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MASTER

**TRANSURANIUM PROCESSING PLANT
SEMIANNUAL REPORT OF PRODUCTION,
STATUS, AND PLANS
FOR PERIOD ENDING DECEMBER 31, 1969**

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

OAK RIDGE NATIONAL LABORATORY
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for the
U.S. ATOMIC ENERGY COMMISSION

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CHEMICAL TECHNOLOGY DIVISION

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JUNE 1970

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SUMMARY

This is the fourth 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 total amounts of transuranium elements recovered during the period July 1, 1969, to December 31, 1969, are: 3 g of ^{242}Pu , 0.8 g of ^{243}Am , 16.8 g of ^{244}Cm , 12.8 mg of ^{252}Cf , 2 mg of ^{249}Bk , 82 μg of ^{253}Es , and 1.5×10^9 atoms of ^{257}Fm . We made 47 shipments, which totaled 0.29 g of ^{242}Pu , 3.2 g of ^{243}Am , 8.4 g of ^{244}Cm , 1 g of ^{244}Cm that contained about 16% ^{246}Cm , 165 μg of ^{248}Cm (95% ^{248}Cm), 119 μg of ^{248}Cm (isotopically pure) that had been "milked" from californium, 1.8 mg of ^{249}Bk , 2.6 mg of ^{252}Cf in a mixture of californium isotopes (75 to 80% ^{252}Cf), 4 mg of ^{252}Cf that had been irradiated to 97% ^{252}Cf , 36.4 μg of isotopically pure ^{253}Es , 69 μg of ^{253}Es containing some ^{254}Es and ^{255}Es , and 8×10^7 atoms of ^{257}Fm .

Our processing schedule has been changed significantly since the previous report. We now plan to process 163 Savannah River Plant (SRP) reactor slugs during fiscal year 1971 (i.e., July 1970 through June 1971). These slugs, which contain americium and curium, are being irradiated in the "Californium-I" phase of a program at SPP to produce ^{252}Cf for use in the USAEC program to evaluate the commercial market for californium. Our original plans for processing irradiated HFIR targets have been modified somewhat in order to accommodate the slugs. However, this modification will not reduce the availability of transuranium elements for research. The schedule for processing the targets will simply be accelerated both before and after the slugs are processed in order that the recovery of californium from HFIR targets as of January 1972 will remain unchanged. The TRU operations group will be expanded during fiscal year 1971 to handle the increased processing load. Research requirements for californium during the processing of the slugs can be satisfied from the californium that will be recovered from the slugs. The total amount of

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berkelium and einsteinium expected to be recovered from the slugs (about 30 mg of ^{249}Bk and 600 μg of ^{253}Es) will be made available to researchers through the Transplutonium Committee.

We expect to produce the following total amounts of transuranium elements during the next 18 months: 72.5 mg of ^{249}Bk , 492 mg of ^{252}Cf (122 mg from HFIR targets for research, and 370 mg from SRP slugs for the program for evaluating the commercial applications of californium), 1.3 mg of ^{253}Es , and 1.8×10^9 atoms of ^{257}Fm .

The sequence of process steps currently being used at TRU has not been changed since the previous report. For convenience, it is reviewed in this report, and the status of each step is summarized.

At TRU, services are also available by which special materials not normally produced in main-line efforts can be prepared. Four neutron sources containing about 1 mg each of ^{252}Cf , and one neutron source containing about 2 mg of ^{252}Cf , were fabricated during this report period. A gamma-ray source, containing 1 mg of ^{244}Cm and 200 mg of ^{13}C , was also fabricated. This source, which emits a sharply-defined 6130-kev gamma ray, is used for calibrating high-energy gamma-ray detectors.

The values that we use 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 fourth report in a series¹⁻³ 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. A significant change has occurred in our plans since the last report period as the result of the recent decision to process 163 SRP reactor slugs at TRU during fiscal year 1971.

TRU operations during the report period are summarized. Quantities of materials that were produced, processed, and shipped are specified, and proposed processing schedules and anticipated yields of various products are presented. Values of nuclear parameters which were used as input data for the calculations of production rates for transuranium elements, along with a tabulation 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. Special processing, fabrication and irradiation programs are described.

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 usually 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

During July-December 1969, we recovered 3 g of ^{242}Pu , 7.8 g of ^{243}Am , 16.8 g of ^{244}Cm , 2.05 mg of ^{249}Bk , 12.8 mg of ^{252}Cf , 81.5 μg of ^{253}Es , and approximately 1.5×10^8 atoms of ^{257}Fm as purified products from the processing of seven irradiated plutonium targets and approximately one-half of an irradiated curium target. The processing of these targets constituted the only major campaign carried out during this report period.

Forty-seven shipments that were made from TRU (see Table 2.1) during this period included: (1) 0.29 g of ^{242}Pu ; (2) 3.17 g of ^{243}Am and 8.41 g of ^{244}Cm that had been separated and purified; (3) 1.07 g of ^{244}Cm that contained about 16% ^{246}Cm ; (4) 165 μg of ^{248}Cm ($\sim 95\%$ ^{248}Cm); (5) 119 μg of ^{248}Cm (isotopically pure) that had been "milked" from californium; (6) 1.8 mg of ^{249}Bk ; (7) 2.6 mg of ^{252}Cf in a mixture of californium isotopes (75 to 80% ^{252}Cf); (8) 4.04 mg of ^{252}Cf that had been irradiated to 97% ^{252}Cf ; (9) 69 μg of ^{253}Es containing some ^{254}Es and ^{255}Es ; (10) 36.4 μg of ^{253}Es (isotopically pure) that had been "milked" from californium; and (11) 8×10^7 atoms of ^{257}Fm .

As of December 31, 1969, an inventory of the transuranium materials on hand (exclusive of HFIR targets) showed the following: 4.8 g of ^{242}Pu , 114 g of ^{243}Am , 184 g of ^{244}Cm , and 10.2 mg of ^{252}Cf .

2.2 Irradiation and Processing Proposals

The estimated future production of transuranium elements is given in Table 2.2. Two factors have caused our future plans to be changed significantly since the previous semiannual report.³ First, the scientist who required 100 μg of ^{253}Es (mixed einsteinium isotopes) postponed his experiment from February until May 1970. Second, the Transuranium Processing Plant will conduct three campaigns during fiscal year 1971 to process americium-curium slugs that are being irradiated as part of the Californium-I program at the Savannah River Plant (SRP). In this program, californium is being produced for use in the USAEC program to evaluate the commercial market for ^{252}Cf .⁴ The plans for the processing of the SRP slugs will be discussed in Sect. 2.3.

Table 2.1. Distribution of Heavy Elements
from the Transuranium Processing Plant
During the Period July 1, 1969, to December 31, 1969

Major Nuclide	Date	TRU File No.	Shipped To	
			Individual	Site
<u>Plutonium-242, g</u>				
0.29 <u>0.29</u>	12-11-69	239	Prod. Support	ORNL-CTD
<u>Americium-243, g</u>				
2.62	8-06-69	204	L. J. Nugent	ORNL-TRL
0.55	8-06-69	205	Isotope Sales	ORNL
<u>3.17</u>				
<u>Curium-244, g</u>				
1.22	8-06-69	203	L. J. Nugent	ORNL-TRL
0.001	8-26-69	210	Prod. Support	ORNL-CTD
0.001 (Gamma source, GS-1)	11-07-69	229	J. K. Dickens	ORNL- Electro- nuclear
2.0 ^a	12-05-69	172	R. O. Budd	PNL
5.0 ^a	12-09-69	170	P. R. Fields	ANL
0.19 (irradiated)	12-11-69	239	Prod. Support	ORNL-CTD
<u>8.412</u>				
<u>Curium-244 (16% ²⁴⁶Cm content), g</u>				
1.07 <u>1.07</u>	10-15-69	207	Isotope Separation	ORNL
<u>Curium-248 (isotopically enriched to ~95% ²⁴⁸Cm), µg</u>				
120	9-25-69	221	R. J. Silva	ORNL-TRL
45	9-25-69	222	O. L. Keller	ORNL-TRL
<u>165</u>				

