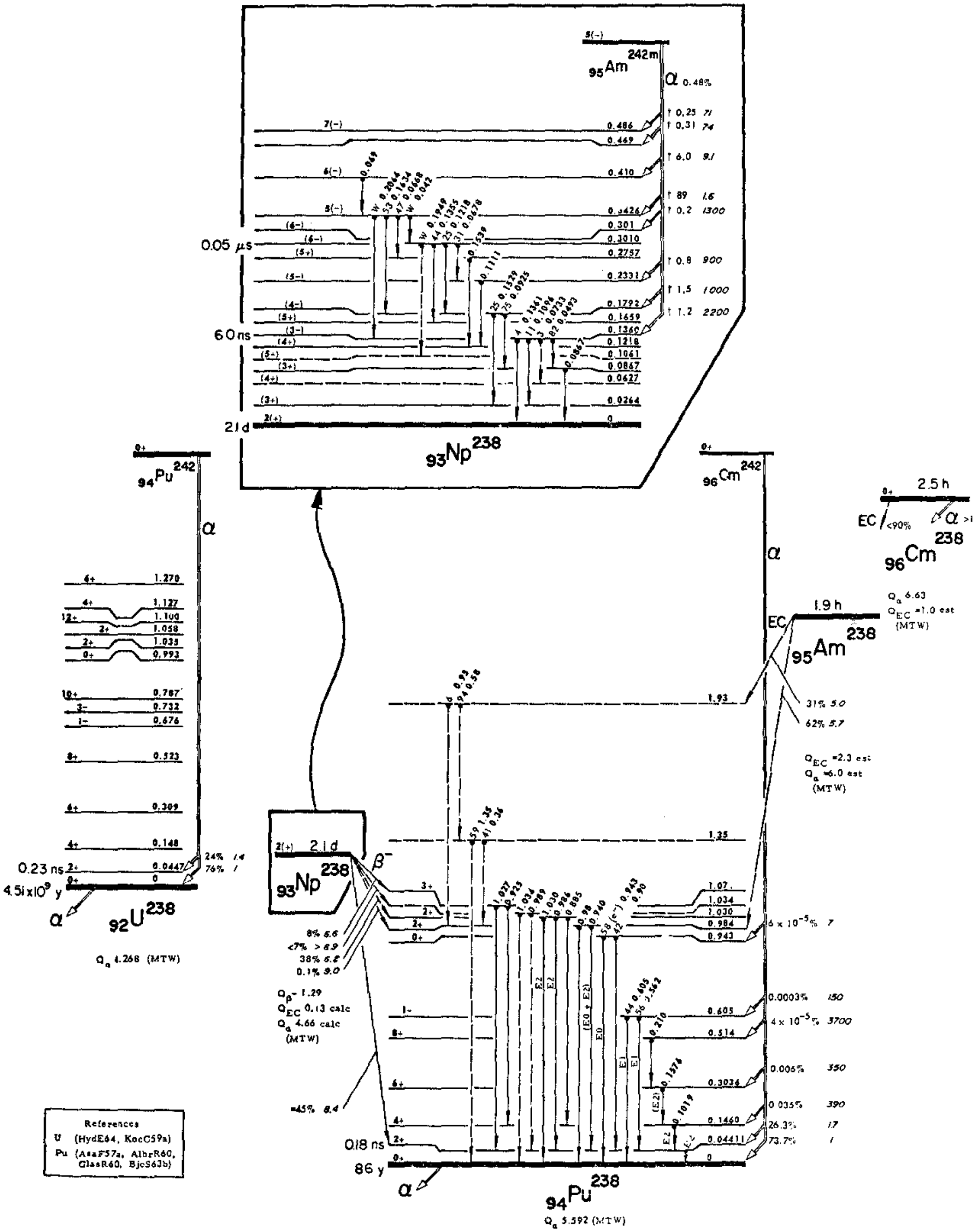
n. With recycle of plutonium and americium.

