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ORNL-4767

TRANSURANIUM PROCESSING PLANT

Semiannual Report of
PRODUCTION, STATUS, AND PLANS
Period Ending December 31, 1971

W. D. Burch

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CHEMICAL TECHNOLOGY DIVISION

TRANSURANIUM PROCESSING PLANT. ^{3#}
SEMIANNUAL REPORT OF
PRODUCTION, STATUS, AND PLANS
FOR PERIOD ENDING DECEMBER 31, 1971

W. D. Burch, J. E. Bigelow, and L. J. King

MAY 1972

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SUMMARY

This is the eighth report in a series that is being issued semiannually to inform the heavy-element community of the status and the future production plans of the Transuranium Element Production Program at ORNL.

During the period of July 1, 1971, through December 31, 1971, we recovered 10 g of ^{242}Pu , 181 g of ^{244}Cm , 26 mg of ^{249}Bk , 210 mg of ^{252}Cf , and 38 μg of ^{253}Es as purified products from nine reactor tubes that had been irradiated as part of the Californium-I program at the Savannah River Plant (SRP) to evaluate the commercial market for ^{252}Cf and to establish an inventory of ^{252}Cf for sale. We made 42 shipments, which contained the following total amounts of nuclides: 500 mg of ^{238}Pu , 500 mg of ^{240}Pu , 0.11 g of ^{244}Cm , 4.4 mg of ^{248}Cm , 3.1 mg of ^{249}Bk and 0.62 mg of isotopically pure ^{249}Cf in an impure transuranium product fraction, 92.2 mg of ^{252}Cf in a mixture of californium isotopes, 36 μg of isotopically pure ^{253}Es , and 6×10^8 atoms of ^{257}Fm .

During the next 18 months, we expect to recover 270 mg of ^{252}Cf from the processing of 11 SRP Pu-Al tubes and 565 mg of ^{252}Cf from the processing of irradiated HFIR targets. A total of 83 mg of ^{249}Bk and 2.8 mg of ^{253}Es will also be recovered as by-products from these campaigns.

We determined that, by irradiating curium from the Californium-I program in the HFIR, we could readily produce all of the ^{252}Cf that would be required to meet the postulated demands (from 0.5 g/year in 1972 up to 1.8 g/year in 1980) of both the Research and the Production Divisions. Alternatively, by irradiating only curium which resulted from the Curium-II campaign at Savannah River, we would be limited to a steady-state production rate of 0.5 g/year.

The processes and equipment used to process the SRP Pu-Al tubes were generally the same as those used to process HFIR targets and the SRP slugs. However, an 11-ft-long dissolver was required to dissolve the 5-ft-4-in.-long sections of SRP tubes, and a different procedure was used to dissolve the aluminum from the tubes.

A new technique was developed for producing uniform particles of curium oxide in the desired size range for use in fabricating HFIR targets. The curium is sorbed on Dowex-50 resin, which is then calcined to convert the curium to the oxide form. Six targets, which were fabricated from oxide produced by this technique, are being irradiated in the HFIR.

Nineteen neutron sources were fabricated during this period, bringing the total fabricated to date to 49.

In special projects, (1) a special target was removed from the HFIR and was cut up to recover the components for various experimental purposes, (2) two additional gamma-ray sources were fabricated (one each from 500 mg of ^{240}Pu and 500 mg of ^{238}Pu) for use in Mössbauer studies, and (3) a rabbit containing 250 μg of ^{240}Pu was fabricated, irradiated, and shipped to F. Asaro at LBL.

The values that we are currently using for transuranium-element decay data and for cross-section data in planning irradiation-processing cycles, calculating production forecasts, and assaying products are tabulated in the Appendix.

1. INTRODUCTION

This is the eighth report in a series that is being issued semi-annually to inform the heavy-element community of the status and the future production plans of the Transuranium Element Production Program at ORNL. The objective of these reports is to provide information that will enable users of the products to obtain maximum service from the production facilities at ORNL. Production plans and schedules are definitely established only for the short term; long-range plans can be (and are) markedly influenced by feedback from researchers and other users of transuranium elements.

TRU operations during the report period are summarized. Quantities of materials that were recovered and shipped are specified, and proposed processing schedules and anticipated yields of various products are presented. Special processing, fabrication, and irradiation programs are described. The original and current contents of all neutron sources that have been made at TRU, as well as the individuals to whom these sources have been loaned, are tabulated. Values of nuclear parameters which were used as input data for the calculations of production rates for transuranium elements, along with a listing of the parameters which were used to calculate the specific activities of the isotopes that are of interest to TRU, are included in the Appendix.

Previous reports in this series are:

1. For period ending June 30, 1968 - ORNL-4376.
2. For period ending December 31, 1968 - ORNL-4428.
3. For period ending June 30, 1969 - ORNL-4447.
4. For period ending December 31, 1969 - ORNL-4540.
5. For period ending June 30, 1970 - ORNL-4588.
6. For period ending December 31, 1970 - ORNL-4666.
7. For period ending June 30, 1971 - ORNL-4718.

2. PROCESSING SUMMARY AND PRODUCTION ESTIMATE

The isotopic concentrations of the various transuranium elements are not constant, but are functions of irradiation histories and decay times. We have selected one isotope of each element to use in making material balances for the isotopic mixtures normally handled in TRU. Thus, we usually trace curium by the isotope ^{244}Cm . Except in special instances, ^{242}Pu , ^{243}Am , ^{249}Bk , ^{252}Cf , and ^{253}Es are the isotopes used for tracing the corresponding elements. Throughout this report section, we are discussing mixtures of isotopes when we do not stipulate "isotopically pure."

2.1 Processing Summary

We recovered 9.6 g of ^{242}Pu , 181 g of ^{244}Cm , 26.4 mg of ^{249}Bk , 209.5 mg of ^{252}Cf , and 37.7 μg of ^{253}Es as purified products from nine reactor tubes that had been irradiated as part of the Californium-I program at the Savannah River Plant (SRP) to evaluate the commercial market for ^{252}Cf and to establish an inventory of material for sale.

Forty-two shipments (Table 2.1) that were made from TRU during this period contained the following amounts of nuclides: (1) 500 mg of ^{238}Pu ; (2) 500.25 mg of ^{240}Pu ; (3) 0.11 g of ^{244}Cm ; (4) 4.43 mg of ^{248}Cm (in products containing 97% ^{248}Cm); (5) 3.1 mg of ^{249}Bk and 0.62 mg of isotopically pure ^{249}Cf in an impure transcurium product fraction; (6) 92.2 mg of ^{252}Cf in a mixture of californium isotopes (40.4 mg of this ^{252}Cf was included in shipments of Californium-I material to SRP); (7) 36.0 μg of isotopically pure ^{253}Es ; and (8) 6×10^8 atoms of ^{257}Fm .

Table 2.1. Distribution of Heavy Elements from the Transuranium Processing Plant During the Period July 1, 1971 - December 31, 1971

Major Nuclide	Date	TRU File Number	Shipped To	
			Individual	Site
<u>Plutonium-238, μg</u>				
500 (GS-6)	11-09-71	460	P. G. Huray	ORNL-TRL
<u>Plutonium-240, mg</u>				
0.25 (irradiated)	8-20-71	434	F. Asaro	LBL
<u>500</u> (GS-5)	9-30-71	459	P. G. Huray	ORNL-TRL
500.25				
<u>Curium-244, g</u>				
0.01	8-06-71	454	M. M. Abraham	ORNL-TRL
<u>0.10</u>	8-19-71	457	M. M. Abraham	ORNL-TRL
0.11				
<u>Curium-248 (97%), mg</u>				
0.003	7-09-71	402	Research Pool	ORNL
3.15	11-01-71	404	R. J. Silva	ORNL-TRL
0.16	12-03-71	405	R. W. Hoff	LLL
<u>1.12</u>	12-17-71	466	S. M. Fried	ANL
4.433				
<u>Berkelium-249, mg</u>				
3.1	10-20-71	411	R. W. Hoff	LLL
<u>Californium-249 (isotopically pure), mg</u>				
0.62	10-20-71	411	R. W. Hoff	LLL

Table 2.1 (continued)