Table 2.1 (continued)

Major Nuclide	Date	TRU File	Shipped To	
		No.	Individual	Site
<u>Curium-248</u> (isotopically pure), μg				
25	9-25-69	220	J. R. Peterson	Leige, Belgium
70	9-25-69	217	J. A. Harris	LRL-B
22	9-30-69	218	P. R. Fields	ANL
2	9-30-69	216	D. C. Hoffman	LASL
<u>119</u>				
<u>Berkelium-249, μg</u>				
35	7-11-69	200	R. W. Hoff	LRL-L
65	8-12-69	202	R. W. Hoff	LRL-L
50	10-17-69	173	F. B. Simpson	INC
100	10-17-69	174	R. O. Budd	PNL
250	10-17-69	175	P. R. Fields	ANL
250	10-17-69	176	J. A. Harris	LRL-B
250	10-17-69	177	R. W. Hoff	LRL-L
275	10-17-69	178	R. A. Penneman	LASL
250	10-17-69	179	R. D. Baybarz	ORNL
250	10-17-69	180	C. H. Ice	SRL
<u>1775</u>				
<u>Californium-252, μg</u>				
3	8-26-69	208	W. J. McDowell	ORNL-CTD
0.25	8-26-69	209	Prod. Support	ORNL-CTD
1	9-25-69	223	W. J. McDowell	ORNL-CTD
10	10-27-69	226	Isotopes Sales	ORNL
1	10-27-69	227	W. J. McDowell	ORNL-CTD
483	11-13-69	181	R. J. Sjoblom	ANL
2087	11-21-69	225	A. P. Boulogne	SRL
19	12-05-69	206	R. O. Budd	PNL
<u>10</u>	12-11-69	239	Prod. Support	ORNL-CTD
2614.25				

Table 2.1 (continued)

Major Nuclide	Date	TRU File No.	Shipped To	
			Individual ¹	Site
<u>Californium-252</u> <u>(irradiated to 97% ²⁵²Cf), μg</u>				
325 (NS-4)	7-15-69	147	C. F. Masters	LASL
2180	9-04-69	136	A. R. Boulogne	SRL
937 (NS-5)	10-31-69	146	F. B. Simpson	INC
<u>4042</u>				
<u>Californium-252</u> <u>(irradiated to 99.23% ²⁵²Cf), μg</u>				
60 ^b	6-16-69	135	M. S. Moore	LASL
<u>Einsteinium-253, μg</u>				
2.0	9-29-69	212	R. D. Baybarz	ORNL-CTD
58.5	9-30-69	213	P. R. Fields	ANL
3.5	9-30-69	211	R. A. Penneman	LASL
5.0	10-08-69	219	C. H. Ice	SRL
<u>69.0</u>				
<u>Einsteinium-253</u> <u>(isotopically pure), μg</u>				
15.0	7-16-69	185	R. W. Hoff	LRL-L
4.4	8-07-69	201	R. W. Hoff	LRL-L
16.0	10-23-69	215	J. A. Harris	LRL-B
1	10-23-69	214	C. H. Ice	SRL
<u>36.4</u>				
<u>Fermium-257, atoms</u>				
8×10^7	9-29-69	224	C. E. Bemis	ORNL-TRL
8×10^7				

^aThese shipments were prepared by ORNL Isotopes Sales. Material originated at Savannah River.

^bThis shipment was inadvertently omitted from the previous report.

Table 2.2. Estimated Future Production of Transcurium Elements

Period	Processing Campaign	Products of Campaigns			²⁵² Cf Production ^a		Date Products Available
		²⁴⁹ Bk (mg)	²⁵² Cf (mg)	²⁵³ Es (μg)	Period (mg)	Cumulative (mg)	
Through December 1969						32	
January - June 1970	7 Pu targets	1.5	12	60(12)			February 1970
	10 Pu targets	3	24	132(24)			May 1970
	2 Cm targets	1	6	33(6)	42	74	May 1970
July - December 1970	35 SRP CF-1 slugs	17	110 ^a	200		74	January 1971
January - June 1971	Irradiation of 100 mg Cf ^b	--	--	500			
	64 SRP CF-1 slugs	18	120 ^a	200			March 1971
	64 SRP CF-1 slugs	21	140 ^a	200			May 1971
	4 Pu targets	1	10	55(10)			June 1971
	10 Cm targets	10	70	385(70)	80	154	June 1971
July - December 1971					~140	~300	
1972					~400 ^c	~700 ^c	
1973					~400 ^c	~1100 ^c	
1974					~500 ^c	~1600 ^c	

^aCalifornium produced in the SRP-slug campaigns is not included in production totals.

^bIt is possible that, concurrently with one of the other campaigns planned for this period, 100 mg of californium will be made available for reirradiation to produce 500 μg of ²⁵³Es.

^cEstimates of californium production rates during 1972, 1973, and 1974 assume that additional curium will be available for fabrication of targets.

Seven irradiated plutonium targets will be processed in January 1970. This processing is expected to yield the following: 3.5 g of ^{242}Pu , 1.5 g of ^{243}Am , 19 g of ^{244}Cm (23 g of total curium), 1.5 mg of ^{249}Bk , 12 mg of ^{252}Cf , and 60 μg of ^{253}Es .

Ten irradiated plutonium targets and two irradiated curium targets will be processed in April 1970. The recovery of 4.8 g of ^{242}Pu , 2.1 g of ^{243}Am , 25.4 g of ^{244}Cm (32.6 g of total curium), 4 mg of ^{249}Bk , 30 mg of ^{252}Cf , and 165 μg of ^{253}Es is anticipated.

Ten irradiated curium targets and four irradiated plutonium targets will be processed following the campaigns to process the SRP slugs. The products — 11 mg of ^{249}Bk , 80 mg of ^{252}Cf , and 440 μg of ^{253}Es — will be available late in June or early in July 1971.

2.3 Processing of Savannah River Slugs

The Division of Production of the USAEC has undertaken an evaluation program to estimate the long-term commercial demand for ^{252}Cf . Californium is now being produced at SRP via the irradiation of materials in two forms: (1) ^{242}Pu contained in 86 tubes that are 4 in. in diameter and have active lengths of about 4 ft, and (2) a mixture of americium and curium contained in 163 slugs that are 1 in. in diameter by 6 in. long.

Arrangements have been made for the slugs to be processed at TRU because of the unique services available here and because facilities for processing such materials are presently unavailable at Savannah River. About 370 mg of ^{252}Cf will be recovered primarily for use in the market evaluation studies; however, some of this californium will also be available to satisfy the research requirements of the National Transplutonium Program of the USAEC's Research Division. Approximately 450 g of curium, assaying 28.5% ^{246}Cm , will be recovered. While this curium is intended for future ^{252}Cf production, some of it may be made available to the research program. About 56 mg of ^{249}Bk and 600 μg of ^{253}Es , by-products of the slug campaigns, will be distributed to researchers by the Transplutonium Committee.

The slugs will be processed in three groups: 35, beginning about September 15, 1970; 64, beginning January 1, 1971; and 64, beginning March 1, 1971. The processing of each group of slugs, designated as a campaign, will require 6 weeks.

We expect to recover about 100 g of ^{244}Cm (150 g of total curium) and 200 μg of ^{253}Es in each of the three campaigns. The successive campaigns are expected to yield 110, 120, and 140 mg of ^{252}Cf , and 17, 18, and 21 mg of ^{249}Bk .

2.4 Estimates of the Availability of Transuranium Elements

Plutonium, americium, and curium are usually considered to be intermediate feed materials rather than products. Estimates of the availability of berkelium, californium, einsteinium, and fermium are given below.

2.4.1 Berkelium

About 72.5 mg of ^{249}Bk will become available during the next 18 months: 1.5 mg in February 1970, 4 mg in May 1970, 17 mg in January 1971, 18 mg in March 1971, 21 mg in May 1971, and 11 mg in June 1971.