APPENDIX D

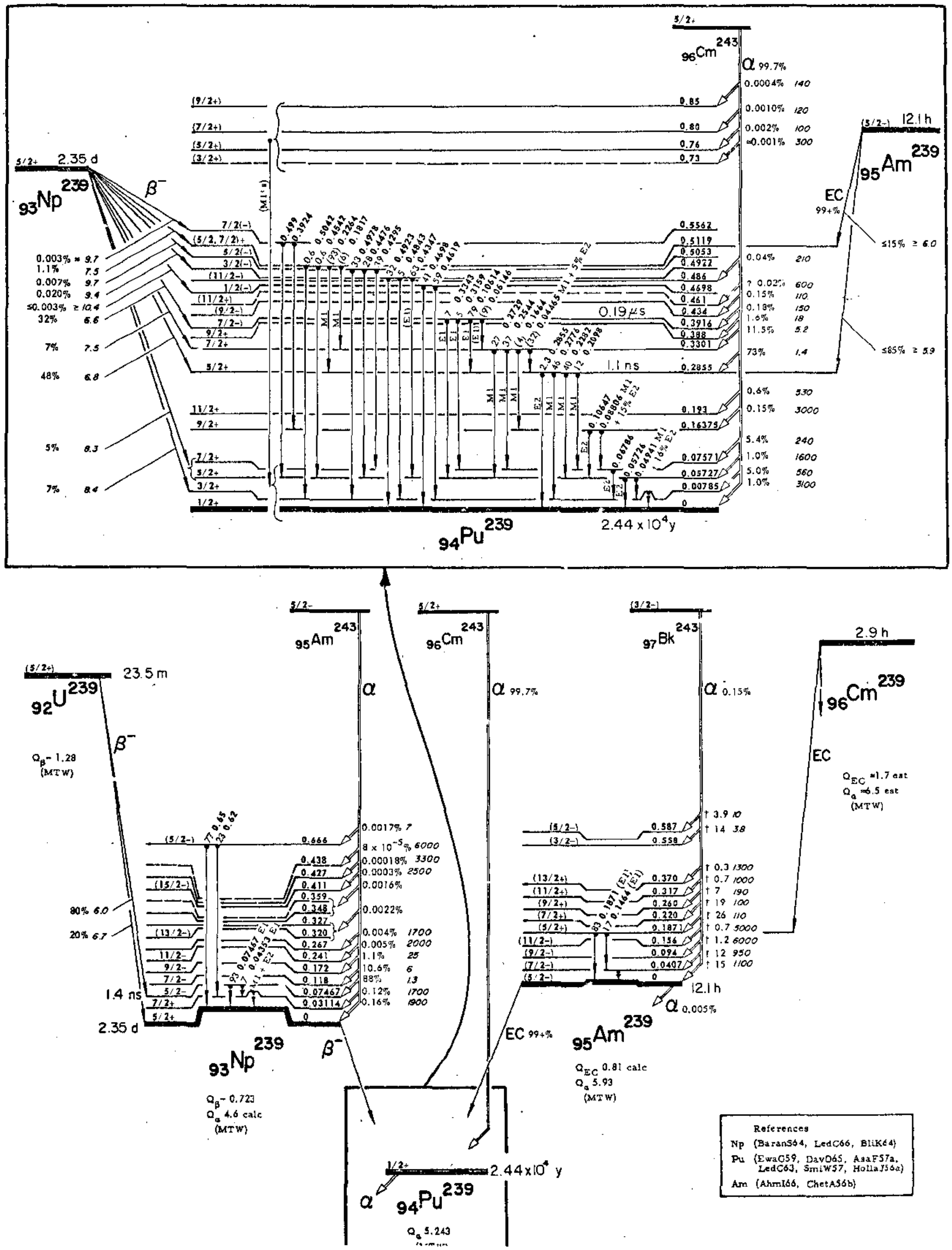
ENERGY LEVELS AND DECAY SCHEMES OF SELECTED ACTINIDES

Reprinted by permission from C. M. Lederer, J. M. Hollander, and I. Perlman. *Table of Isotopes*. 6th ed. John Wiley & Sons, Inc., New York (1968).