Major Nuclide	Date	TRU File Number	Shipped To	
			Individual	Site
<u>Californium-252, mg</u>				
0.01	7-09-71	450	Isotopes Sales	ORNL
0.197 (NSD-47)	7-20-71	446	J. L. Cason	Gamma Ind. ^a
0.192 (NSD-48)	7-20-71	446	J. L. Cason	Gamma Ind. ^a
0.196 (NSD-49)	7-20-71	446	J. L. Cason	Gamma Ind. ^a
0.465 (NSD-20)	8-04-71	455	F. F. Haywood	ORNL-DOSAR
1.95	9-07-71	373	J. A. Harris	LBL
9.69 (NSD-37)	9-07-71	384	R. W. Perkins	PNL
4.81 (NSS-17)	9-07-71	438	L. W. Dahlke	Sandia-Liv.
0.276 (NSD-52)	9-07-71	375	E. D. Clayton	PNL
1.75 (NSD-45)	9-07-71	441	K. L. Swint	PNL
0.027	9-20-71	462	Isotopes Sales	ORNL
21.16	10-05-71	458	A. R. Boulogne	SRL
0.012	10-15-71	463	Isotopes Sales	ORNL
11.22 (NSD-29)	11-04-71	374	E. M. Murray	Y-12
0.928 (NS-39)	11-12-71	395	V. Spiegel	NBS
5.04 (NSD-41)	11-12-71	443	C. J. Emert	BAPL
4.37 (NSD-42)	11-12-71	443	C. J. Emert	BAPL
0.359 (NSD-51)	11-12-71	444	L. C. Nelson, Jr.	New Brunswick
1.04 (NSD-53)	11-12-71	461	L. J. Esch	KAPL
0.136 (NS-50)	11-19-71	439	S. G. Carpenter	ANL (NRTS)
1.73 (NZD-31)	11-29-71	382	C. N. Jackson	WADCO
1.77 (NZD-32)	11-29-71	382	C. N. Jackson	WADCO
1.86 (NZD-33)	11-29-71	382	C. N. Jackson	WADCO
1.90 (NZD-34)	11-29-71	382	C. N. Jackson	WADCO
1.88 (NZD-35)	11-29-71	382	C. N. Jackson	WADCO
19.24	12-13-71	464	A. R. Boulogne	SRL
<u>0.018</u>	12-14-71	465	Isotopes Sales	ORNL
92.226				

Table 2.1 (continued)

Major Nuclide	Date	TRU File Number	Shipped To	
			Individual	Site
<u>Einsteinium-253</u> (isotopically pure), µg				
25.8	8-04-71	429	R. W. Hoff	LLL
10.0	8-04-71	456	R. D. Baybarz	ORNL-CTD
<u>0.2</u>	8-04-71	452	T. C. Parsons	LBL
36.0				
<u>Fermium-257, atoms</u>				
~6 x 10 ⁸	7-13-71	431	R. M. Latimer	LBL

^aFor the use of P. L. Johnson, Mound Laboratory.

2.2 Processing Proposed Prior to June 1973

The amounts of transcurium elements that will be produced at TRU during the next few years will depend upon: (1) the needs of researchers for various isotopes, (2) the needs for ^{252}Cf in the Production Division's market evaluation and sales program, and (3) the capabilities at TRU to produce the required materials.

Table 2.2 lists the estimated future production of transcurium elements from a series of likely processing campaigns which are scheduled through June 1973. The campaigns to process HFIR targets are scheduled essentially as dictated by the times required for target fabrication, irradiation, and processing. We have firm plans to process six SRP Pu-Al tubes during the spring of 1972 and have tentative plans to process five more in the fall of 1972. We requested approval of a proposal to irradiate 200 g of Californium-I curium to increase the amount of materials available to researchers. This quantity of irradiated curium would produce approximately 30 mg of ^{249}Bk , 1500 μg of ^{253}Es in mixed einsteinium isotopes, and 300 μg of isotopically pure ^{253}Es for the Research Division, along with 300 mg of ^{252}Cf for the Production Division. We have already received approval to irradiate 100 g of the Californium-I curium and, therefore, have started to fabricate ten targets, which will be irradiated for seven months and then processed by December 1972. This does not represent an optimum irradiation; processing is scheduled to permit return of the curium to SRP during 1972. If approved, a second 100-g batch of the curium would be irradiated and processed by April 1973.

2.3 Estimates of the Availability of Transuranium Elements Prior to June 1973

The plutonium, americium, and curium that are separated from the transcurium elements during the processing of irradiated targets are considered to be intermediate feed materials rather than products. Isotopes of these elements are not normally discussed in this report. However, we will soon be able to "milk" about 9 mg of ^{248}Cm every six months from 90 mg of ^{252}Cf , which has been made available for this purpose through an agreement between the Production and Research Divisions.

Table 2.2. Estimated Future Production of Transcurium Elements

Period	Processing Campaign	Products of Campaigns			²⁵² Cf Production ^b		
		²⁴⁹ Bk (mg)	²⁵² Cf (mg)	²⁵³ Es ^a (μg)	During the Period (mg)	Cumul. (mg)	Date Products Available
Through December 1971						135 ^b	
January-June 1972	9 TRU-HFIR targets	7	70	350(70)			February 1972
	6 SRP Cf-I tubes	15	150 ^b	0 ^c	70	205	June 1972
July-December 1972	10 TRU-HFIR targets	5	50	250(50)			July 1972
	5 SRP Cf-I tubes	12	120 ^b	0 ^c			November 1972
	10 SRP-Cm targets	15	150	750(150)	200	405	December 1972
January-June 1973	5 TRU-HFIR targets	6	60	300(60)			January 1973
	10 SRP-Cm targets	15	150	750(150)			April 1973
	13 TRU-HFIR targets	8	85	425(85)	295	700	June 1973
July-December 1973					100-380 ^d		
1974					300-760 ^d		

^aAmounts from initial separation. Amounts "milked" from californium product fraction after decay period are given in parentheses.

^bCalifornium produced in the SRP campaigns is not included in production totals. A total of 416 mg has been recovered from 164 SRP slugs and 9 SRP tubes processed through December 1971.

^c²⁵⁴Es (~3 μg) can be recovered.

^dThe lesser amount will be produced if feed curium is limited to current TRU stock plus Curium-II curium (95% ²⁴⁴Cm). The higher amount will be produced if Californium-I curium is used as feed for the HFIR.

The amounts of ^{249}Bk , ^{252}Cf , and ^{253}Es that are anticipated to be produced in the next 18 months are listed in Table 2.2. About 1×10^9 , 7.5×10^8 , 2×10^9 , 9×10^8 , 2×10^9 , and 1.3×10^9 atoms of ^{257}Fm will become available in February 1972, July 1972, December 1972, January 1973, April 1973, and June 1973, respectively.

2.4 Processing After June 1973

The rate at which transuranium elements will be produced at TRU after June 1973 will depend heavily on how much ^{252}Cf is produced and recovered in the TRU-HFIR complex for the Production Division's Sales-Loan Program. The paragraphs below discuss the production rates of ^{252}Cf in the HFIR for: (1) large-scale irradiations of curium from the Californium-I program, and (2) irradiation of curium from the Curium-II program. Also, the corresponding rates of production of ^{249}Bk , ^{253}Es , and ^{257}Fm are given in the following section of this report.

However, the HFIR will not be loaded to its capacity with either type of curium, pending a decision by the AEC concerning the quantity of each type to be irradiated in this reactor.

The long-term capability of the TRU-HFIR complex to produce transuranium elements was discussed in a previous report¹ in this series. In October 1971, we reexamined that capability using more recent HFIR-effective neutron cross sections and more recent predictions of the isotopic content of the Californium-I program curium. We did not optimize the irradiation-processing cycles for maximum ^{252}Cf production but merely determined that the postulated demands could be achieved by using only about 80% of the total HFIR capacity (since we did not include the use of the reflector).

The objective of this new study was to determine whether TRU-HFIR could produce enough ^{252}Cf to meet the postulated demands of both the Research Division and the Production Division through 1980. The top curve in Fig. 2.1 shows the total demands for ^{252}Cf , which increase steadily from 0.5 g/year in 1972 to 1.3 g/year in 1977. Although not shown on this curve, the demands continue to increase to 1.8 g/year in 1980. In the study it was

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Fig. 2.1. ^{252}Cf Production Rates - Total Required and Production Rates in HFIR Using Curium from Californium-I or Curium-II Programs.

assumed that all of the SRP Pu-Al tubes would be processed at TRU within five years. Our approach was to assume that curium having the composition of the curium in the tubes would be irradiated in the HFIR as long as the ^{252}Cf demand could be met without having to recycle the "better" curium produced by the first-cycle irradiations.

The second curve from the top shows the rates of production of ^{252}Cf in TRU-HFIR according to our study. This curve does not represent our maximum capability for two reasons: (1) we could produce more ^{252}Cf in the period 1974-1976 by recycling the heavier curium (produced by initial irradiations) as soon as it becomes available, and (2) results of a HFIR target campaign completed since the calculations were made indicate that we currently underestimate ^{252}Cf production from irradiation (in the HFIR) of curium containing large amounts of the heavier isotopes. Apparently the destruction of ^{247}Cm and ^{251}Cf by fission is reduced by self-shielding at high concentrations.

According to our study, although only half of the mass of Californium-I curium would be left in 1980, it would still contain 77% of the "potential californium" (i.e., the total californium that eventually can be produced from the presently available curium feeds). This curium could be used to sustain a production rate of 1.5 to 2 g/year at least through 1990 or to produce ^{252}Cf at an increasing rate, reaching nearly 3 g/year in 1982.

The two bottom curves in Fig. 2.1 represent the ^{252}Cf production rates that would result from irradiating present TRU stocks of curium (including 200 g of Californium-I curium), plus 200 g of curium (95% ^{244}Cm) per year from the Curium-II program. The next-to-the-bottom curve is our earlier prediction, while the bottom curve corresponds to the new projection. In each case, the steady-state production rate of ^{252}Cf is 0.5 g/year. The difference between the curves shows the short-term effects that the processing of Californium-I materials has had on the production of research materials.

The diminished rate of production of ^{252}Cf in the HFIR results in a reduction in the availability of other transuranium elements. The requirements for ^{252}Cf for research in the near future can be met by using ^{252}Cf

recovered from the Californium-I tubes; however, short-lived isotopes, such as ^{253}Es and ^{257}Fm , cannot be supplied by the processing of SRP tubes.