2.4.2 Californium

We expect to produce about 492 mg of ^{252}Cf contained in a mixture of californium isotopes (about 80% ^{252}Cf) during the next 18 months. About 370 mg will be primarily for use in the USAEC's market evaluation program, and 122 mg (12 mg in February 1970, 30 mg in May 1970, and 80 mg in June 1971) will be available for research purposes.

2.4.3 Einsteinium

We expect to recover about 1.3 mg of ^{253}Es contained in a mixture of einsteinium isotopes during the next 18 months: 60 μg in February 1970; 165 μg in May 1970; 200 μg in each of the SRP slug campaigns in January 1971, March 1971, and May 1971; and 440 μg in June 1971. If more einsteinium is needed, an additional 500 μg of ^{253}Es (in a mixture of isotopes) could be produced by irradiating 100 mg of ^{252}Cf , assuming

that this operation did not interfere with the processing schedule and the needs dictated by the californium market evaluation program. This irradiation could be scheduled so that it would coincide with one of the processing campaigns during the period January-June 1971.

We will be able to recover some "second-growth" einsteinium from the HFIR target campaigns. That is, after the mixture of einsteinium isotopes has been separated from the californium, the latter will be stored to allow ^{253}Es to "grow in" from the decay of ^{253}Cf ; then the "second growth" ^{253}Es will be recovered. About 12 μg and 30 μg of isotopically pure ^{253}Es will be "milked" from the californium that will be isolated in February 1970 and in May 1970 respectively. The campaign in June 1971 should produce 80 μg of isotopically pure ^{253}Es .

No isotopically pure ^{253}Es will be recovered from the SRP slug campaigns because most of the ^{253}Cf will decay during the long cooling time prior to processing.

2.4.4 Fermium

Each batch of californium that is recovered from the processing of HFIR targets will contain about 1.5×10^7 atoms of ^{257}Fm per milligram of ^{252}Cf . Thus, about 1.8×10^8 atoms will be available in February 1970, 4.5×10^8 atoms will be available in May 1970, and 1.2×10^9 atoms will be available in June 1971. Significant quantities should also be recovered from the SRP slugs; however, data that would permit us to predict the yields in the SRP reactor are not currently available.

3. PROCESSES AND EQUIPMENT

Figure 3.1 is a block diagram showing the processing steps that are required for transuranium element production. These steps are: (1) the preparation of a feed solution by dissolving irradiated targets; (2) the recovery of plutonium; (3) the decontamination of the transplutonium elements from fission products; (4) the separation of americium-curium from the transcurium elements; (5) the separation and purification of berkelium, californium, einsteinium, and fermium; (6) the preparation

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Fig. 3.1. Processing Steps Used for Transuranium Element Production.

of americium-curium oxide microspheres; and (7) the fabrication of targets to be irradiated in the HFIR.

The target dissolution and plutonium recovery steps were modified during this report period to permit iodine removal and trapping techniques to be evaluated. The iodine experiments were performed in conjunction with the Liquid Metal Fast Breeder Reactor (LMFBR) Program.

3.1 Target Dissolution

A 3 M NaOH--1.5 M NaNO₃ solution was used to dissolve the aluminum from the seven plutonium targets and about one-half of a curium target processed during this report period. The resulting solution was decanted through a filter, leaving the undissolved particles of actinides in the dissolver. Then, in order to simulate the dissolution step in the LMFBR fuel recovery process, the residue of actinide oxides was dissolved in T-70 (the dissolver), using 12 M HNO₃. (The normal dissolution procedure is to slurry the oxides, transfer them to a tantalum-lined evaporator, and dissolve them in HCl.)

After the oxides were completely dissolved, the acid was diluted to 5 M HNO₃ and an iodine removal experiment was performed. The experiments and results are described in Sect. 3.9.2.

3.2 Plutonium Removal

A batch extraction, which is best described as a combined Cleanex-Pubex process, was used to separate the plutonium from the other actinides and to prepare the solution of transplutonium actinides for feed to the Transex process. In the Pubex process, plutonium is extracted from 5 to 6 M acid (either HCl or HNO₃), using 1 M di(2-ethylhexyl)phosphoric acid (HDEHP) in diethylbenzene (DEB). The transplutonium actinides remain in the aqueous phase. In the Cleanex process, transplutonium elements are purified (from metallic impurities) by extraction from dilute acid, using about 1 M HDEHP in Amco 125-82 diluent; the metallic impurities are left in the aqueous phase, which is discarded.

In the combined Cleanex-Pubex process, the plutonium is extracted from an acid solution that is at least 1 M (to prevent polymerization of the plutonium); then the acid is neutralized (to about 0.03 M) to promote the extraction of the transplutonium elements, and the aqueous raffinate, which still contains the metallic impurities (in this case, the potassium that was added during the iodine experiments), is discarded. The next step is the stripping of the transplutonium elements. Although either HNO_3 or HCl (5 M) can be used as the stripping agent for these elements, HCl is preferred because the subsequent process step (the Tramex process) requires a chloride medium. The stripping of plutonium requires the use of a reductant in an organic-phase modifier, di-tert-butylhydroquinone (DBHQ) in 2-ethylhexanol, to reduce the plutonium in the organic phase; also, HCl must be used instead of HNO_3 since the latter interferes with the reducing power of the DBHQ.

In the campaign, the composite dissolver solution was adjusted to about 10 liters of 1.6 M acid by boiling, addition of water during boiling, and by dilution. The plutonium was extracted into 20 liters of 1 M HDEHP in 95% Amoco 125-82--5% DEB diluent. The DEB was added to enhance the retention of iodine by the solvent. Sodium hydroxide was added to neutralize the acid.

We attempted to adjust the acidity to 0.03 M; however, we were unable to reduce it below 0.06 M even after three moles of NaOH in excess of the amount required (based on the original acidity) had been added. Apparently, some impurity (we suspect potassium) was being extracted and buffering the acid concentration at 0.06 to 0.07 M. As a result of the higher acidity, nearly 2% of the curium remained in the raffinate, which was subsequently stored as rework material for future recovery. The solvent was scrubbed, successively, with 10 liters of 0.03 M HNO_3 and 10 liters of 0.03 M HCl . The transplutonium elements were then stripped using three 10-liter batches of 6 M HCl --0.5 M H_2O_2 . The H_2O_2 was added to enhance the stripping of the berkelium. About 5% of the berkelium had remained with the plutonium in the Pubex processing step in the previous campaign; however, less than 1% was measured in the plutonium product obtained during the campaign made in this report period.

In the final step of the combined Pubex-Cleanex process, a reductant in an organic-phase modifier, 0.2 M DBHQ in 2-ethylhexanol, was added to the solvent to reduce the plutonium; then the plutonium was stripped using 6 M HCl--0.05 M hydroxylamine. (The hydroxylamine is a holding reductant.) Only 71% of the plutonium was stripped after one addition of the organic-phase modifier and three stripping cycles of 10 liters each. Additional stripping cycles (i.e., three more additions of organic-phase modifier, each of which was followed by stripping with 10 liters of 6 M HCl--0.5 M HONH₂) only increased the total plutonium recovery to 78% of the amount that was measured in the feed. No plutonium was detected in the organic waste.

3.3 First-Cycle Solvent Extraction Process (Tramex)

We made two changes in the Tramex process: an equipment modification and a change in process chemistry.

Modification of the feed system successfully resolved the feed line plugging problems we had encountered in nearly every previous Tramex run. In the original feed system, the feed solution was pumped to a pressurized feed pot that overflowed through a jackleg to the feed tank, and the feed was introduced to the columns through an orifice that frequently plugged. In the modified system, the orifice has been replaced with a metering pump. No difficulty was encountered with the feed system during this campaign.