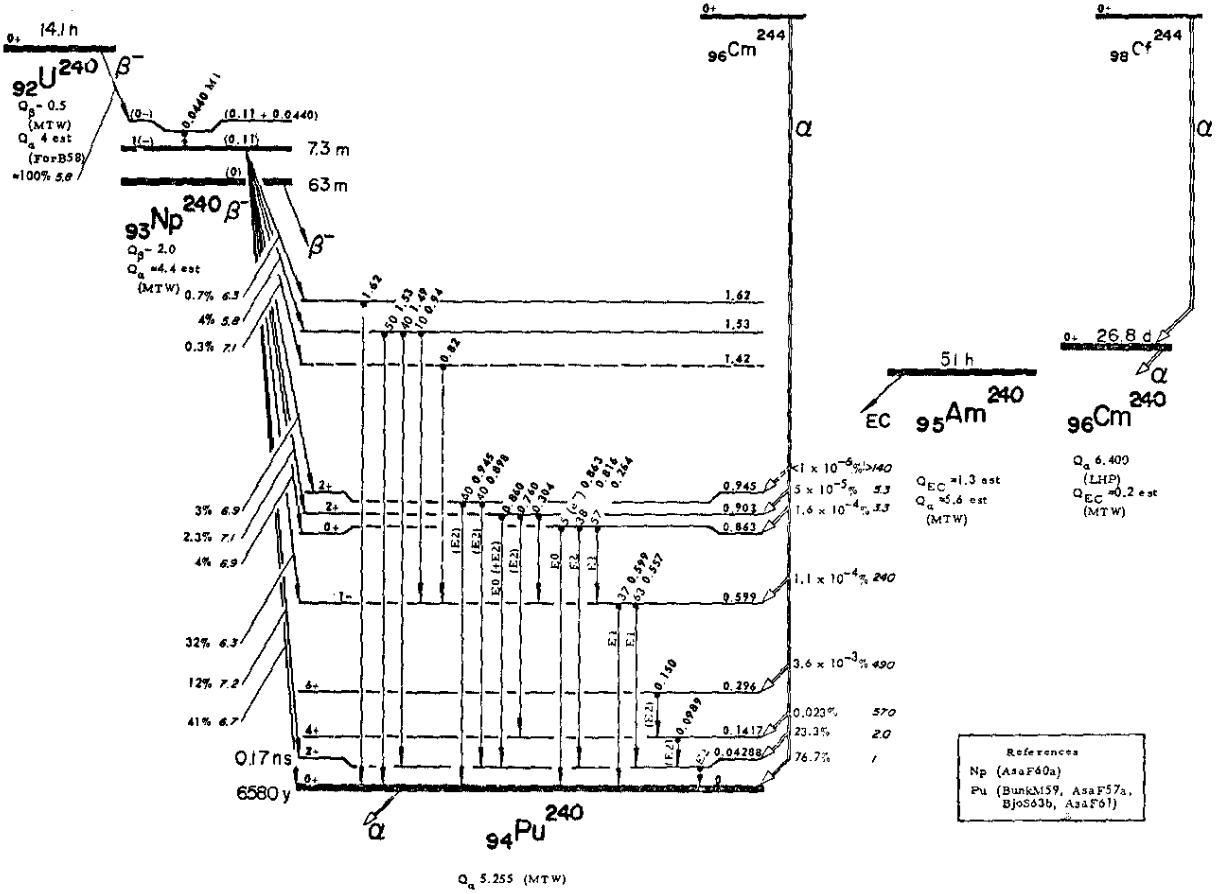




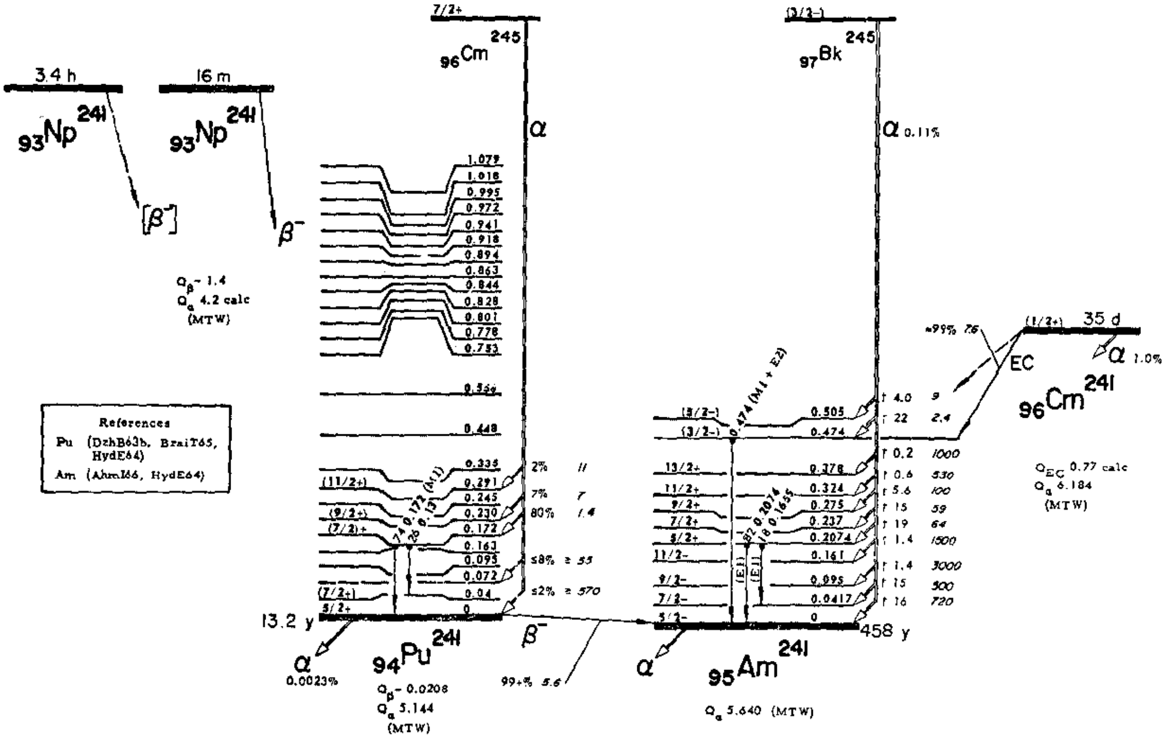
$_{95}Am^{242m}$
5(-)
α 0.48%
$_{93}Np^{238}$
0.05 μs
6.0 ns
2.1 d
7(-) 0.486
0.469
6(-) 0.410
5(-) 0.3426
0.301
0.3010
0.2757
0.2331
0.1792
0.1659
0.1360
0.1218
0.1061
0.0867
0.0627
0.0264
2(+) 0
↑ 0.25 71
↑ 0.31 74
↑ 6.0 9.1
↑ 89 1.6
↑ 0.2 1300
↑ 0.8 900
↑ 1.5 1000
↑ 1.2 2200
$_{94}Pu^{242}$
0+
α
$_{92}U^{238}$
6+ 1.270
4+ 1.127
12+ 1.100
2+ 1.058
2+ 1.033
0+ 0.993
10+ 0.787
3- 0.732
1- 0.676
8+ 0.523
6+ 0.309
4+ 0.148
0.23 ns 2+ 0.0447
0+ 0
4.51×10⁹ y
24% 1.4
76% 1
α
$Q_α$ 4.268 (MTW)
2(+) 2.1 d
$_{93}Np^{238}$
β⁻
8% 6.6
<7% >6.9
38% 6.8
0.1% 9.0
$Q_{β^-}$ 1.29
Q_{EC} 0.13 calc
$Q_α$ 4.66 calc
(MTW)
≈45% 8.4
$_{96}Cm^{242}$
0+
α
$_{96}Cm^{238}$
0+ 2.5 h
EC <90%
α >10%
$Q_α$ 6.63
Q_{EC} ≈1.0 est
(MTW)
1.9 h
EC
$_{95}Am^{238}$
31% 5.0
62% 5.7
Q_{EC} ≈2.3 est
$Q_α$ ≈6.0 est
(MTW)
1.93
1.35
3+ 1.07
1.034
2+ 1.030
2+ 0.984
0+ 0.943
6 × 10⁻⁵% 7
1- 0.605
8+ 0.514
6+ 0.3036
4+ 0.1460
2+ 0.04411
0+ 0
0.0003% 150
4 × 10⁻⁵% 3700
0.006% 350
0.035% 390
26.3% 1.7
73.7% 1
0.18 ns
86 y
α
$_{94}Pu^{238}$
$Q_α$ 5.592 (MTW)
E2
E0
(E0 + E2)
E1
(E2)
References
U (HydE64, KocC59a)
Pu (AsaF57a, AlbrR60, GlasR60, BjoS63b)



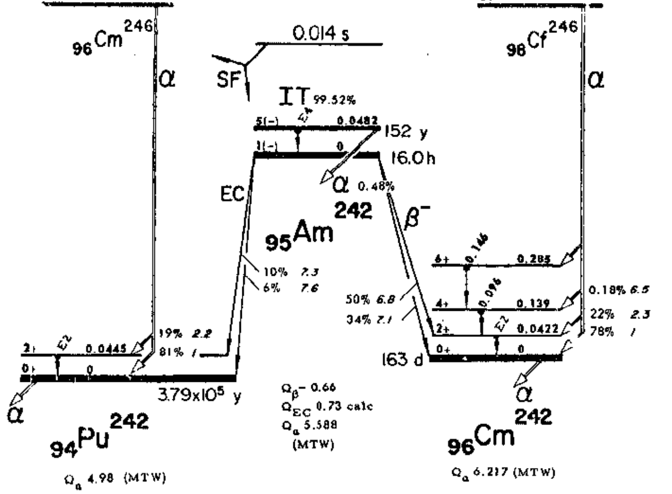
References
Np (BaranS64, LedC66, BliK64)
Pu (EwaG59, DavD65, AsaF57a, LedC63, SmiW57, HollaJ56a)
Am (AhmI66, ChetA56b)



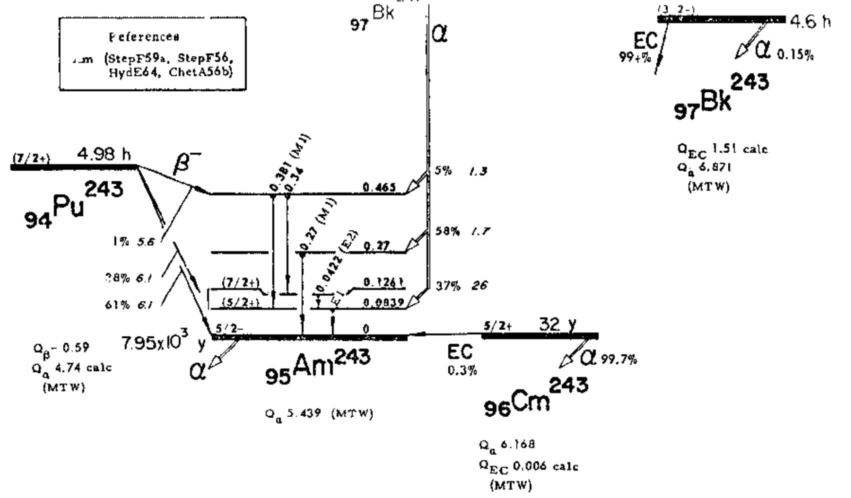
References
Np (AsaF60a)
Pu (BunkM59, AsaF57a, BjoS63b, AsaF61)