2.5 Availability of ^{249}Bk , ^{253}Es , and ^{257}Fm After June 1973

The other isotopes of major interest to the Research Division program, ^{249}Bk , ^{253}Es , and ^{257}Fm , are produced in TRU-HFIR at rates that are nearly proportional to the rate of ^{252}Cf production. For our usual irradiation cycles, the rates are: ^{249}Bk , 10% of the ^{252}Cf rate; ^{253}Es , 0.5% of the ^{252}Cf rate; and ^{257}Fm , 1.5×10^7 atoms per mg of ^{252}Cf . Of these, only ^{249}Bk can be recovered from the SRP Pu-Al tubes (about 2 mg per tube). Table 2.3 contains estimates of the amounts of these isotopes that would be produced by (1) full-scale production of ^{252}Cf at TRU-HFIR for both the Research Division and the Production Division, using curium from the Californium-I program, and (2) production of ^{252}Cf for the Research Division alone. These estimates were obtained from the curves of Fig. 2.1.

3. PROCESSES AND EQUIPMENT

The steps that were used in processing the SRP Pu-Al reactor tubes were: (1) dissolution of the aluminum using NaNO_3 and NaOH followed by dissolution of the actinide oxide using concentrated HNO_3 ; (2) recovery of plutonium using the Pubex batch extraction process; (3) removal of ionic impurities, and conversion from a nitrate to chloride medium, using the Cleanex batch extraction process; (4) decontamination of the transplutonium elements from fission products, using batch extractions based on the Tramex process; (5) separation of curium from the transcurium elements, using LiCl -based anion exchange; and (6) separation and purification of berkelium, californium, einsteinium, and fermium. Final purification of the plutonium and conversion of the curium to curium oxide have not yet been done.

The processes and equipment used were generally the same as those used to process HFIR targets and the SRP slugs. The major differences were:

Table 2.3. Estimated Production of ^{249}Bk , ^{253}Es , and ^{257}Fm in the HFIR

Year	^{249}Bk		^{253}Es		^{257}Fm	
	Prod. and Res. ^a (mg)	Res. Only ^b (mg)	Prod. and Res. ^a (μg)	Res. Only ^b (μg)	Prod. and Res. ^a (atoms)	Res. Only ^b (atoms)
1973	85	30	3000	1600	0.9×10^{10}	0.5×10^{10}
1974	90	35	3900	1700	1.2×10^{10}	0.5×10^{10}
1975	100	40	4300	2000	1.3×10^{10}	0.6×10^{10}
1976	115	45	5800	2300	1.7×10^{10}	0.7×10^{10}
1977	130	50	6900	2500	2.0×10^{10}	0.75×10^{10}
1978	150	50	7800	2500	2.3×10^{10}	0.75×10^{10}
1979	170	50	8800	2500	2.6×10^{10}	0.75×10^{10}
1980	160	50	8100	2500	2.4×10^{10}	0.75×10^{10}

^aProduction and research.

^bResearch only.

(1) We replaced our dissolver vessel with an 11-ft-long dissolver to permit dissolution of the 5-ft-4-in.-long sections of SRP tubes. The new vessel has a small-diameter bottom section to make it useful for dissolving HFIR targets.

(2) The procedure used to dissolve the aluminum from the tubes was different from that used for dissolving aluminum from HFIR targets and SRP slugs. The capacity of the dissolver off-gas system is insufficient to handle hydrogen at the rate that it would be evolved if we contacted a tube (about 4 kg of aluminum) with sufficient 3 M NaOH--1.5 M NaNO₃ solution (about 120 liters) to dissolve the aluminum and then heated the solution to initiate the reaction. Therefore, we placed a tube, along with 85 liters of 2.1 M NaNO₃ solution (heated to 92°C), in the dissolver. Then we slowly added 36.5 liters of 10 M NaOH solution. The addition rate was limited in order to keep the temperature difference between the solution and the dissolver heating-cooling jacket below 12°C and thereby keep the aluminum dissolution rate sufficiently low that the dissolver off-gas vacuum would be maintained and the hydrogen content in the dissolver vapor space would be less than 4%.

(3) We were surprised by the large amounts of solids present and, at first, had difficulties filtering both the waste dejacketing solutions and the nitric acid dissolver solutions. We had already increased the sizes of the filters considerably, based on extrapolation of our experiences with the SRP slugs; however, we had to fabricate even larger filters to use in processing the tubes.

(4) Aluminum that was present in the acidic dissolver solutions (up to 200 g in the composite feed solution for one three-tube campaign) caused minor difficulties in all of the succeeding process steps: Pubex, Cleanex, Tramex, and LiCl-based anion exchange.

3.1 Preparation of Curium Oxide for HFIR Targets

A new technique has been developed for producing uniform particles of curium oxide in the desired size range for use in fabricating HFIR targets. This technique has replaced the sol-gel method, in which excessive amounts of recycle were required.

Curium is loaded onto Dowex-50 resin from a dilute (0.05-0.5 M) nitric acid solution, and the column is rinsed with water. The resin (approximately 7.0 ml of resin per gram of curium) is contained in a quartz column, which can be heated in a vertical cylindrical furnace after loading. The column design provides for a positive gas flow through the resin bed during the firing procedure. After the resin has been loaded and washed, the column is placed in the furnace and heated at 140°C for approximately 2 hr in an air stream to thoroughly dry the resin. The resin is then heated in an air stream for 5 hr at 800°C to burn the carbon, 5 more hr at 800°C in 4% H₂--argon, and an additional 5 hr at 800°C in air. The product is then removed from the quartz column and heated in a platinum crucible for 5 hr at 1100°C.

The hydrogen-argon firing step is required to produce curium oxide having a satisfactory density and crushing strength. The reason for this behavior appears to be related to sulfur removal. Analysis of curium oxide products indicates that the sulfur content is decreased from 5% to about 1.5% when the hydrogen-argon firing step is used.

This procedure produces curium oxide having an adequate density. X-ray and metallographic examinations of sections of pellets made from curium oxide formed by this technique indicated that the dispersion within the pellet was satisfactory.

3.2 Fabrication of Targets

During this report period, six HFIR targets were fabricated using actinide oxide formed by the resin loading--calcination technique. Each target contained 10 g of actinide metal in the form of actinide oxide--aluminum pellets that were pressed to 80% of the theoretical density of the pellet core. In two of the targets, the actinide content was about 3 g of ²⁴³Am and 7 g of curium (7.4% ²⁴⁶Cm). Each of the other four contained 0.7 g of ²⁴⁰Pu-²⁴³Am and 9.3 g of curium (17.8% ²⁴⁶Cm). These targets are now being irradiated in the HFIR.

4. NEUTRON SOURCES

Much of the californium recovered at TRU is incorporated into sources, which are subsequently loaned to researchers. Data for all of the neutron sources that have been fabricated at TRU are listed in Table 4.1. Those that are standard models, NSS (singly encapsulated) and NSD (doubly encapsulated), are indicated in the table. A "standard" source is one which has been fabricated in the form shown in Fig. 4.1 of ref. 2 and has been cataloged by the AEC Division of Materials Licensing. Thus far, only sources fabricated from type 304L stainless steel have been cataloged. The characteristics of standard sources were listed in the previous report in this series.²

4.1 Sources Fabricated During July-December 1971

Nineteen sources were fabricated during this report period. Eleven of these, NSD-29, -37, -41, -42, -45, -47, -48, -49, -51, -52, and -53 are standard TRU sources doubly encapsulated in type 304L stainless steel. Source NSS-17 is a standard source singly encapsulated in type 304L stainless steel. Sources NSD-31, -32, -33, -34, and -35 were fabricated in standard TRU form but are doubly encapsulated in Zircaloy-2. Sources NS-39 and NS-50 were fabricated in nonstandard forms specified by the users. Also, during this period, source NSS-20 was converted from the standard singly encapsulated type to the standard doubly encapsulated type (NSD-20) to permit its use in an area which is not equipped as a containment zone for radioactivity.