Tramex product is purified using a batch extraction, similar to the extraction step in the Pubex process, to remove zirconium and residues of Tramex process solvent. During the campaign that was made in this report period, we added DBHQ to the product-cleaning solvent to prevent the extraction of berkelium.

3.4 Partitioning of Actinides

The only change that has been made in the LiCl-based anion exchange process is the addition of 0.1 M H₂O₂ to the transcurium-element elutriant. This change was made to ensure the elution of berkelium (see Sect. 3.6).

3.5 Separation of Transcurium Elements

No modifications have been made in the procedures that are used to separate and purify the transcurium elements.

3.6 Berkelium Material Balance

During the campaign to process the irradiated HFIR targets, 97.5% of the berkelium that was measured in the composite dissolver solution was found in the final transcurium product fraction plus the rework material from the LiCl-based anion exchange processing step. In previous campaigns we had been able to account for only 50 to 75% of the berkelium measured in the dissolver solution.

In the recent campaign we took the following precautions to enhance berkelium recovery: (1) H₂O₂ was added to the transplutonium-element strip solutions in the Cleanex-Pubex step to ensure that the berkelium was stripped, (2) DBHQ was added to the solvent that was used to remove zirconium and organic residues from the Tramex product in order to prevent the extraction of berkelium (DBHQ is normally added to the Tramex extractant to prevent the extraction of cerium), and (3) H₂O₂ was added to the transcurium-element elutriant (8 M HCl) in the LiCl-based anion exchange process to ensure that all of the berkelium would be eluted.

3.7 Preparation of Actinide Oxides

Most of the americium-curium that is recovered from irradiated targets is incorporated into recycle targets (in the form of americium-curium oxides in pressed aluminum pellets), which are subsequently irradiated in the HFIR to produce transcurium elements. During this report period, we produced 136 g of oxide (91 g of ^{244}Cm) that was suitable for incorporation into targets; at the same time, we continued our attempts to improve the sol-gel process being used. As in previous runs, the yield of product was low (less than 70%) and it contained oversize material that had to be ground and screened in order to produce the desired particle size distribution.

Oxalate precipitation is used to remove lithium and other impurities from the americium-curium product from the LiCl-based anion exchange process. This purified material is converted to a feed solution that is 0.1 M in HNO_3 and about 0.1 M in actinides. The feed solution is added to a 10 M NH_4OH solution to form a precipitate, which is subsequently washed, using a fluidized-bed technique, and is then concentrated and digested to form a sol. The sol is converted to gel particles by agitation, in the presence of a drying solvent, in a stirred pot. The gel particles are calcined at 1150°C to give oxide particles.

Feed Preparation. - The oxalate precipitation procedure has not been changed since the previous report.

Feed Adjustment. - Nitric acid having a concentration of 4 M or greater is required to dissolve the oxalate precipitate; however, the acid concentration that is required for the sol-forming step is only 0.1 M. Previously, we removed excess acid by precipitating the actinides with NH_4OH and then redissolving them in 0.1 M HNO_3 . This procedure resulted in plugged lines and equipment and losses of the actinides to the filtrate. Now we evaporate the concentrated acid solution to near dryness to remove all except about 0.8 mole of HNO_3 ; then we add 0.7 mole of NH_4OH and dilute with water to give an acidity of about 0.1 M.

Sol Preparation. - The feed is added to 10 M NH_4OH , which is agitated vigorously, in a conical vessel. The resulting precipitate of americium-curium oxides is fluidized and washed continuously with water. The washed precipitate is allowed to settle, the supernate is removed, and the remaining slurry is concentrated and digested to form a sol.

About 10 to 20% of the actinides is lost (to rework) during the washing step, and the washing effluent is usually cloudy. We tried a number of variations in the process in an effort to reduce the washing losses, which were as low as 3% in the development runs. None proved to be effective. In one run we washed with demineralized water that had been purified by double distillation over potassium permanganate. There was no significant reduction in the losses due to washing. Apparently, these losses are not caused by impurities in our demineralized water. Attempts to remove possible harmful impurities from the feed solution (to the sol-forming step) included: (1) purification of the oxalate precipitate by calcination at 650°C to remove any volatile impurities, followed by dissolution in nitric acid, and (2) purification of this solution by cation exchange (in addition to, or instead of, oxalate precipitation) using Dowex 50 resin. The following two process variations were evaluated: (1) washing with 2 M NH_4OH prior to the water wash in an attempt to remove some impurity that might be more soluble in caustic than in water, and (2) adding the wash water in eight 3-liter volumes, followed by stirring, settling, and decanting, instead of using the fluidized-bed washing technique.

Formation of Microspheres. - A stainless steel baffle was placed in the apex of the inverted-cone vessel, and the agitator containing three blades of different diameters was replaced by a single 3-in.-diam, pitched-blade impeller. These modifications have improved our ability to lift solids out of the bottom (apex area) of the conical vessel. An adjustable dip probe, which is used as a baffle at the top of the liquid, prevents excessive swirl.

The use of a different drying solvent and the stepwise drying of the sol to form gel particles have reduced the tendency of particles to

stick to the wall of the sphere-forming vessel. However, these modifications have not resulted in a decrease in the fraction of particles that are oversize ($>210 \mu$). To date, the fraction of such particles in our oxide products has averaged about 65%. This material must, of course, be ground to give acceptable product.

The drying solvent that we now use is isoamyl alcohol containing the following additives: 0.6 vol % Span 80, 0.6 vol % Amine O, 1 vol % additional water (isoamyl alcohol, as received, contains approximately 0.2% water), and enough HNO_3 to give a final concentration of 0.002 M. This drying solvent reduces the tendency of particles to stick to the walls of the conical vessel. This vessel is not sufficiently large to permit the required amount of solvent to be added at one time; therefore, two to four equal-volume portions of it are added sequentially. The solvent is agitated, allowed to settle, and then decanted between additions.

3.8 Fabrication of Targets

Eleven HFIR targets - one plutonium target and 10 americium-curium targets - were fabricated during this report period. The plutonium target contained 9.33 g of plutonium (9.2 g of ^{242}Pu), and each of the americium-curium targets contained 10 g of actinides (from 72 to 92% ^{244}Cm) in the form of actinide oxide--aluminum pellets that were pressed to 80% of the theoretical density of the pellet core.

3.9 Iodine Studies

We reported previously³ that we had successfully demonstrated the processing of short-cooled targets (to maximize the recovery of ^{253}Es) without releasing excessive amounts of ^{131}I to the environment. For that demonstration, we installed an experimental iodine removal system in a 3-cfm off-gas stream from the condensate collection system serving the dissolver and the seven process evaporators. The system consisted of a caustic scrubber (to remove acid vapors), a heater, a bed of

Hopcalite* (to convert organic materials to CO_2 and H_2O , thus protecting the charcoal trap from organic substances), a cooler, and a charcoal trap.

A larger system to handle the total vessel and cubicle off-gas system (450 cfm) is now being fabricated; installation is planned for July 1970.

3.9.1 Experiments for the LMFBR Program

During the current report period we performed some iodine experiments in conjunction with the LMFBR program. By making only minor changes in our dissolution and plutonium removal steps (i.e., to dissolve in a HNO_3 medium instead of HCl), we were able to closely approximate the proposed LMFBR dissolution process. A special equipment rack was installed in the dissolver off-gas line in cubicle 7, which contained equipment to study iodine-removal techniques that are pertinent to LMFBR fuel processing.

Two aspects of iodine handling in LMFBR head-end processing were studied: (1) the evolution of iodine from the nitric acid dissolver solution, and (2) the removal of iodine from the dissolver off-gas. The experiments and results have been described in detail in an LMFBR progress report.⁵

In three tests, 97 to 98.5% of the iodine was volatilized from nitric acid dissolver solutions by air sparging at elevated temperatures for 8 to 10 hr while adding potassium nitrite continuously and potassium iodide intermittently.