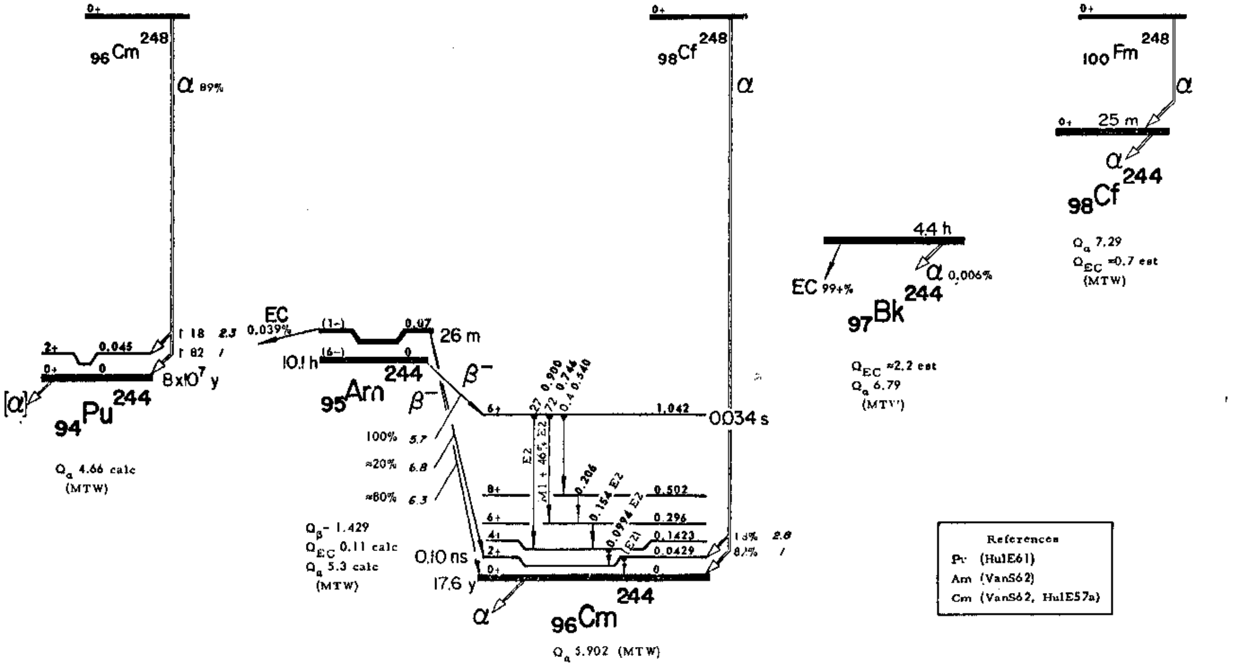
References
Pu (DzhB63b, BraiT65, HydE64)
Am (AhmI66, HydE64)



References
Pu (HulE61, AsaF60, HoffR55)
Am (AsaF60, LineA65)
Cm (ChetA56b, HumJ55, AsaF60, HoffR55)