4.2 Calibration of ^{252}Cf Sources Using a Fast-Neutron Detector

Two additional matters relating to the assay of ^{252}Cf sources and shipping containers were investigated. For the first time, we had time available and a source of sufficient strength to investigate the depression of count rates due to coincidence in the counter system. We compared a shipping package (containing about 39 mg of ^{252}Cf) with our counting standard, NSS-19, in three different counting geometries. Nearly 17% of the counts were lost at the highest count rate (about 15,000 counts/sec). The following empirical relationship between the fraction of the counts lost (FL) and

Table 4.1. Data for Neutron Sources Prepared at TRU

Source	Date of Calibration	^{252}Cf	^{252}Cf	^{248}Cm	On Loan To	
		Content at Calibration (μg)	Content as of 12-31-71 (μg)	Content as of 12-31-71 (μg)	Individual	Site
NS-1 ^a	8-28-68	311	130	b	K. L. Swint	PNL
NS-2	8-23-68	250	104	b	J. E. Powell	Sandia-NM
NS-3	5-13-69	~90	~46	b	H. E. Banta	ORAU
NS-4	7-09-69	870	457	397	C. F. Masters	LASL
NS-5 ^c	8-16-69	932	500	412	F. B. Simpson	ANC
NS-6	11-21-69	736	424	298	R. W. Hoff	LLL
NS-7	1-21-70	776	467	295	T. F. Handley	ORNL
NS-8	12-17-69	1811	1062	714	H. Berger	ANL
NSD-9	4-17-70	1694	1083	583	N. D. Wogman	PNL
NSS-10	3-11-70	111	69	b	J. P. Balagna	LASL
NS-11	3-10-70	8	5	b	R. R. Fullwood	LASL
NSS-12	6-30-70	1840	1241	571	R. W. Hoff	LLL
NSD-13	3-19-71	4578	3729	810	H. O. Menlove	LASL
NSS-14	6-29-70	4545	3063	1413	D. C. Stewart	ANL
NS-15 ^c	6-25-70	917	616	287	F. B. Simpson	ANC
NSD-16	10-08-70	1632	1182	429	R. Yoshimura	Sandia-NM
NSS-17	8-31-71	4612	4409	384	L. W. Lahlke	Sandia-Liv.
NS-18 ^c	6-24-70	947	636	297	F. B. Simpson	ANC
NSS-19	6-26-70	486	327	152	J. E. Bigelow	ORNL-TRU
NSD-20	7-01-70	620	419	192	F. F. Haywood	ORNL
NSS-21	10-21-70	18	13	b	F. Cross	PNL
NS-22	9-10-70	13	10	b	W. S. Lyon	ORNL
NSD-24	10-17-70	8	6	b	J. B. Davidson	ORNL
NS-25	11-09-70	57	42	b	F. J. Muckenthaler	ORNL
NSD-26	2-11-71	14	11	b	H. O. Menlove	LASL
NSD-27	1-29-71	2430	1910	496	J. E. Powell	Sandia-NM
NSD-28	2-12-71	11	9	b	E. E. Hicks	Rocky Flats
NSD-29	9-10-71	11220	10126	1043	E. M. Murray	Y-12

Table 4.1 (continued)

Source	Date of Calibration	²⁵² Cf Content at Calibration (μ g)	²⁵² Cf Content as of 12-31-71 (μ g)	²⁴⁸ Cm Content as of 12-31-71 (μ g)	On Loan To	
					Individual	Site
NSD-30	3-31-71	866	715	144	E. B. Darden	ORNL
NZD-31	11-23-71	1729	1682	45	C. N. Jackson	WADCO
NZD-32	11-23-71	1773	1725	45	C. N. Jackson	WADCO
NZD-33	11-23-71	1859	1809	48	C. N. Jackson	WADCO
NZD-34	11-23-71	1895	1844	49	C. N. Jackson	WADCO
NZD-35	11-23-71	1875	1625	48	C. N. Jackson	WADCO
NS-36 ^c	3-23-71	2039	1665	357	F. B. Simpson	ANC
NSD-37	9-04-71	9689	8707	936	R. W. Perkins	PNL
NSD-38	6-16-71	100	87	b	H. O. Menlove	LASL
NS-39	11-07-71	928	893	33	V. Spiegel	NBS
NSD-41	11-08-71	5039	4851	179	C. J. Emert	BAPL
NSD-42	11-02-71	4367	4186	173	C. J. Emert	BAPL
NSD-45	8-18-71	1749	1588	154	K. L. Swint	PNL
NSD-47	7-14-71	197	174	22	P. L. Johnson	Mound
NSD-48	7-14-71	191	169	21	P. L. Johnson	Mound
NSD-49	7-14-71	196	173	22	P. L. Johnson	Mound
NS-50	8-23-71	136	124	11	S. G. Carpenter	ANL-NRTS
NSD-51	11-02-71	359	344	14	L. C. Nelson, Jr.	New Brunswick
NSD-52	9-02-71	276	248	27	E. D. Clayton	PNL
NSD-53	10-25-71	1035	986	47	L. J. Esch	KAPL
SR-Cf-167 ^d	5-26-71	3915	3346	543	H. W. Dickson	ORAU

^aThis source is encapsulated in aluminum.

^bThis source is not suitable for recovery of ²⁴⁸Cm.

^cThis source is encapsulated in type 405 stainless steel.

^dThis source was fabricated at TRU in standard Savannah River SR-Cf-100 series hardware.

the observed count rate (CR) was determined:

$$\ln FL = 0.679 \ln CR - 8.309.$$

This relationship predicts a loss of only 0.77% when source NSS-19 is counted in position "C" (i.e., the normal position); however, 7.6% of the counts would be lost from a 10-mg source counted in that position. This explains most of the bias we have seen between wet radiochemical measurements and assays made by neutron counting for large shipments.

The second matter that we investigated was the effect of moisture in shipping packages. The shipping package containing 39 mg of ^{252}Cf was first assayed as loaded (i.e. with wet resin) and then assayed again after the resin was calcined. The presence of the moisture during the first set of counts depressed the count rate an average of 0.32%, an insignificant amount.

5. SPECIAL PROJECTS

The primary functions of TRU are: (1) to fabricate targets for irradiation in the HFIR to produce transuranium elements, and (2) to isolate and purify transuranium elements for use by research workers. However, the facilities that are available at TRU are also used for a variety of other purposes such as nonroutine productions, special preparations, and special irradiations in HFIR; in each case, a unique service can be provided to assist a research program at ORNL or another site.

5.1 Special Target

Special target ST-6 (K-32), which was discharged from the HFIR on July 1, was cut up at TRU for distribution to various experiments. This target initially contained the following:

- 1 ^{252}Cf pellet ($\sim 10 \mu\text{g}$ of ^{252}Cf)
- 1 ^{242}Pu pellet ($\sim 286 \text{ mg}$ of ^{242}Pu)
- 1 ^{244}Cm pellet ($\sim 286 \text{ mg}$ of ^{244}Cm)
- 2 ^{248}Cm Phoenix ($\sim 150 \mu\text{g}$ of ^{248}Cm) capsules
- 2 silicon pellets
- 2 SPAMS (spacers-flux monitors)
- 16 ^{244}Cm pellets to fill space

The first three pellets were analyzed for heavy-element content to aid in cross-section interpretation. The two Phoenix capsules are being held for future irradiation. This marks the third step in a continuing program to produce long-lived ^{250}Cm .³ The silicon pellets were shipped to H. Diamond (ANL), who will mass-separate the 700-year ^{32}Si . The SPAMS must be allowed to cool for several months before analysis. In the next HFIR target campaign, the 16 curium pellets will be processed to recover the actinides.

5.2 Preparation of Gamma-Ray Sources for Mössbauer Studies

Two additional Mössbauer sources, GS-5 and GS-6, were made for experimenters working at the Transuranium Research Laboratory. The first Mössbauer source, GS-3, was described in the previous report of this series.² In these sources, plutonium oxide was deposited in a very thin layer (only a few mils thick) over a disk which is about 1 in. in diameter. The disk was doubly encapsulated in aluminum, making the overall container dimensions 1-3/8 in. in diameter by 1 in. high. Source GS-5 was loaded with 0.50 g of ^{240}Pu as the oxide, and GS-6 was loaded with 0.50 g of ^{238}Pu as the oxide. Loading and welding of the inner containers were done in-cell as before; the outer container of GS-5 was welded out of cell. In the case of GS-6, however, the outer container was also welded in-cell due to the extreme hazard of the ^{238}Pu . In-cell welding proved to be considerably more difficult than welding in the laboratory, and several attempts were made before a reasonably good weld was obtained. Even then, after decontamination of the capsule, we covered the weld area with a special low-temperature epoxy.

We also prepared three 1-3/8-in.-diam by 1/16-in.-thick absorbers, of ^{238}U , ^{236}U , and ^{234}U , to be used in the same Mössbauer studies. Each of the absorbers was potted in epoxy.

5.3 Plutonium-240 Rabbit

A sample containing about 250 μg of ^{240}Pu was sealed in a quartz ampule by F. Asaro of LBL. We encapsulated this ampule in a HFIR rabbit and irradiated it 5 days in the HFIR rabbit facility. After discharge

and transfer of the rabbit to TRU, we found that its radioactivity level (probably due to ^{140}La radiation) was too high to permit shipment. After a cooling period of about 2 weeks, we finally shipped it in the same type of carrier that we use for ^{253}Es rabbits — a heavy 4-in.-diam lead pig contained inside a 6M container. Asaro is searching for a short-lived (approximately 1/3 year) nuclide which has been previously observed in a plutonium sample. The unknown nuclide was originally thought to be an isomer of ^{241}Pu , but it might be americium or neptunium instead.

6. REFERENCES

1. W. D. Burch, J. E. Bigelow, and L. J. King, Transuranium Processing Plant Semiannual Report of Production, Status, and Plans for Period Ending December 31, 1970, ORNL-4666.
2. W. D. Burch, J. E. Bigelow, and L. J. King, Transuranium Processing Plant Semiannual Report of Production, Status, and Plans for Period Ending June 30, 1971, ORNL-4718.
3. W. D. Burch, J. E. Bigelow, and L. J. King, Transuranium Processing Plant Semiannual Report of Production, Status, and Plans for Period Ending June 30, 1969, ORNL-4447.
4. C. M. Lederer, J. M. Hollander, and I. Perlman, Table of Isotopes, 6th ed., Wiley, New York, 1967.
5. A. H. Wapstra, "Actinide Fingerprints," Actinides Rev. 1, 39-53 (1967).