Iodine-removal equipment in the dissolver off-gas stream consisted of two liquid scrubbers in series, followed by a Hopcalite bed--charcoal bed system. The iodine DF for this system varied from 4×10^6 to

*Hopcalite is an oxidation catalyst consisting of a CuO-MnO_2 mixture that has been treated to give a large surface area. It is a proprietary material of the Mine Safety Appliance Company.

1×10^5 in three tests. The lowest DF, which was obtained in the third test, was believed to be due to moisture in the charcoal bed.

3.9.2 Removal of Iodine from Off-Gas in TRU

In our most recent campaign at TRU, about 3.3 Ci of ^{131}I was collected in the effluent from the caustic scrubber in cell 3. In previous campaigns, in which the actinides had been dissolved using HCl, very little iodine had been collected in the scrubber. This demonstrates clearly that the chemical form of the iodine in the off-gas is dependent on the medium in which it was generated.

During the recent campaign, we were unable to evaluate the performance of the Hopcalite bed--charcoal bed portion of the experimental system (3 cfm) that was installed in cell 3 because we could not sample the condensate-collection system off-gas stream between the scrubber and the Hopcalite bed. Also, we were unable to interpret the data that were collected by the radiation monitor on the charcoal bed. Of course, very little iodine was introduced to the Hopcalite-charcoal part of the system in cell 3, since it was being removed by the LMFBR scrubbers and the Hopcalite-charcoal system on the dissolver off-gas, as well as by the cell 3 caustic scrubber.

Inspection of the cell 3 system after the campaign had been completed (i.e., to locate and repair leaks that were causing a loss of vacuum in the evaporator off-gas system) revealed that the charcoal bed outlet was black. Subsequent examination showed that there was no charcoal in the bed. Although it was obvious that the charcoal bed had burned, we could not determine when the burning had occurred. Recorder charts for a thermocouple located in the gas stream on the inlet side of the bed did not register a temperature rise that could be interpreted as the result of a fire in the charcoal bed.

It is obvious that no iodine was collected on the charcoal during the campaign. If the charcoal had been intact at the start of the campaign and had sorbed iodine and then burned, there would have been a sharp increase in the rate of iodine release. No such release was noted either on the gamma spectrometer in the TRU vessel off-gas filter plenum or in the daily samples from the HFIR stack.

The iodine removal system in cell 3 was found to contain numerous leaks caused by corrosion of the stainless steel equipment. Evidently, the HCl vapors had not been completely removed by the caustic scrubber.

The entire system has now been removed and discarded. We do not believe that its replacement will be necessary. Prior to installation of a large iodine removal unit in the 450-cfm vessel off-gas system, we can dissolve targets in HNO_3 , evolve iodine into a dissolver off-gas treatment system (as we did in the current campaign), and collect the iodine that is evolved from other process steps in the large permanent caustic scrubber located in cell 7.

4. SPECIAL PROJECTS

Projects that are being carried out by various groups at ORNL and at other sites require the services available at TRU for the preparation of special materials which are not normally produced in main-line efforts. The phases of these projects involving activities at TRU are reported here; the end results of the research are reported elsewhere.

4.1 Fabrication of Neutron Sources Using Californium

Five neutron sources, in which ^{252}Cf was contained in a pressed aluminum pellet that was encapsulated in stainless steel, were fabricated during this report period. The sources, denoted as NS-4 through NS-8, contained 925 μg , 992 μg , 816 μg , 826 μg , and 1.99 mg of ^{252}Cf respectively. All of the sources except NS-5 were encapsulated in type 304L stainless steel. NS-5 was encapsulated in low-nickel stainless steel (type 405) because nickel would interfere with some of the experiments for which the source will be used. These experiments involve the use of an iron "filter." The source will be surrounded by a massive block of iron, and the neutrons will be scattered into an energy region where iron has a very low cross section. Unfortunately, nickel has a very strong resonance absorption in this energy region; thus an ordinary stainless steel capsule would provide a significant sink for neutrons of this energy and would cause an undesirable flux depression.

4.2 Preparation of a High-Energy Gamma-Ray Source

An interesting new application of transuranium elements is a high-energy gamma-ray source (GS-1), which was fabricated in a manner similar to that of our neutron sources. The gamma radiation of interest (a monoenergetic 6130-keV gamma ray) arises from the reaction $^{13}\text{C}(\alpha, n)^{16}\text{O}$. The alpha particles are supplied by ^{244}Cm ($\alpha_0 = 5.802$ MeV). This particular reaction is valuable because the 6130-keV gamma ray is produced from an excited state of ^{16}O ; the excited state has a half-life of 17 picoseconds, which is sufficiently long to permit the recoil ^{16}O nucleus to come to rest before emitting the photon. In this way, the 6130-keV line is not broadened by the Doppler effect due to the motion of the nucleus.

The source was prepared by adding an alcohol solution of curium nitrate, containing 1 mg of ^{244}Cm , to 200 mg of carbon highly enriched (i.e., 92%) in ^{13}C and subsequently converting the curium nitrate to curium oxide by heating the mixture in a stream of argon. The curium-impregnated carbon was then placed in an aluminum casing, and the resulting pellet was pressed to a minimum size. The pellet was encapsulated in a type 304L stainless steel can and welded shut to ensure complete containment.

The source strength of the 6130-keV gamma ray is about 3700 photons per second. Undesired radiation arises from the spontaneous fission of the ^{244}Cm and from the decay of a few radioactive impurities that are present; however, this amounts to only about 5 mrem/hr at a distance of 2.5 cm from the source and does not interfere with the use of the source. The source is especially useful for calibrating high-quality Ge(Li) detector systems.

4.3 Irradiation of ^{252}Cf to Produce ^{253}Es

About 19.4 μg of isotopically pure ^{253}Es for use in an underground nuclear test, "Physics-8," was produced by irradiating and subsequently processing four ^{252}Cf rabbits that originally contained 6.01 mg of

^{252}Cf . Although the production method is the one normally used, this project was specially designed to meet the schedule of the customer's experiment.

The californium had been irradiated in the HFIR hydraulic rabbit facility and had been processed (during the previous report period) to remove the mixture of einsteinium isotopes produced by the irradiation. About 7.6 μg of ^{253}Es (in the mixture of isotopes) was distributed, and the purified californium was stored about one month to allow the ^{253}Cf (about 1% of the total californium) to decay to ^{253}Es . About 15 μg of pure ^{253}Es was recovered by "milking" the californium. When the Physics-8 shot was delayed, it became necessary to "milk" the californium a second time in order to obtain more ^{253}Es to replace the amount lost by decay during the delay. About 4.4 μg of ^{253}Es was obtained from the second milking.

Other samples provided by TRU for "Physics 8" were: (1) 60 μg of ^{252}Cf (99.23% ^{252}Cf), (2) 41 μg of ^{245}Cm (~75% ^{245}Cm), (3) 21 μg of ^{247}Cm (~26% ^{247}Cm), and (4) 77 μg of ^{248}Cm (~90% ^{248}Cm).

5. 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 June 30, 1968, ORNL-4376.
2. 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, 1968, ORNL-4428.
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. F. P. Baranowski, "Development and Production of Californium-252," pp. 201-11 in Californium-252, CONF-681032 (1969).
5. W. E. Unger et al., LMFBR Fuel Cycle Studies Progress Report for September 1969, No. 7, ORNL-TM-2748 (November 1969).
6. C. M. Lederer, J. M. Hollander, and I. Perlman, Table of Isotopes, 6th ed., Wiley, New York, 1967.
7. A. H. Wapstra, "Actinide Fingerprints," Actinides Rev. 1, 39-53 (1967).

6. 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 Lederer⁶ and Wapstra⁷, 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.

6.1 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

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

Underlines indicate new values since the previous report.