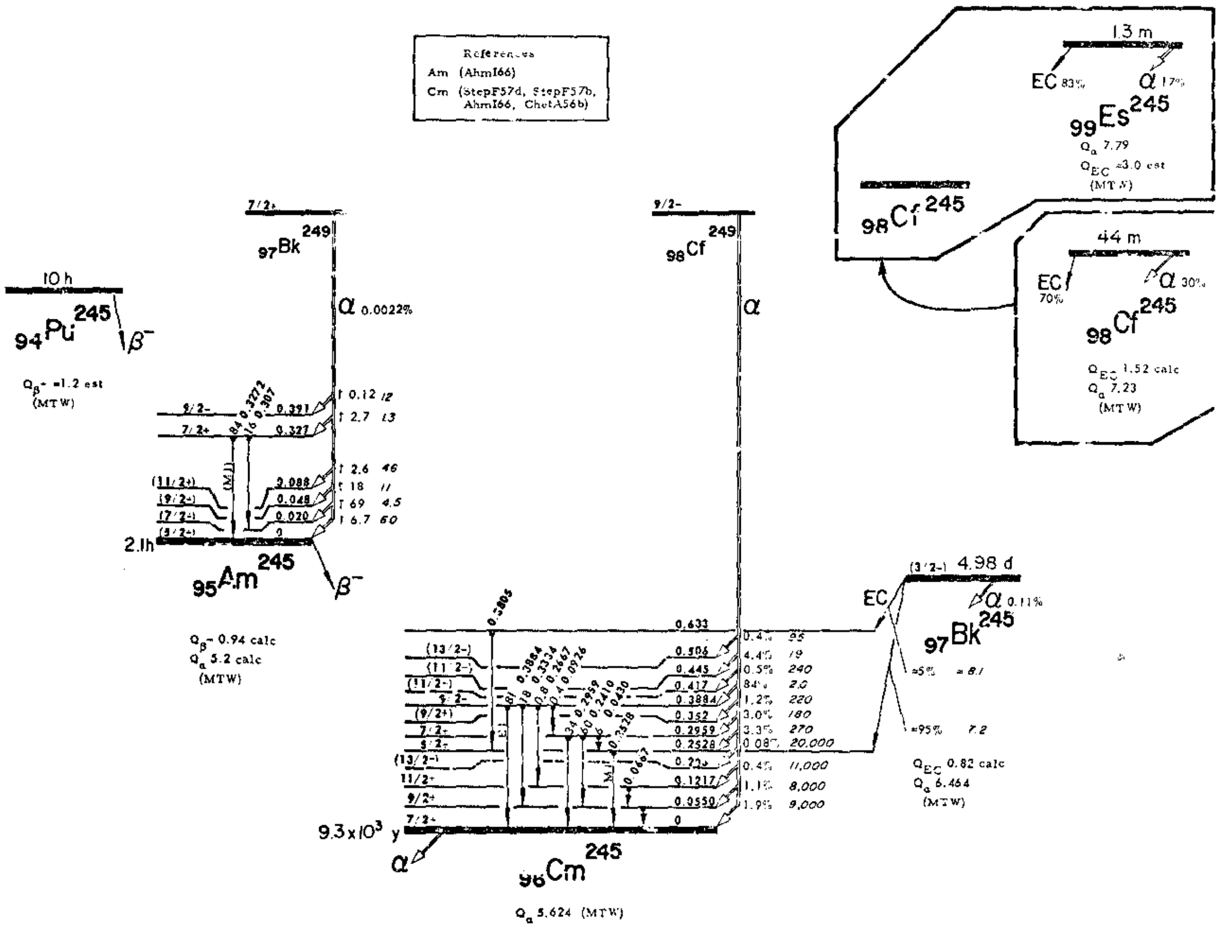


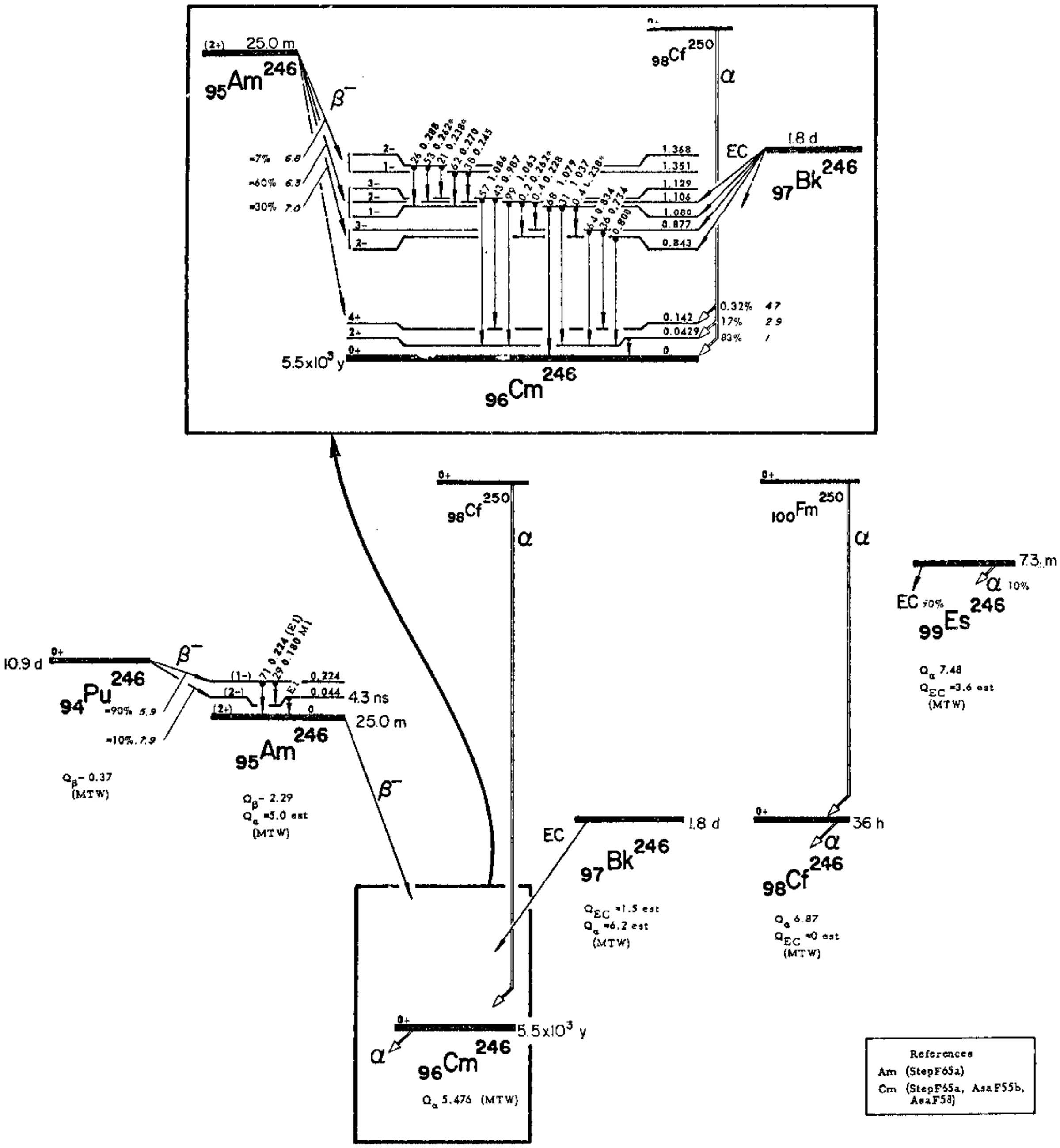
References
Am (StepF59a, StepF56, HydE64, ChetA56b)



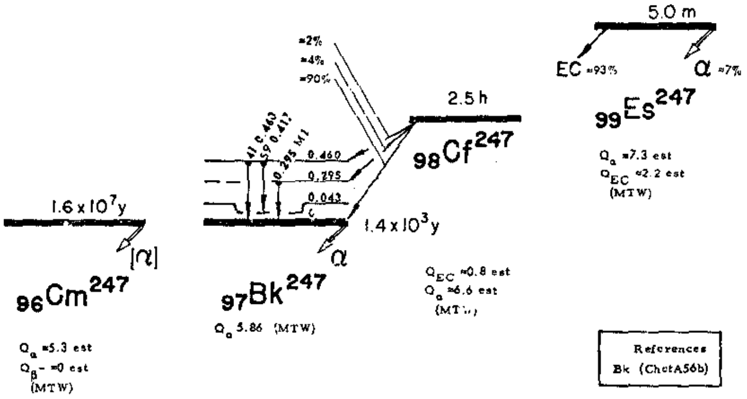
References
Pu (HulE61)
Am (VanS62)
Cm (VanS62, HulE57a)

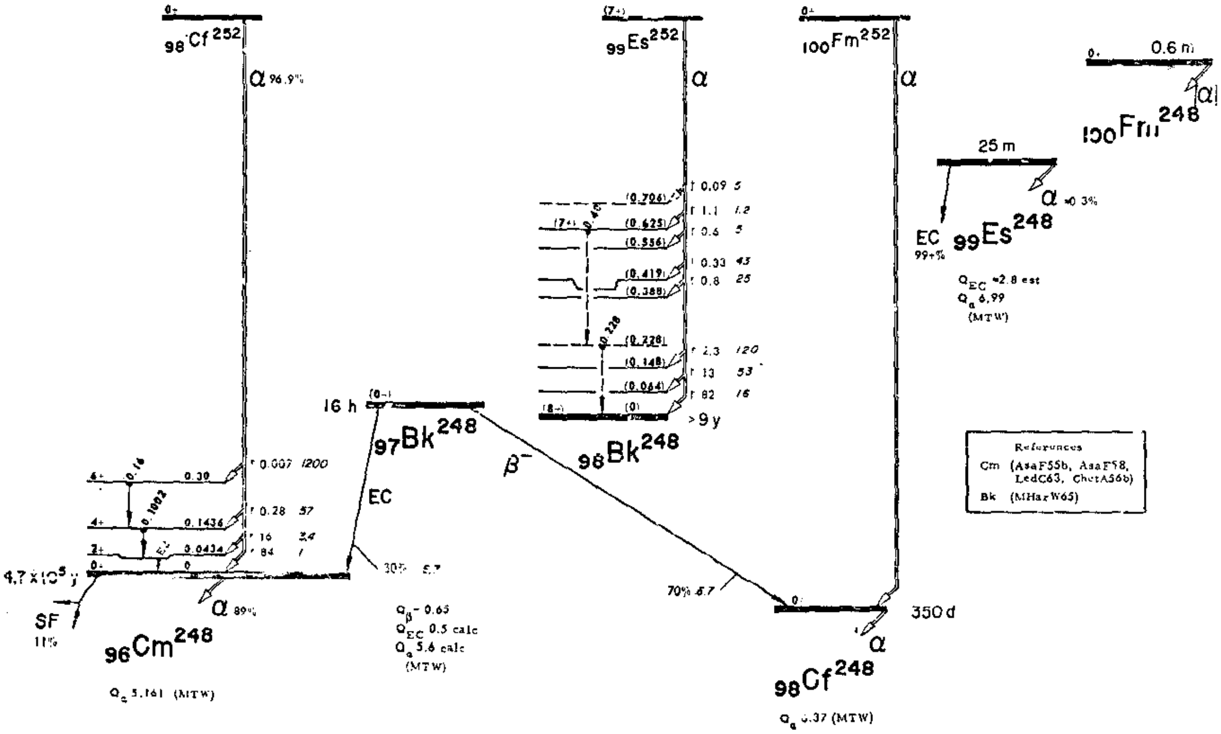
References
Am (AhmI66)
Cm (StepF57d, StepF57b, AhmI66, ChetA56b)