7. APPENDIX

We have tabulated the decay data and the cross-section data that we use in planning irradiation-processing cycles, calculating production forecasts, and assaying products. The tables will be reproduced completely in each of these semiannual reports, and changes made since the preceding report will be indicated. We wish to state clearly that these data merely represent numbers being used in our calculations and that the data are presented on a "best efforts" basis. Although the information is intended to be definitive, it has not been checked and cross-checked sufficiently to be considered "publishable."

The Transplutonium Element Production Program is now making nuclides available in increasing abundance and purity; therefore, in the next few years, we anticipate a burgeoning literature concerning nuclear constants for the transuranium nuclides. However, since we need such data at the present time, it will not be feasible for us to wait until highly reliable sources, such as Lederer⁴ and Wapstra,⁵ can publish data that have been fully evaluated.

We welcome telephone calls to point out errors or indicate additional sources of information. Please contact John Bigelow, FTS 615-483-1872 or, by commercial telephone service, 615-483-8611, ext. 3-1872.

7.1 Decay Data

Table A-1 is a list of all nuclides of interest to the Transplutonium Element Production Program, (i.e., all that can be produced by neutron bombardment of ^{238}U). The list includes values for half-lives and branching ratios or partial decay half-lives, along with literature references where available. In many cases, the half-life of an isotope was determined by relating that isotope's half-life to the half-life of some other, reference isotope. In a few of these cases, a newer value has been accepted for the half-life of the reference isotope, and the values of the half-lives that were dependent upon it have been recalculated. Such cases

Table A-1. Half-Life Values^a for Isotopes of Transuranium Elements

Underlines indicate new values since the previous report.

Nuclide	Total Half-Life	Partial Half-Life for α Decay	Branching Ratios	Partial Half-Life for Spontaneous Fission	Neutrons per Fission	References ^b
^{237}Np		$(2.14 \pm 0.01) \times 10^6 \text{ y}$		$\sim 10^{18} \text{ y}$	2.00^d	60Br12, 61Br04
^{238}Np	$2.10 \pm 0.01 \text{ d}$					58Fr53
^{239}Np	$2.350 \pm 0.010 \text{ d}$					59Cz03
^{240}Np	$63 \pm 2 \text{ m}$					60Le03
^{240m}Np	$7.3 \pm 0.3 \text{ m}$					48Py61
^{241}Np	16 m					60Le03
^{242}Np	3.4 h					60Le03
^{238}Pu	$87.404 \pm 0.041 \text{ y}$			$(5 \pm 0.6) \times 10^{10} \text{ y}$	2.33 ± 0.00	61Br04, 64Je15, 56Ni02
^{239}Pu		$(2.4413 \pm 0.003) \times 10^4 \text{ y}$		$5.5 \times 10^{15} \text{ y}$	2.24^d	52Se07, 59Ma26
^{240}Pu		$6500 \pm 40 \text{ y}$		$(1.340 \pm 0.015) \times 10^{11} \text{ y}$	2.257 ± 0.046	51In03, 62Ma13, 56Ni02
^{241}Pu	$14.90 \pm 0.33 \text{ y}$	$(5.72 \pm 0.1) \times 10^5 \text{ y}$				60Ca19, 60Br15
^{242}Pu		$(3.860 \pm 0.016) \times 10^5 \text{ y}$		$(7.45 \pm 0.17) \times 10^{10} \text{ y}$	2.18 ± 0.09	63Ma50, 69Ma06, 56Ni01
^{243}Pu	$4.955 \pm 0.003 \text{ h}$					60Br09
^{244}Pu		$(8.28 \pm 0.10) \times 10^7 \text{ y}$		$(6.55 \pm 0.32) \times 10^{10} \text{ y}$	2.84^d	64Fi07, 69Ma06
^{245}Pu	$10.6 \pm 0.4 \text{ h}$					56Br02
^{246}Pu	$10.85 \pm 0.02 \text{ d}$					54Ma23
^{241}Am		$432.7 \pm 0.7 \text{ y}$		$(2.3 \pm 0.8) \times 10^{14} \text{ y}$	2.48^d	61Br04, 67Ma01
^{242}Am	$16.01 \pm 0.02 \text{ h}$		$\text{EC}/\beta = 0.19$			53Ka58
^{243}Am	$144 \pm 7 \text{ y}$	$(2.92 \pm 0.15) \times 10^4 \text{ y}$				59Ma21 ^c
^{243m}Am		$7370 \pm 40 \text{ y}$				60Br22
^{244}Am	$10.1 \pm 0.1 \text{ h}$					62Va08
^{246m}Am	25 m					54Ch24
^{245}Am	$2.07 \pm 0.02 \text{ h}$					56Br02
^{246}Am	$25.9 \pm 0.2 \text{ m}$					55En16
^{248m}Am	$40 \pm 7 \text{ m}$					67Or02
^{247}Am	$24 \pm 3 \text{ m}$					67Or02
^{242}Cm	$162.7 \pm 6.1 \text{ d}$			$7.2 \times 10^6 \text{ y}$	2.65 ± 0.09	51Ma07, 57Pe52, 56Ni01
^{243}Cm		32 y				57Ma00
^{244}Cm	$10.009 \pm 0.015 \text{ y}$		$\alpha/\text{SF} = (7.43 \pm 0.01) \times 10^5$		2.84 ± 0.09	65Ma02, 60Br26, 56Ni01
^{245}Cm		$6265 \pm 100 \text{ y}$				60Ma01
^{246}Cm		$4711 \pm 22 \text{ y}$	$\alpha/\text{SF} = 3822 \pm 10$		3.00^d	60Ma01
^{247}Cm		$(1.64 \pm 0.20) \times 10^7 \text{ y}$				63Fi08
^{248}Cm		$(3.84 \pm 0.04) \times 10^5 \text{ y}$	$\alpha/\text{SF} = 11.0 \pm 3$		3.32^d	69Ma01
^{249}Cm	$64 \pm 3 \text{ m}$					58Ea06
^{250}Cm				$(1.76 \pm 0.24) \times 10^4 \text{ y}$	3.56^d	66Mc01

Table A-1 (continued)

Nuclide	Total Half-Life	Partial Half-Life for α Decay	Branching Ratios	Partial Half-Life for Spontaneous Fission	Neutrons per Fission	References ^b
²⁴⁰ Ba	314 $\pm$ 8 d		$\lambda_\alpha = (1.45 \pm 0.08) \times 10^{-5}$	$(1.07 \pm 0.09) \times 10^9$ y	3.72 $\pm$ 0.16	57En01, 68Mc08, 64Py02
²⁵⁰ Ba	3.222 $\pm$ 0.005 h					59Va02
²⁵¹ Ba	57 $\pm$ 1.7 m					66Mc04
²⁴⁰ Cf		352 $\pm$ 6 y	$\alpha/SF = (1.992 \pm 0.040) \times 10^8$		3.44 ^d	68Mc01, 68Mc08
²⁵⁰ Cf		13.08 $\pm$ 0.09 y	$\alpha/SF = 1260 \pm 40$		3.56 ^d	63Ph01, 68Mc01
²⁵¹ Cf		900 $\pm$ 50 y				68Mc01
²⁵² Cf	2.646 $\pm$ 0.004 y		$\alpha/SF = 31.3 \pm 0.2$		3.796 $\pm$ 0.031	65Mc02, 68Mc04
²⁵³ Cf	17.812 $\pm$ 0.002 d		$\alpha/\beta = (3.1 \pm 0.4) \times 10^{-3}$			68Mc02, 66Mc01
²⁵⁴ Cf	49.5 $\pm$ 0.2 d		$\alpha/SF = (3.10 \pm 0.16) \times 10^{-3}$		3.90 $\pm$ 0.14	63Ph01, 64Py02, 68Mc01
²⁵⁵ Cf	1.5 $\pm$ 0.5 h					70Lo19
²⁵³ Es	20.067 $\pm$ 0.024 d		$\alpha/SF = (1.15 \pm 0.03) \times 10^7$		3.92 ^d	65Mc02, 68Mc02
²⁵⁴ Es	276 d			$> 2.5 \times 10^7$ y	4.04 ^d	67Fi03, 67Mc01
^{254m} Es	30.3 $\pm$ 0.2 h		$\left\{ \begin{array}{l} \alpha/\beta = 382 \pm 30 \\ \text{E.C.}/\beta = 0.00078 \pm 0.00020 \end{array} \right.$			62Mc01, 63Ph01
²⁵⁵ Es	30.8 $\pm$ 1.2 d		$\left\{ \begin{array}{l} \alpha/\beta = 0.0046 \pm 0.0043 \\ \alpha/SF = (2.22 \pm 0.10) \times 10^4 \end{array} \right.$		4.16 ^d	66Mc01, 67Fi03
²⁵⁶ Es	25 $\pm$ 3 m					68Lo11
²⁵⁴ Pu	3.24 $\pm$ 0.01 h		$\alpha/SF = 1695 \pm 8$		4.05 $\pm$ 0.19	56Ja09, 67Fi03, 56Ch03
²⁵⁵ Pu	20.07 $\pm$ 0.07 h		$SF/\alpha = (2.4 \pm 1.1) \times 10^{-7}$		4.16 ^d	63Ph01, 64As01
²⁵⁶ Pu	2.62 $\pm$ 0.03 h		$\sim 100\%$ SF		4.27 ^d	68Mc03
²⁵⁷ Pu	94 $\pm$ 10 d					66Mc01
²⁵⁸ Pu	380 $\pm$ 60 ms		$\sim 100\%$ SF			70Mc17

^aThe half-life values used in this table were being used at TMS at the end of the report period.