Isotope	Total half-life	Partial half-life for α decay	Branching Ratios	Partial half-life for Spontaneous Fission	Neutrons per Fission	References ^b
^{237}Pu		$(2.14 \pm 0.01) \times 10^8 \text{ y}$		$>10^{16} \text{ y}$	<u>2.00^d</u>	60Br12, 61Dr4
^{238}Pu	$2.80 \pm 0.01 \text{ d}$					50F53
^{239}Pu	$2.350 \pm 0.010 \text{ d}$					59C93
^{240}Pu	$63 \pm 2 \text{ m}$					60Le3
^{240m}Pu	$7.3 \pm 0.3 \text{ m}$					48Ho1
^{241}Pu	16 m					60Le3
^{241m}Pu	3.4 h					60Le3
^{238}Pu	$87.404 \pm 0.041 \text{ y}$			$(5 \pm 0.6) \times 10^{10} \text{ y}$	<u>2.33 $\pm$ 0.08</u>	61Dr4, 68Je15, 54h101
^{239}Pu		$(2.4413 \pm 0.003) \times 10^4 \text{ y}$		$5.5 \times 10^{15} \text{ y}$	<u>2.24^d</u>	52Se7, 55u126
^{240}Pu		$6500 \pm 40 \text{ y}$		$(1.340 \pm 0.015) \times 10^{11} \text{ y}$	<u>2.257 $\pm$ 0.046</u>	51j3, 67Ne13, 54h102
^{241}Pu	$14.90 \pm 0.35 \text{ y}$	<u>$(5.72 \pm 0.1) \times 10^5 \text{ y}$</u>				68Ca19, 60Br15
^{242}Pu		$(3.869 \pm 0.010) \times 10^5 \text{ y}$		$(7.45 \pm 0.17) \times 10^{10} \text{ y}$	<u>2.18 $\pm$ 0.09</u>	68Ne50, 69Ne06, 54h101
^{243}Pu	$4.955 \pm 0.003 \text{ h}$					62u109
^{244}Pu		$(8.28 \pm 0.10) \times 10^7 \text{ y}$		$(6.55 \pm 0.32) \times 10^{10} \text{ y}$	<u>2.84^d</u>	64Fi07, 69Ne06
^{245}Pu	$10.6 \pm 0.4 \text{ h}$					56h92
^{246}Pu	$10.75 \pm 0.02 \text{ d}$					56h23
^{241}Am		$432.7 \pm 0.7 \text{ y}$		$(2.3 \pm 0.8) \times 10^{14} \text{ y}$	<u>2.48^d</u>	61Dr4, 67Ne01
^{242}Am	$16.01 \pm 0.02 \text{ h}$		<u>EC/B = 0.19</u>			53E38
^{242m}Am	$144 \pm 7 \text{ y}$	<u>$(2.92 \pm 0.15) \times 10^4 \text{ y}$</u>				52S221 ^c
^{243}Am		$7340 \pm 50 \text{ y}$				
^{244}Am	$10.1 \pm 0.1 \text{ h}$					62Ve0
^{244m}Am	26 m					54G24
^{245}Am	$2.07 \pm 0.02 \text{ h}$					56h92
^{246}Am	$25.0 \pm 0.2 \text{ m}$					55E16
^{246m}Am	$40 \pm 7 \text{ m}$					67Or02
^{247}Am	$24 \pm 3 \text{ m}$					67Or02
^{242}Cm	$162.7 \pm 0.1 \text{ d}$			$7.2 \times 10^6 \text{ y}$	<u>2.65 $\pm$ 0.09</u>	51h07, 57PS2, 54h101
^{243}Cm		32 y				57A70
^{244}Cm	$18.000 \pm 0.015 \text{ y}$		<u>$\alpha/\text{SF} = (7.43 \pm 0.01) \times 10^5$</u>		<u>2.84 $\pm$ 0.09</u>	62Ne02, 68Ne26, 54h101
^{245}Cm		$8265 \pm 100 \text{ y}$				62Ne01
^{246}Cm		$4711 \pm 22 \text{ y}$	<u>$\alpha/\text{SF} = 3822 \pm 10$</u>		<u>3.08^d</u>	68Ne01
^{247}Cm		$(1.64 \pm 0.20) \times 10^7 \text{ y}$				63Fi08
^{248}Cm		<u>$(3.14 \pm 0.04) \times 10^5 \text{ y}$</u>	<u>$\alpha/\text{SF} = 11.0 \pm 3$</u>		<u>3.32^d</u>	68Ne01
^{249}Cm	$64 \pm 3 \text{ m}$					58E06
^{250}Cm				$(1.74 \pm 0.24) \times 10^4 \text{ y}$	<u>3.56^d</u>	64BC01

Table A-1 (continued)

Isotope	Total half-life	Partial half-life for α decay	Branching fraction	Partial half-life for spontaneous fission	Neutrons per fission	References ^b
^{238}Pu	134 ± 6 d		$\alpha/\lambda = (1.45 \pm 0.05) \times 10^{-5}$	$(1.67 \pm 0.05) \times 10^3$ y	3.72 ± 0.10	57101, 68100, 68702
^{239}Pu	3.523 ± 0.006 a					58902
^{240}Pu	17 ± 1.7 a					68004
^{240}Cf		26.2 ± 0.7 y	$\alpha/\lambda = (1.1 \pm 0.2) \times 10^{-5}$		3.25^d	68101, 68102
^{242}Cf		15.00 ± 0.05 y	$\alpha/\lambda = 1.300 \pm 0.01$		3.50^d	63701, 68101
^{244}Cf		900 ± 50 y				68101
^{244}Cf	1.040 ± 0.004 y		$\alpha/\lambda = 34.5 \pm 0.2$		3.700 ± 0.03	68102, 68104
^{245}Cf	17.042 ± 0.002 d					68102
^{246}Cf	60.5 ± 0.2 d			60.5 ± 0.2 d	3.00 ± 0.10	63701, 68702
^{248}Pu	20.007 ± 0.004 d		$\alpha/\lambda = (1.15 \pm 0.05) \times 10^{-7}$		3.02^d	68101, 68102
^{248}Pu	271 d			2.5 ± 10^4 y	2.04^d	67103, 67105
^{249}Pu	50.5 ± 0.2 d		$\alpha/\lambda = 254 \pm 20$ $\lambda_{\text{C.F.}} = 0.00070 \pm 0.00010$			63101, 63701
^{249}Pu	20.3 ± 1.2 d		$\alpha/\lambda = 0.0000 \pm 0.0003$ $\alpha/\lambda = (12.22 \pm 0.05) \times 10^6$		2.46^d	68101, 67103
^{250}Pu	25 ± 3 d					68101
^{250}Pu	1.21 ± 0.04 d		$\alpha/\lambda = 3000 \pm 0$		2.05 ± 0.10	56100, 67103, 68103
^{251}Pu	20.47 ± 0.07 d		$\alpha/\lambda = (2.0 \pm 1.1) \times 10^{-7}$		2.50^d	63701, 68101
^{252}Pu	1.61 ± 0.05 d			2.62 ± 0.05 d	2.77^d	68103
^{253}Pu	94 ± 10 d					68101

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

^bReferences are listed in Table A-2.

^cSpontaneous values are reported for ^{240}Pu half-life of 433.7 y.

^dValues estimated by linear interpolation of the values for ^{240}Pu and ^{242}Cf , based on systematic error.

upon it have been recalculated. Such cases 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.