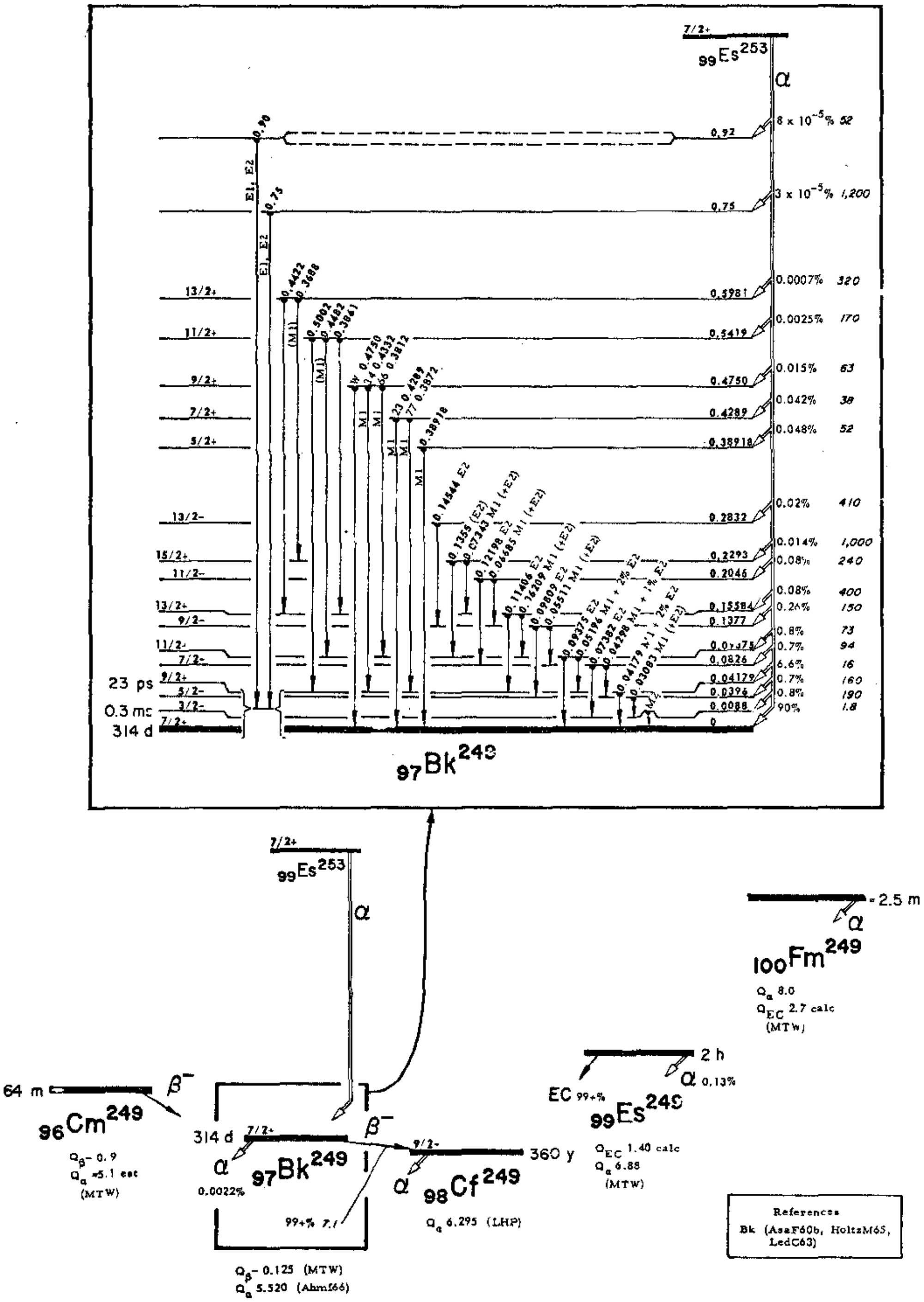




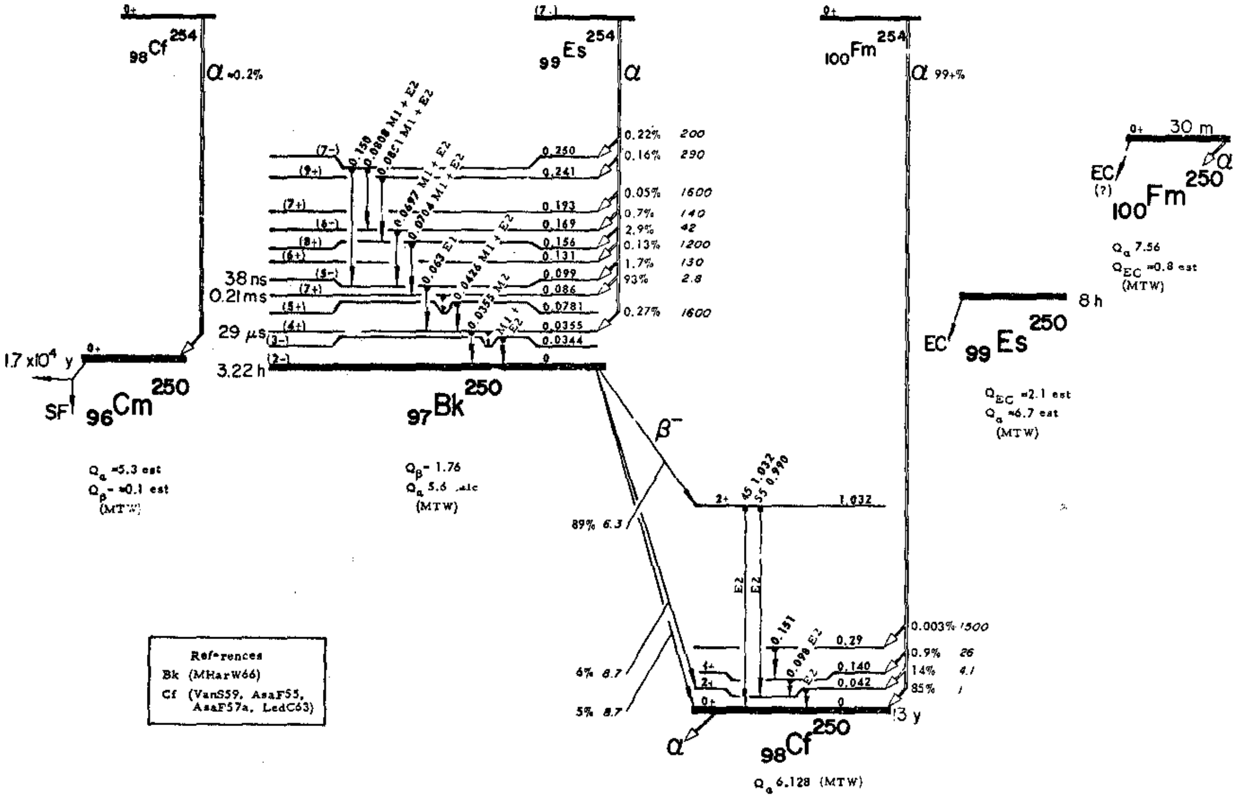
References
Am (StepF65a)
Cm (StepF65a, AsaF55b, AsaF58)