^bReferences are decoded in Table A-2.

^cPublished values are adjusted for ²⁴¹Am half-life of 432.7 y.

^dValue estimated by linear interpolation of the values for ²⁴⁴Cm and ²⁵²Cf, based on nucleonic mass.

are footnoted because the half-life value in our table no longer agrees with the value given in the reference. However, we did use the relationship given in the referenced work.

The references used in Table A-1 are decoded in Table A-2. The system of references is that used by the Nuclear Data Project here at ORNL in their widely distributed "Nuclear Data Sheets." Table A-3 lists derived data, such as specific activities, along with information concerning the hazard associated with handling these nuclides.

7.2 Neutron Cross-Section Data

The values of neutron cross sections used to compute transmutations in HFIR target irradiations are listed in Table A-4. This table shows six parameters describing the neutron interactions. The first is the thermal-neutron capture cross section, and the third is the neutron capture resonance integral. The second parameter is a constant that is a function of the target geometry; it is used to estimate the resonance self-shielding effect. The effective capture cross section, $\sigma_{\text{eff}}^{\text{C}}$, would be:

$$\sigma_{\text{eff}}^{\text{C}} = \sigma_{2200}^{\text{C}} + \frac{\phi_{\text{res}}}{\phi_{2200}} \frac{RI}{\sqrt{1 + CN}},$$

where σ_{2200}^{C} is the thermal-neutron capture cross section, N is the number of grams of the particular nuclide in one target rod, ϕ_{res} is the average flux per unit lethargy width in the resonance region, and ϕ_{2200} is the equivalent flux of 2200-m/sec neutrons that would give the same reaction rate with a 1/v absorber as would the actual reactor flux. The effective cross section for fission is computed by a similar relationship among the last three parameters.

These cross sections are to be regarded as a self-consistent set whereby one can compute overall transmutation effects, and as a set of arbitrary constants to be used to obtain the best fit to our data. Hopefully, these numbers and the cross sections experimentally measured on pure isotopes will agree; however, we will not allow the possibility of a discrepancy to confine us.

Table A-2. References for Table A-1

Code	Reference	Code	Reference
40Hy1	E. E. Hyde, M. H. Studier, and W. H. Manning, <i>AML-4143</i> (April 15, 1948) and <i>AML-4182</i> (August 4, 1948).	62He13	D. E. Hutt, F. J. Bonister, J. B. Laidler, and F. Brown, <i>Phys. Rev.</i> , 126 , 264-265 (1962).
50Fr53	M. S. Freedman, A. H. Jaffey, and F. Wagner, Jr., <i>Phys. Rev.</i> , 79 , 410-411 (1950).	63Fr108	P. R. Fields, A. M. Friedman, J. Lerner, D. Metta, and R. Sjöblom, <i>Phys. Rev.</i> , 131 , 1249-1250 (1963).
51He87	G. C. Hanna, B. G. Harvey, H. Moss, and P. R. Tummelcliffe, <i>Phys. Rev.</i> , 81 , 466-467 (1951).	63He50	L. Z. Malkin, I. D. Alkhasov, A. S. Krivokhatskii, and K. A. Petrichak, <i>At. Energy. (USSR)</i> , 15 , 158-159 (1963).
51He83	M. G. Inghram, D. C. Moss, P. R. Fields, and G. L. Pyle, <i>Phys. Rev.</i> , 83 , 1250 (1951).	63Ph81	L. Phillips, R. Gatti, R. Brandt, and S. G. Thompson, <i>J. Inorg. Nucl. Chem.</i> , 25 , 1085-1087 (1963).
52Se67	E. Segre, <i>Phys. Rev.</i> , 86 , 21-28 (1952).	64As81	F. Asaro, S. Björholm, and I. Perlman, <i>Phys. Rev.</i> , 133 , B291-B300 (1964).
53He38	T. E. Kernen, D. A. Fenneman, and D. B. McInteer, <i>J. Chem. Phys.</i> , 21 , 1002-1003 (1953).	64Py82	R. V. Pyle, unpublished results as reported in E. E. Hyde, "Fission Phenomena", Prentice Hall, Inc., (1964).
54Ch24	A. Chiorso, S. G. Thompson, G. R. Choppin, and B. G. Harvey, <i>Phys. Rev.</i> , 94 , 1001 (1954).	65He82	D. Metta, M. Diamond, R. F. Barnes, J. Milsted, J. Gray, Jr., D. J. Henderson, and C. M. Stevens, <i>J. Inorg. Nucl. Chem.</i> , 27 , 33-35 (1965).
55He16	D. Engelkemeir, P. R. Fields, T. Fried, G. L. Pyle, C. M. Stevens, L. B. Asprey, C. I. Bruno, R. Louise Smith, and R. W. Spence, <i>J. Inorg. Nucl. Chem.</i> , 1 , 345-351 (1955).	64Fr87	P. R. Fields, A. M. Friedman, J. Milsted, J. Lerner, C. M. Stevens, D. Metta, and W. K. Sabine, <i>Nature</i> , 212 , 131 (1966).
56He82	J. P. Butler, T. A. Eastwood, T. L. Collins, M. E. Jones, F. N. Bourke, and R. P. Schuman, <i>Phys. Rev.</i> , 103 , 634 (1956).	64RC81	Combined Radiochemistry Group, LBL, LANS, and ARL, <i>Phys. Rev.</i> , 140 , No. 3, 1192-1198 (1966).
56Ch83	G. R. Choppin, B. G. Harvey, D. A. Hicks, J. Iso, Jr., and R. V. Pyle, <i>Phys. Rev.</i> , 102 , 766 (1956).	64RC84	Argonne Heavy Element Group (unpublished data).
56He62	B. C. Diven, M. C. Martin, R. F. Tachak, and J. Terrell, <i>Phys. Rev.</i> , 101 , 1012-1015 (1956).	67Fr83	P. R. Fields, M. Diamond, A. M. Friedman, J. Milsted, J. L. Lerner, R. F. Barnes, R. E. Sjöblom, D. H. Metta, and E. P. Horvitz, <i>Nucl. Phys.</i> , A96 , 440-448 (1967).
56He81	D. A. Hicks, J. Iso, Jr., and R. V. Pyle, <i>Phys. Rev.</i> , 101 , 1016-1020 (1956).	67He81	F. L. Oetting and S. R. Gunn, <i>J. Inorg. Nucl. Chem.</i> , 29 , 2659-2664 (1967).
56He23	B. C. Hoffman and C. I. Bruno, <i>J. Inorg. Nucl. Chem.</i> , 2 , 200 (1956).	67Or82	C. J. Orth, W. R. Daniels, B. W. Erkhila, F. O. Lawrence, and D. C. Hoffman, <i>Phys. Rev. Letters</i> , 19 , No. 3, 128-131 (1967).
56Je80	M. Jones, R. P. Schuman, J. P. Butler, G. Couper, T. A. Eastwood, and M. G. Jackson, <i>Phys. Rev.</i> , 102 , 203-207 (1956).	67Un81	J. Unib, private communication to P. Fields (1967).
57As70	F. Asaro, S. G. Thompson, F. S. Stephens, Jr., and I. Perlman, <i>Bull. Am. Phys. Soc.</i> , 8 , 303 (1957).	68Be21	C. E. Bemis, Jr. and J. Halperin, <i>Nucl. Phys.</i> , A121 , 433-439 (1968).
57E81	T. A. Eastwood, J. P. Butler, M. J. Cabell, M. G. Jackson, R. P. Schuman, F. N. Bourke, and T. L. Collins, <i>Phys. Rev.</i> , 102 , 1635-1638 (1957).	68Be26	M. C. Bentley, <i>J. Inorg. Nucl. Chem.</i> , 30 , 2007-2009 (1968).
57Pe52	R. A. Fenneman, L. M. Treiman, and D. Brown, as reported by D. C. Hoffman, G. P. Ford, and F. O. Lawrence, <i>J. Inorg. Nucl. Chem.</i> , 5 , 6-11 (1957).	68Br22	L. C. Brown and R. C. Probst, <i>J. Inorg. Nucl. Chem.</i> , 30 , 2591-2594 (1968).
58He86	T. A. Eastwood and R. P. Schuman, <i>J. Inorg. Nucl. Chem.</i> , 6 , 261-262 (1958).	68Ca19	M. J. Cabell, <i>J. Inorg. Nucl. Chem.</i> , 30 , 2583-2589 (1968).
58He21	R. F. Barnes, D. J. Henderson, A. L. Markness, and M. Diamond, <i>J. Inorg. Nucl. Chem.</i> , 9 , 105-107 (1958).	68Di80	M. Diamond, J. J. Nines, R. E. Sjöblom, R. F. Barnes, D. H. Metta, J. L. Lerner, and P. R. Fields, <i>J. Inorg. Nucl. Chem.</i> , 30 , 2553-2559 (1968).
58Ca83	D. Cohen, J. C. Sullivan, and A. J. Zielen, <i>J. Inorg. Nucl. Chem.</i> , 11 , 159-161 (1958).	68He13	R. W. Hoff, J. E. Evans, E. K. Mulet, R. J. Dupzyk, and B. J. Qualheim, <i>Nucl. Phys.</i> , A115 , 225-235 (1968).
58He26	T. L. Markin, <i>J. Inorg. Nucl. Chem.</i> , 9 , 320-322 (1958).	68Je15	K. C. Jordan, <i>MLN-1443</i> , 11-30 (1968).
58V82	S. E. Vandenberg, M. Diamond, R. E. Sjöblom, and P. R. Fields, <i>Phys. Rev.</i> , 115 , 115-121 (1958).	68Lo11	R. W. Loughood, private communication to J. E. Bigelow (1968).
60Br12	F. P. Brauer, R. W. Serenett, J. B. Ludrick, F. P. Roberts, and W. L. Lyon, <i>J. Inorg. Nucl. Chem.</i> , 12 , 234-236 (1960).	68Wh84	P. W. White and E. J. Aston, <i>J. Nucl. Energy</i> , 22 , 73-77 (1968).
60Br15	F. Brown, G. G. George, D. E. Green, and D. E. Hutt, <i>J. Inorg. Nucl. Chem.</i> , 13 , 192-195 (1960).	68Wh86	C. E. Bemis, Jr., J. Halperin, and R. Eby, <i>J. Inorg. Nucl. Chem.</i> , 31 , 599-604 (1968).
60Le85	R. H. Loeblor and M. C. Michel, <i>Phys. Rev.</i> , 110 , 263-264 (1960).	68Dr82	R. E. Drushel, J. Halperin, and C. E. Bemis, Jr., <i>ORNL-4437</i> , 28-29 (1968).
61Br84	V. A. Brin, V. P. Porolyskin, and G. I. Shchepilov, <i>Sov. Phys. JETP</i> , 13 , 913-914 (1961).	68He81	D. H. Metta, M. Diamond, and F. R. Kelly, <i>J. Inorg. Nucl. Chem.</i> , 31 , 1245-1250 (1968).
62He81	J. Unib, P. Day, and S. Vandenberg, <i>Nucl. Phys.</i> , 36 , 284-304 (1962).	68He88	J. Milsted, E. P. Horvitz, A. M. Friedman, and D. H. Metta, <i>J. Inorg. Nucl. Chem.</i> , 31 , 1561-1568 (1968).
62V88	S. E. Vandenberg and P. Day, <i>Nucl. Phys.</i> , 36 , 177-190 (1962).	70He17	E. K. Mulet, J. F. Wild, R. W. Loughood, J. E. Evans, B. J. Qualheim, M. Markin, and A. Chiorso, <i>UCRL-72858</i> (1970). To be published in <i>Phys. Rev. Letters</i> .
		70Le19	R. W. Loughood, J. E. Evans, and E. K. Mulet, private communication to J. E. Bigelow (1970).