6.2 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

Table A-2. References for Table A-1

Code	Reference	Code	Reference
48001	E. K. Hyde, M. H. Studier, and W. M. Sherring, <i>AML-4743</i> (April 15, 1948) and <i>AML-4182</i> (August 4, 1948).	62001	J. Unik, P. Day, and S. Vandenbosch, <i>Nucl. Phys.</i> , 36 , 284-304 (1962).
50F53	M. S. Freedman, A. H. Jaffey, and F. Wagner, Jr., <i>Phys. Rev.</i> , 79 , 410-411 (1950).	62V08	S. E. Vandenbosch and P. Day, <i>Nucl. Phys.</i> , 30 , 177-190 (1962).
51007	G. C. Hanna, B. G. Harvey, M. Moss, and P. R. Turniciff, <i>Phys. Rev.</i> , 81 , 466-467 (1951).	62W13	D. E. Matt, F. J. Zimmister, J. B. Laidler, and F. Brout, <i>Phys. Rev.</i> , 126 , 264-265 (1962).
5113	M. G. Inghram, P. C. Hess, P. R. Fields, and G. L. Pyle, <i>Phys. Rev.</i> , 83 , 1250 (1951).	63F108	P. R. Fields, A. M. Friedman, J. Lerner, D. Metta, and R. Sjöblom, <i>Phys. Rev.</i> , 131 , 1249-1250 (1963).
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55E16	D. Engelkemeir, P. R. Fields, T. Froel, G. L. Pyle, C. M. Stevens, L. B. Asprey, C. I. Brown, M. Louise Smith, and R. W. Spruce, <i>J. Inorg. Nucl. Chem.</i> , 1 , 345-351 (1955).	64P02	R. V. Pyle, Unpublished results, as reported in E. K. Hyde, "Fission Phenomena", Prentice Hall, Inc., (1964).
56002	J. P. Butler, T. A. Eastwood, T. L. Collins, M. E. Jones, F. M. Bourke, and R. P. Schuman, <i>Phys. Rev.</i> , 103 , 634 (1956).	65M02	D. Metta, H. Diamond, R. F. Barnes, J. L. Lerner, J. Gray, Jr., D. J. Henderson, and C. M. Stevens, <i>J. Inorg. Nucl. Chem.</i> , 27 , 33-35 (1964).
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56062	B. C. Biven, M. C. Martin, R. F. Taschek, and T. Terrell, <i>Phys. Rev.</i> , 101 , 1012-1015 (1956).	66R01	Combined Radiochemistry Group, LRL, LAGL, and AML, <i>Phys. Rev.</i> , 140 , No. 3, 1182-1196 (1966).
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56023	D. C. Hoffman and C. I. Brown, <i>J. Inorg. Nucl. Chem.</i> , 2 , 209 (1956).	67F103	P. R. Fields, H. Diamond, A. M. Friedman, J. Milsted, J. L. Lerner, R. F. Barnes, R. K. Sjöblom, D. M. Metta, and E. P. Horvitz, <i>Nucl. Phys.</i> , A96 , 440-448 (1967).
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57E01	T. A. Eastwood, J. P. Butler, M. J. Cabell, M. G. Jackson, R. P. Schuman, F. M. Bourke, and T. L. Collins, <i>Phys. Rev.</i> , 107 , 1635-1638 (1957).	67U01	J. Unik, private communication to P. Fields (1967).
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59V02	S. E. Vandenbosch, H. Diamond, R. K. Sjöblom, and P. R. Fields, <i>Phys. Rev.</i> , 115 , 115-121 (1959).	68L011	R. W. Loughood, private communication to J. E. Bigelow (1968).
60F12	F. P. Brout, R. N. Strom, J. D. Ludwig, F. P. Roberts, and M. L. Lyon, <i>J. Inorg. Nucl. Chem.</i> , 12 , 234-235 (1960).	68M04	P. H. White and E. J. Aron, <i>J. Nucl. Energy</i> , 22 , 73-77 (1968).
60L03	R. M. Lessler and M. C. Michel, <i>Phys. Rev.</i> , 118 , 263-264 (1960).	68O06	C. E. Emsis, Jr., J. Halperin, and R. Eby, <i>J. Inorg. Nucl. Chem.</i> , 31 , 599-604 (1968).
61D04	V. A. Drin, V. P. Pereygin, and G. I. Khlebnikov, <i>Sov. Phys. JETP</i> , 13 , 913-914 (1961).	69O02	R. E. Drushel, J. Halperin, and C. E. Emsis, Jr., <i>ORNL-4437</i> , 28-29 (1969).
		69M01	D. M. Metta, H. Diamond, and F. R. Kelly, <i>J. Inorg. Nucl. Chem.</i> , 31 , 1245-1250 (1969).
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Table A-3. Properties^a of Transactinium Nuclides

Underlines indicate new values since the previous report.

Nuclide	Half-Life	Energy of Prim. Emissions (MeV)		Specific Activity				Reactivity ^b		
		α	β	(Ci/g)	(μ /g)	(μ cps/mg ^c)	(μ cps/mg)	(Sources μ ls ⁻¹ mg ⁻¹)	$\frac{R}{\alpha(\text{Ci})}$ (μ Cl/cm ³)	$\frac{R}{\beta(\text{Ci})}$ (μ Cl)
²³⁷ Pu	2.14×10^6 y	4.79		7.07×10^{-4}	2.07×10^{-5}	8.01×10^5		7×10^{-6}	4×10^{-12}	0.76
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	0.132 0.427		2.32×10^5	5.86×10^2		5.14×10^{14}		8×10^{-7}	30
²⁴⁰ Pu	63 m	0.89		1.24×10^7	1.03×10^5		2.76×10^{16}			
^{240m} Pu	7.3 m	2.12 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}			
^{242m} Pu	3.4 h			3.82×10^6			8.49×10^{15}			
²³⁸ Am	87.604 y	5.49		17.2	0.570	1.94×10^{10}		155	2×10^{-12}	0.04
²³⁹ Am	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
²⁴⁰ Am	6580 y	5.16		0.227	7.897×10^{-3}	2.57×10^8		55.7	2×10^{-12}	5.04
²⁴¹ Am	4.32×10^4 y	4.9	0.02	19.1	4.08×10^{-2}	2.94×10^8	2.20×10^{12}		9×10^{-11}	0.5
²⁴² Am	3.869×10^5 y	4.90		1.82×10^{-3}	1.13×10^{-4}	4.32×10^6		95.9	2×10^{-12}	0.05
²⁴³ Am	4.555 h	0.49 0.58		2.60×10^6	3.34×10^3		5.78×10^{15}		2×10^{-6}	7.2
²⁴⁴ Am	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
²⁴⁵ Am	10.6 h			1.21×10^6			7.68×10^{15}		2×10^{-7}	3.0
²⁴⁶ Am	10.05 d	0.15		4.91×10^4	66.9		1.09×10^{16}			
²⁴¹ Am	432.7 y	5.48		3.43	0.1145	3.08×10^9		1.55×10^{-4}	4×10^{-12}	0.1
²⁴² Am	16.01 h	0.62 0.67		8.11×10^5	2.08×10^3		1.80×10^{15}		5×10^{-8}	3.06
^{243m} Am	164 y	5.207	1.7.	10.3	3.08×10^{-2}	5.53×10^7	2.28×10^{10}		4×10^{-12}	0.07
²⁴³ Am	7340 y	5.27		0.300	6.45×10^{-3}	2.27×10^8			4×10^{-12}	0.05
²⁴⁴ Am	10.1 h	0.387		1.27×10^6	8.74×10^3		2.82×10^{15}		2×10^{-7}	0.18
^{244m} Am	26 m	1.5		1.97×10^7	8.58×10^4		6.58×10^{16}		4×10^{-6}	0.18
²⁴⁵ Am	2.07 h	0.91		6.17×10^6	1.70×10^4		1.37×10^{16}		1×10^{-6}	12
²⁴⁶ Am	25.0 m	1.31		3.74×10^7	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}			
²⁴¹ Cm	162.7 d	6.11		3.32×10^3	122	3.76×10^{12}		1.21×10^4	1×10^{-16}	0.05
²⁴³ Cm	32 y	5.79		45.9	1.677	5.20×10^{10}	3.27×10^8		6×10^{-12}	0.09
²⁴⁴ Cm	18.099 y	5.80		80.54	2.832	9.16×10^{10}		6.87×10^5	9×10^{-12}	0.1
²⁴⁵ Cm	8365 y	5.36		0.177	5.89×10^{-3}	2.00×10^8			5×10^{-12}	0.04
²⁴⁶ Cm	4711 y	5.4		0.177	1.51×10^{-2}	1.50×10^8		3.74×10^5	1×10^{-12}	0.05
²⁴⁷ Cm	1.64×10^7 y			8.83×10^{-5}	2.8×10^{-6}	1.00×10^5			5×10^{-12}	0.04
²⁴⁸ Cm	3.52×10^5 y	5.05		4.09×10^{-3}	5.32×10^{-4}	4.25×10^6		2.34×10^6	6×10^{-13}	0.01
²⁴⁹ Cm	64 m	0.9		1.18×10^7	2.06×10^5		7.42×10^{16}		8×10^{-6}	0.8
²⁵⁰ Cm	1.74×10^4 y			8.20×10^{-2}	4.1			6.49×10^8		
²⁴⁹ Bk	314 d	5.4	0.125	1.67×10^3	0.358	4.70×10^7	3.71×10^{12}	8.34×10^3	9×10^{-10}	0.7
²⁵⁰ Bk	3.272 h	0.23		3.89×10^6	2.75×10^4		8.63×10^{15}		1×10^{-7}	0.04
²⁵¹ Bk	57 m			1.32×10^7			7.89×10^{16}			