5.0 m
EC ≈93%
α ≈7%
$_{99}Es^{247}$
Q_α ≈7.3 est
Q_EC ≈2.2 est
(MTW)
≈2%
≈4%
≈90%
2.5 h
$_{98}Cf^{247}$
41 0.460
59 0.417
0.295 M1
0.460
0.295
0.043
0
1.4 x 10³y
α
1.6 x 10⁷y
[α]
$_{96}Cm^{247}$
$_{97}Bk^{247}$
Q_α 5.86 (MTW)
Q_EC ≈0.8 est
Q_α ≈6.6 est
(MTW)
Q_α ≈5.3 est
Q_β- ≈0 est
(MTW)
References
Bk (ChetA56b)

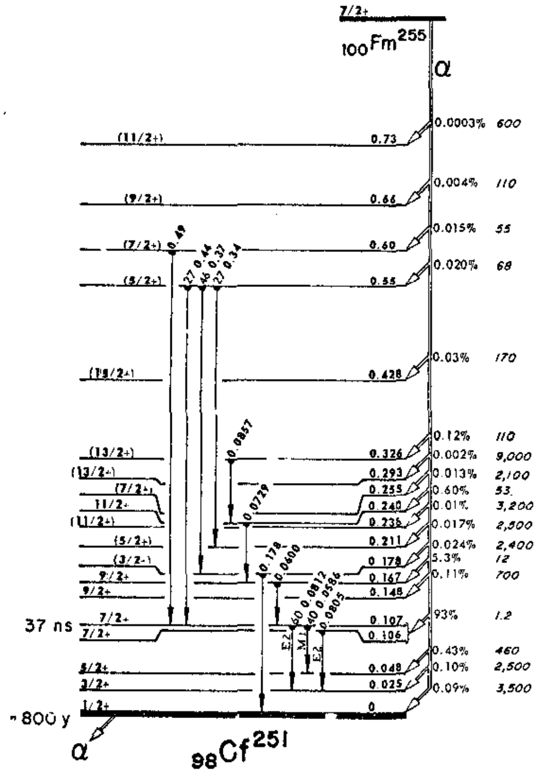

0+
$_{98}Cf^{252}$
α 96.9%
(7+)
$_{99}Es^{252}$
α
0+
$_{100}Fm^{252}$
α
0+
0.6 m
[α]
$_{100}Fm^{248}$
25 m
α ≈0.3%
EC
99+%
$_{99}Es^{248}$
Q_EC ≈2.8 est
Q_α 6.99
(MTW)
(0.706)
(7+)
0.40
(0.625)
(0.556)
(0.419)
(0.388)
0.228
(0.228)
(0.148)
(0.064)
(8+)
(0)
0.09 5
1.1 1.2
0.6 5
0.33 43
0.8 25
2.3 120
13 53
82 16
>9 y
$_{98}Bk^{248}$
16 h
(0+)
$_{97}Bk^{248}$
β⁻
EC
30% 5.7
70% 5.7
6+
0.16
0.30
0.007 1200
4+
0.1002
0.1436
0.28 57
2+
E2
0.0434
16 3.4
84 1
0+
0
4.7 x 10⁵ y
SF
11%
α 89%
$_{96}Cm^{248}$
Q_α 5.161 (MTW)
Q_β- 0.65
Q_EC 0.5 calc
Q_α 5.6 calc
(MTW)
0+
350 d
α
$_{98}Cf^{248}$
Q_α 6.37 (MTW)
References
Cm (AsaF55b, AsaF58, LedC63, ChetA56b)
Bk (MHarW65)



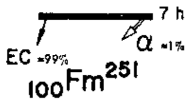
References
Bk (AsaF60b, HoltzM65, LedC63)



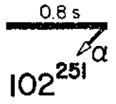
References
Bk (MHarW66)
Cf (VanS59, AsaF55, AsaF57a, LedC63)



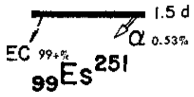
Q_α 5.94 (GrouCR66)



Q_{EC} ≈1.6 est
Q_α ≈7.4 est
(MTW)



Q_α 8.8
(LHP, MTW)



Q_{EC} ≈0.4 est
Q_α 6.59
(MTW)

References
Cf (AsaF64, AhmI66)

Cf^{252} (2.65 y):

α: α_0 **6.119** (84.3%), α_{43} **6.076** (15.5%), α_{144} **5.975** (0.28%) mag spect (AsaF55b, AsaF58)
others (MagL54, EasT57, LedC63)

γ: γ_1 **0.0433**, γ_2 **0.1002** mag spect conv (AsaF64a)
γ_1 **0.0434** mag spect conv (HollaJ56b)
γ_1 (γ 0.014%), γ_2 (γ 0.01%) scint spect, αγ coinc (AsaF55b)
γ_3 **0.16** (γ 2 x 10^{-3}%) γα coinc (LedC63)

Es^{252} (≈140 d):

α: α_0 **6.639** († 82), α_{64} **6.58** († 13), α_{148} **6.493** († 2.3), α_{388} **6.26** († 0.8), α_{419} **6.23** († 0.33), α_{556} **6.09** († 0.6), α_{582} **6.07** ? († 0.11), α_{625} **6.02** († 1.1), α_{659} **5.99** († 0.07), α_{706} **5.94** ? († 0.09) semicond spect (MHarW65)

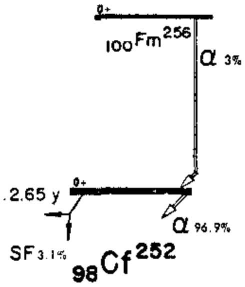
γ: **0.074** (γ 0.3%), **0.154** (γ 0.07%), **0.198** (γ 0.08%), **0.228** (γ 0.23%), **0.278** (γ 0.21%), **0.40** (γ 1.1%, complex), **0.52** (γ 0.15%), **0.57** (γ 0.14%) scint spect, γα coinc (MHarW65)

Fm^{252} (23 h):

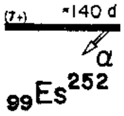
α: **7.05** ion ch (FrieA56)
7.06 ion ch (AmiS57a)
others (StuM54)

102^{252} (2.1 s):

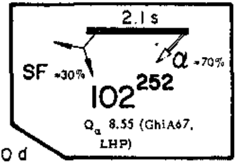
α: **8.41** semicond spect (GhiA67, MikV66a)
others (GhiA59, GhiA62)



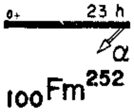
Q_α 6.217 (MTW)