Table A-3. Properties^a of Transuranium Nuclides
Underlines indicate new values since the previous report.

Nuclide	Half-Life	Energy of Prin. Emission (MeV)		Specific Activity				Hazard ^b		
		α	β	(Ci/g)	(Bq/g)	(μ Ci/mg)	(Bq/mg)	Neutrons $\text{min}^{-1} \text{mg}^{-1}$	$\text{MPC}_d(\text{L})$ ($\mu\text{Ci/cm}^3$)	Body Burden (μCi) (mg)
²³⁷ Pu	2.14×10^6 y	4.78		7.07×10^{-4}	2.07×10^{-5}	8.01×10^5		4.7×10^{-6}	4×10^{-12}	0.06 0.9
²³⁸ Pu	2.10 d		0.25 1.24	2.61×10^5	1.27×10^3		5.80×10^{14}			
²³⁹ Pu	2.359 d		0.332 0.427	2.32×10^5	5.86×10^2		5.14×10^{14}		8×10^{-7}	30 1.29×10^{-4}
²⁴⁰ Pu	63 m		0.89	1.24×10^7	1.03×10^5		2.76×10^{16}			
^{240m} Pu	7.3 m		2.18 1.6	1.07×10^8	5.33×10^5		2.38×10^{17}			
²⁴¹ Pu	16 m			4.86×10^7			1.08×10^{17}			
^{241m} Pu	3.4 h			3.82×10^6			8.49×10^{15}			
²³⁸ Pu	87.404 y	5.49		17.2	0.570	1.94×10^{10}		155	2×10^{-12}	0.04 2.32×10^{-3}
²³⁹ Pu	2.4413×10^4 y	5.15		6.13×10^{-2}	1.913×10^{-3}	6.94×10^7		1.35×10^{-3}	2×10^{-12}	0.04 0.654
²⁴⁰ Pu	6580 y	5.16		0.227	7.097×10^{-3}	2.57×10^8		55.7	2×10^{-12}	0.04 0.176
²⁴¹ Pu	14.98 y	4.9	0.02	99.1	4.06×10^{-3}	2.94×10^6	2.20×10^{11}		9×10^{-11}	0.9 9.08×10^{-3}
²⁴² Pu	3.869×10^5 y	4.90		3.82×10^{-3}	1.13×10^{-4}	4.32×10^6		95.9	2×10^{-12}	0.05 13.0
²⁴³ Pu	4.955 h		0.49 0.58	2.60×10^6	3.34×10^3		5.78×10^{15}		2×10^{-6}	7.2 2.78×10^{-6}
²⁴⁴ Pu	8.28×10^7 y	4.587		1.77×10^{-5}	4.93×10^{-7}	2.00×10^4		141	2×10^{-12}	0.04 2.23×10^3
²⁴⁵ Pu	10.6 h			1.21×10^6			2.68×10^{15}		2×10^{-7}	3.0 4.03×10^5
²⁴⁶ Pu	10.95 d		0.15	4.91×10^4	66.9		1.09×10^{14}			
²⁴¹ Am	432.7 y	5.48		3.43	0.1145	1.08×10^9		3.55×10^{-2}	6×10^{-12}	0.1 0.0292
²⁴² Am	16.01 h		0.63 0.67	8.11×10^5	2.08×10^3		1.80×10^{15d}		4×10^{-6}	0.06 7.39×10^{-8}
^{242m} Am	144 y	5.207	1.7.	10.3	3.08×10^{-2}	5.53×10^7	2.28×10^{10e}		6×10^{-12}	0.07 6.80×10^{-3}
²⁴³ Am	7370 y	5.27		0.200	6.42×10^{-3}	2.24×10^8			6×10^{-12}	0.05 0.25
²⁴⁴ Am	10.1 h		0.387	1.27×10^6	8.74×10^3		2.82×10^{15}		2×10^{-7}	0.18 1.42×10^{-7}
^{244m} Am	26 m		1.5	2.96×10^7	8.98×10^4		6.58×10^{16f}		4×10^{-6}	0.18 6.08×10^{-9}
²⁴⁵ Am	2.07 h		0.91	6.17×10^6	1.20×10^4		1.37×10^{16}		3×10^{-6}	12 1.94×10^{-6}
²⁴⁶ Am	25.0 m		1.31	3.06×10^7	2.48×10^5		6.79×10^{16}			
^{246m} Am	40 m			1.91×10^7			4.24×10^{16}			
²⁴⁷ Am	24 m			3.17×10^7			7.04×10^{16}			
²⁴² Cm	162.7 d	6.11		3.32×10^3	122	3.76×10^{12}		1.21×10^5	1×10^{-10}	0.05 6.27×10^{-7}
²⁴³ Cm	32 y	5.79		45.9	1.677	5.20×10^{10}	3.27×10^8		6×10^{-12}	0.09 1.96×10^{-3}
²⁴⁴ Cm	18.099 y	5.80		80.94	2.832	9.16×10^{10}		6.87×10^5	9×10^{-12}	0.1 1.24×10^{-3}
²⁴⁵ Cm	8265 y	5.36		0.177	5.89×10^{-3}	2.00×10^8			5×10^{-12}	0.04 0.263
²⁴⁶ Cm	4711 y	5.4		0.009	1.01×10^{-2}	3.50×10^8		3.52×10^5	5×10^{-12}	0.05 0.194
²⁴⁷ Cm	1.64×10^7 y			8.83×10^{-5}	2.8×10^{-6}	1.00×10^5			5×10^{-12}	0.04 453
²⁴⁸ Cm	3.52×10^5 y	5.05		4.09×10^{-3}	5.32×10^{-4}	4.25×10^6		2.51×10^6	6×10^{-13}	0.01 2.91
²⁴⁹ Cm	64 m		0.9	1.18×10^7	2.06×10^4		2.62×10^{16}		8×10^{-6}	0.8 6.78×10^{-8}
²⁵⁰ Cm	1.74×10^4 y			8.20×10^{-2}	40.1			6.49×10^8		
²⁴⁹ Bk	314 d	5.4	0.125	1.67×10^3	0.358	2.74×10^7	3.71×10^{12}	6.34×10^3	9×10^{-10}	0.7 4.19×10^{-4}
²⁵⁰ Bk	3.222 h		0.23	3.89×10^6	2.75×10^4		8.62×10^{15}		1×10^{-7}	0.04 1.03×10^{-7}
²⁵¹ Bk	57 m			1.32×10^7			2.92×10^{16}			