Table A-3 (continued)

Sample	Date-1490	Sample of Pyr. Substrate (mg)		Specific Activity				Results		
		a	b	(C/L/g)	(cp/g)	(a cpm/mg ¹)	(b cpm/mg)	(C/L/g)	(a cpm/mg ¹)	(b cpm/mg)
²⁵² Cf	202 g	1.82	4.28	9.192	4.60 × 10 ⁸		2.20	2 × 10 ⁻¹²	0.04	0.08 × 10 ⁻³
²⁵² Cf	17.08 g	6.03	140	4.06	1.23 × 10 ¹¹		0.75 × 10 ⁸	5 × 10 ⁻¹²	0.04	1.70 × 10 ⁻³
²⁵² Cf	900 g		1.30	5.79 × 10 ⁻²	1.70 × 10 ⁹			2 × 10 ⁻¹²	0.04	2.30 × 10 ⁻³
²⁵² Cf	2.046 g	6.11	136	30.0	9.40 × 10 ¹¹		1.40 × 10 ¹¹	6 × 10 ⁻¹²	0.04	1.07 × 10 ⁻³
²⁵² Cf	17.042 g		0.27	2.00 × 10 ⁴	13.00	6.42 × 10 ¹¹		0 × 10 ⁻¹⁰	0.04	1.40 × 10 ⁻⁴
²⁵² Cf	07.5 g	5.04	0.51 × 10 ³	1.06 × 10 ⁴			0.75 × 10 ¹¹	5 × 10 ⁻¹²	0.04	1.10 × 10 ⁻⁴
²⁵² Cf	20.407 g	6.63	2.30 × 10 ⁴	1.06 × 10 ³	2.06 × 10 ¹¹		1.04 × 10 ⁷	7 × 10 ⁻¹⁰	0.04	1.30 × 10 ⁻⁴
²⁵² Cf	276 g	6.42	1.50 × 10 ³	72.0	2.11 × 10 ¹²		0.70 × 10 ¹¹	2 × 10 ⁻¹¹	0.04	1.00 × 10 ⁻³
²⁵² Cf	20.7 g		0.10	1.40 × 10 ³	1.40 × 10 ³	6.97 × 10 ¹⁴		5 × 10 ⁻⁹	0.04	0.27 × 10 ⁻⁴
²⁵² Cf	30.6 g			1.20 × 10 ⁴		2.06 × 10 ¹¹	0.90 × 10 ¹¹	6 × 10 ⁻¹⁰	0.04	1.10 × 10 ⁻⁴
²⁵² Cf	25 g			2.00 × 10 ⁷		6.32 × 10 ¹⁴				
²⁵² Cf	1.20 g	7.20	1.00 × 10 ⁶	1.00 × 10 ³	4.30 × 10 ¹³		2.00 × 10 ¹¹	4 × 10 ⁻⁸	0.04	5.25 × 10 ⁻⁹
²⁵² Cf	20.07 g	7.03	6.13 × 10 ³	2.79 × 10 ⁴	6.00 × 10 ¹⁴		1.00 × 10 ¹¹	2 × 10 ⁻⁸	0.04	6.53 × 10 ⁻⁸
²⁵² Cf	2.60 g		4.07 × 10 ⁶	5.00 × 10 ⁶			0.50 × 10 ¹⁶	2 × 10 ⁻⁹	0.04	2.14 × 10 ⁻⁹
²⁵² Cf	04 g		5.40 × 10 ³	~200	6.12 × 10 ¹²					

The values for properties listed in this table are those in use at the end of the report period.
 From IAEA Publication 2, "Report of Committee II on Possible Uses for Internal Radiation (1959)" and the 1960 Supplement.
 Counting geometry, 20%.
 Decay data by 0.0100 (252) and orbital electron capture (252).
 Decay data almost entirely by isomeric transition to the 16-hr ground state, ^{252g}Cf.
 Decay data primarily by 0.0100, but 0.0100 decay by electron capture to ^{252g}Cf.

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

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	87.404 y	560	0	150	16.5	0	25
²³⁹ Pu	24.413 y	265.7	0	195	742.4	0	324
²⁴⁰ Pu	6580 y	290	0	8453	3.05	0	0
²⁴¹ Pu	14.98 y	360	0	166	1011	0	541
²⁴² Pu	373,000 y	18.5	7.509	1280	0	0	0
²⁴³ Pu	4.955 h	60	0	0	210	0	0
²⁴⁴ Pu	8.26×10^7 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	1340 y	75	2.126	1500	0	0	0
²⁴⁴ Am ^a	21.3 m	0	0	0	130	0	0
²⁴⁵ Am	2.07 h	0	0	0	0	0	0
²⁴⁶ Am	25.6 m	0	0	0	0	0	0
²⁴⁰ Cm	18.099 y	10	6.9	650	2.7	6.9	72
²⁴¹ Cm	8265 y	445.3	1.4	90	1544	1.4	600
²⁴² Cm	4711 y	5	0.866	170	0	0	0
²⁴³ Cm	1.64×10^7 y	77.63	0	260	281.33	0	1000
²⁴⁴ Cm	352,000 y	5.2	1.477	250	0	0	0
²⁴⁵ Cm	61 m	2.8	0	0	50	0	0
²⁴⁶ Cm	17,400 y	2	0	0	0	0	0
²⁴⁰ Bk	314 d	1706	0	1850	553.5	0	0
²⁴¹ Bk	1.227 h	350	0	0	950	0	0
²⁴² Cf	352 y	300	0	0	1735	0	0
²⁴³ Cf	13.08 y	2000	0	0	0	0	0
²⁴⁴ Cf	900 y	1948	0	0	5126	0	0
²⁴⁵ Cf	2.646 y	19.8	0	44	0	0	0
²⁴⁶ Cf	17.812 d	12.6	0	0	1330	0	0
²⁴⁷ Cf	60.5 d	75	0	1650	0	0	0
²⁴³ Bk	20.467 d	345	0	0	0	0	0
²⁴⁴ Bk	276 d	20	0	0	3060	0	0
²⁴⁴ Ge	39.3 h	1.26	0	0	1840	0	0
²⁴⁵ Ge	39.8 d	60	0	0	0	0	0
²⁴⁶ Ge	25 m	0	0	0	0	0	0
²⁴⁴ Pb	3.24 h	76	0	0	0	0	0
²⁴⁵ Pb	20.07 h	26	0	0	100	0	0
²⁴⁶ Pb	2.62 h	20	0	0	0	0	0
²⁴⁷ Pb	94 d	10	0	0	100	0	0

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

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.

It should be pointed out that $^{244}\text{C}_{\text{Am}}$ is a fictitious isotope that combines the properties of $^{244}\text{B}_{\text{Am}}$ and $^{244}\text{S}_{\text{Am}}$ according to their relative rates of production from ^{243}Am . The use of this isotope simplifies the calculations of the main transmutation chain involving ^{244}Am .