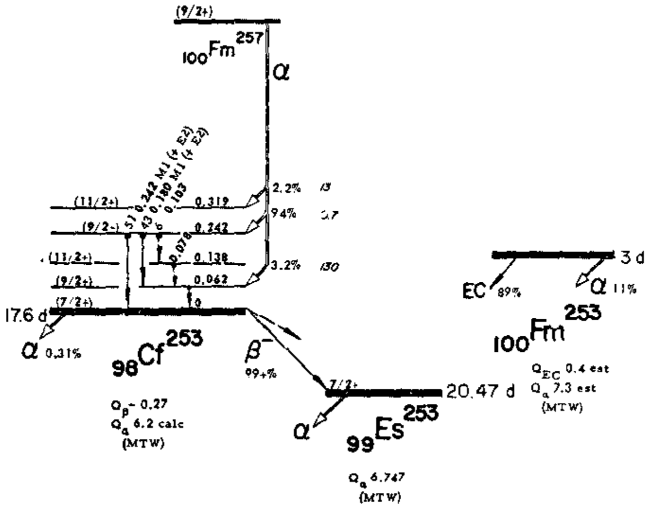
Q_α 6.75
(MHarW65, MTW)
Q_{EC} 1.1 calc
Q_{β^-} 0.2 calc
(MTW)

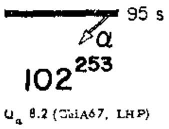


Q_α 8.55 (GhiA67, LHP)

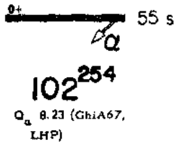


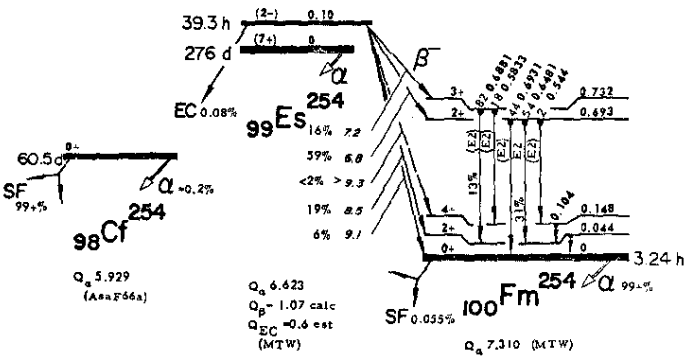
Q_α 7.17 (MTW)



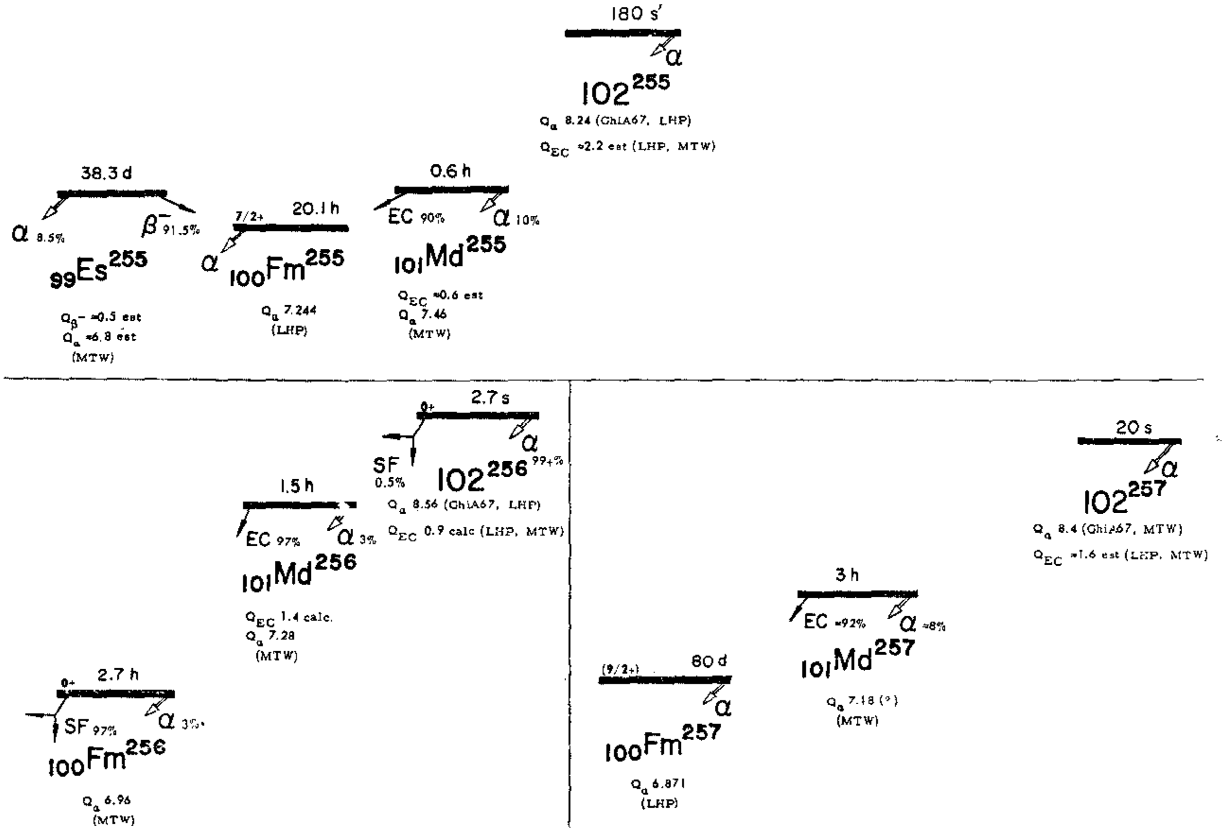


References
Cf (AsaF66b)





References
Es (MHarW65)
Fm (UniJ62, HollaJ63)


180 s′
α
102^255
Qα 8.24 (GhiA67, LHP)
QEC ≈2.2 est (LHP, MTW)
38.3 d
α 8.5%
β⁻ 91.5%
99Es^255
Qβ⁻ ≈0.5 est
Qα ≈6.8 est
(MTW)
7/2+
20.1 h
α
100Fm^255
Qα 7.244
(LHP)
0.6 h
EC 90%
α 10%
101Md^255
QEC ≈0.6 est
Qα 7.46
(MTW)
0+
2.7 s
α
SF
0.5%
102^256 99+%
Qα 8.56 (GhiA67, LHP)
QEC 0.9 calc (LHP, MTW)
1.5 h
EC 97%
α 3%
101Md^256
QEC 1.4 calc.
Qα 7.28
(MTW)
0+
2.7 h
SF 97%
α 3%
100Fm^256
Qα 6.96
(MTW)
20 s
α
102^257
Qα 8.4 (GhiA67, MTW)
QEC ≈1.6 est (LHP, MTW)
3 h
EC ≈92%
α ≈8%
101Md^257
Qα 7.18 (?)
(MTW)
(9/2+)
80 d
α
100Fm^257
Qα 6.871
(LHP)

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