Table A-3 (continued)

Nuclide	Half-life	Energies of Principal Emissions (keV)		Specific Activity				Dose	
		α	β	(Ci/g)	(Bq/g)	(μ Ci/mg)	(Bq/mg)	(Neutrons $\text{min}^{-1}\text{mg}^{-1}$)	$\text{MPC}_{\text{a(60)}}^{\text{b}}$ ($\mu\text{Ci/cm}^3$)
^{209}Fr	152 y	5.81		4.00	0.152	4.62×10^9		156	2×10^{-12}
^{210}Fr	13.00 y	6.03		109	4.06	1.25×10^{11}		6.85×10^8	5×10^{-12}
^{251}Fr	900 y			1.59	5.79×10^{-2}	1.78×10^9			2×10^{-12}
^{252}Fr	2.600 y	6.11		530	39.0	5.88×10^{11}		1.40×10^{11}	6×10^{-12}
^{253}Fr	17.012 d	5.96	0.27	2.90×10^8	15.09	1.02×10^{11}	6.41×10^{10}		8×10^{-10}
^{254}Fr	60.5 d	5.84		8.09×10^3	1.08×10^4	2.89×10^{10}		7.35×10^{13}	5×10^{-12}
^{255}Fr	1.5 h			1.8×10^9					
^{253}Ra	20.467 d	6.63		2.52×10^4	1.01×10^3	2.86×10^{13}		1.91×10^7	7×10^{-10}
^{254}Ra	2.76 d	6.42		1.06×10^3	71.9	2.11×10^{12}		45.06×10^5	2×10^{-11}
^{256}Ra	30.3 h		0.48	3.14×10^5	1.18×10^3		6.97×10^{14}		5×10^{-9}
^{255}Ra	30.8 d			1.29×10^4			2.06×10^{13}	4.94×10^9	6×10^{-10}
^{256}Ra	25 h			2.94×10^7			6.52×10^{16}		
^{254}Po	3.24 h	7.20		3.01×10^6	1.60×10^3	4.31×10^{15}		2.02×10^{13}	6×10^{-8}
^{255}Po	20.07 h	7.03		6.15×10^5	2.79×10^4	6.94×10^{16}		1.36×10^3	2×10^{-8}
^{256}Po	2.62 h			4.67×10^6	5.85×10^6			4.43×10^{16}	2×10^{-9}
^{257}Po	94 d			5.41×10^3	~200	6.12×10^{12}			
^{258}Po	300 ms			1.15×10^{11}					

^aThe values for properties included in this table are those in use at TNU at the end of the report period.

^bFrom ICRP Publication 2, "Report of Committee II on Permissible Dose for Internal Radiation (1959)" and the 1962 Supplement.

^cCounting geometry, 516

^d ^{251}Fr decays by β emission (84%) and orbital electron capture (16%).

^e ^{252}Fr decays almost entirely by isomeric transition to the 16-hr ground state, ^{252}Fr .

^f ^{253}Fr decays primarily by β emission, but 0.030% decays by electron capture to ^{253}Ra .

Table A-4. Neutron Cross Sections Used to Compute Transmutations in HFIR Target Irradiations

Underlines indicate new values since the previous report.

Nuclide	Half-Life	Capture			Fission		
		2200-m/s Cross Section (barns)	Resonance Self-Shielding Constant	Resonance Integral (barns)	2200-m/s Cross Section (barns)	Resonance Self-Shielding Constant	Resonance Integral (barns)
²³⁸ Pu	87.494 y	560	0	150	16.5	0	25
²³⁹ Pu	2.4413 x 10 ⁴ y	265.7	0	195	742.4	0	324
²⁴⁰ Pu	6580 y	290	0	8453	0.05	0	0
²⁴¹ Pu	14.98 y	360	0	166	1011	0	541
²⁴² Pu	3.869 x 10 ⁵ y	<u>19.5</u>	<u>6.20</u>	1280	0	0	0
²⁴³ Pu	4.955 h	80	0	0	210	0	0
²⁴⁴ Pu	8.28 x 10 ⁷ y	1.6	0	0	0	0	0
²⁴⁵ Pu	10.6 h	277	0	0	0	0	0
²⁴⁶ Pu	10.85 d	0	0	0	0	0	0
²⁴³ Am	7370 y	<u>105</u>	<u>0</u>	1500	0	0	0
²⁴⁴ Am	10.1 h	0	0	0	2300	0	0
^{244m} Am	26 m	0	0	0	0	0	0
^{244c} Am ^a	49 m	0	0	0	1128	0	0
²⁴⁵ Am	2.07 h	0	0	0	0	0	0
²⁴⁶ Am	25.0 m	0	0	0	0	0	0
²⁴⁴ Cm	18.099 y	<u>10.0</u>	<u>4.0</u>	650	1.2	<u>4.0</u>	12.5
²⁴⁵ Cm	8265 y	<u>343</u>	<u>2.4</u>	<u>120</u>	<u>1727</u>	<u>2.4</u>	1140
²⁴⁶ Cm	4711 y	1.25	<u>0</u>	121	0	0	0
²⁴⁷ Cm	1.64 x 10 ⁷ y	60	0	500	120	0	1060
²⁴⁸ Cm	3.52 x 10 ⁵ y	<u>3.56</u>	<u>2.0</u>	<u>170</u>	0	0	0
²⁴⁹ Cm	64 m	2.8	0	0	50	0	0
²⁵⁰ Cm	1.74 x 10 ⁴ y	2	0	0	0	0	0
²⁴⁹ Bk	314 d	1451	2.4	1240	0	0	0
²⁵⁰ Bk	3.222 h	350	0	0	3000	0	0
²⁵¹ Bk	<u>57 m</u>	<u>0</u>	<u>0</u>	<u>0</u>	<u>0</u>	<u>0</u>	<u>0</u>
²⁴⁹ Cf	352 y	450	1.46	750	1690	5.8	2920
²⁵⁰ Cf	13.08 y	1900	<u>20</u>	<u>11600</u>	0	0	0
²⁵¹ Cf	900 y	<u>2850</u>	14	<u>1600</u>	<u>3750</u>	<u>14</u>	<u>5400</u>
²⁵² Cf	2.646 y	19.8	0	44	<u>32</u>	0	<u>110</u>
²⁵³ Cf	17.812 d	12.6	0	0	1300	0	0
²⁵⁴ Cf	60.5 d	50	0	1650	0	0	0
²⁵⁵ Cf	1.5 h	0	0	0	0	0	0
²⁵³ Es	20.467 d	345	0	0	0	0	0
²⁵⁴ Es	276 d	20	0	0	3060	0	0
^{254m} Es	39.3 h	1.26	0	0	1840	0	0
²⁵⁵ Es	39.8 d	60	0	0	0	0	0
²⁵⁶ Es	25 m	0	0	0	0	0	0
²⁵⁴ Fm	3.24 h	76	0	0	0	0	0
²⁵⁵ Fm	20.07 h	26	0	0	100	0	0
²⁵⁶ Fm	2.62 h	45	0	0	0	0	0
²⁵⁷ Fm	94 d	10	0	0	5500	0	0
²⁵³ Fm	380 μ s	0	0	0	0	0	0

^aTo simplify calculations we use a fictitious isotope, ^{244c}Am, which combines the properties of ^{244m}Am and ²⁴⁴Am according to their relative rates of production from ²⁴³Am.

It should be pointed out that $^{244}\text{C}\text{Am}$ is a fictitious isotope which is used to simplify the calculation of the main transmutation chain involving ^{244}Am . The properties of $^{244}\text{C}\text{Am}$ were calculated from the properties of the real isomers $^{244}\text{g}\text{Am}$ and $^{244}\text{m}\text{Am}$ by assuming that: (1) the number of atoms of $^{244}\text{C}\text{Am}$ present equals the total number of atoms of the real isomers; (2) the β decay from $^{244}\text{C}\text{Am}$ equals the total β decay from the real isomers; (3) the fissions from $^{244}\text{C}\text{Am}$ equal the total fissions from the real isomers; (4) the isomers are in equilibrium with their common parent ^{243}Am while the reactor is operating; and (5) the only significant production and removal factors are the removal of the isomers by decay and neutron absorption, and the production of the isomers by transmutation from ^{243}Am . Thus,

$$(1) \quad N_c = N_g + N_m,$$

$$(2) \quad \lambda_c N_c = \lambda_g N_g + \lambda_m N_m,$$

$$(3) \quad \sigma_c^f N_c = \sigma_g^f N_g + \sigma_m^f N_m,$$

$$(4) \quad \frac{dN_c}{dt} = \frac{dN_g}{dt} = \frac{dN_m}{dt} = 0, \text{ and}$$

$$(5) \quad (\lambda_i + \sigma_i^a \phi) N_i = f_i \sigma_i^c N_{243},$$

where superscripts f, a, and c refer to fission, neutron absorption, and neutron capture; subscript i refers to the i th isomer, c, g, or m; and f_i is the fraction of neutron captures in ^{243}Am resulting in the i th isomer, such that $f_c = f_g + f_m = 1$.