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THE PRODUCTION OF ²⁴¹Am BY NEUTRON IRRADIATION OF ²⁴¹Am

by

D. C. Stewart, R. W. Anderson,
and John Milsted

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ANL-6933
Physics
(TID-4500, 37th Ed.)
AEC Research and
Development Report

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60440

THE PRODUCTION OF CURIUM BY
NEUTRON IRRADIATION OF Am^{241}

by

D. C. Stewart, R. W. Anderson,
and John Milsted

Chemistry Division

November 1964

Operated by The University of Chicago
under
Contract W-31-109-eng-38
with the
U. S. Atomic Energy Commission

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THE PRODUCTION OF CURIUM BY NEUTRON IRRADIATION OF Am^{241}

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INTRODUCTION

The Chemistry Division at Argonne National Laboratory is currently interested in the production of multicurie amounts of highly alpha-active Cm^{242} for use in two different development projects, each of which establishes quite different requirements for the Am^{241} irradiation used to produce the curium. For the first study, the preparation of intensely active sources for an alpha-scattering device to be used for remote analysis of the lunar surface,⁽¹⁾ a highly isotopically pure material is needed so that even after considerable Cm^{242} decay the percentage of alphas emitted from other curium isotopes in the source will remain very small. In the other application, the preparation of a series of intense (α, n) sources for a neutron irradiation facility, the primary interest is in obtaining a maximum Cm^{242} yield with no particular concern for the presence of other curium isotopes. Because of these two problems, and because of an anticipated continuing need for Cm^{242} for varying research purposes, the present computer study of production of the isotope has been undertaken.

THE PRODUCTION PROCESS

Basically, Cm^{242} is produced by irradiation of Am^{241} in a neutron flux, a single neutron capture forming Am^{242} which decays to Cm^{242} with a 16-hr half-life. The actual process is somewhat more complicated. Figure 1 shows the total buildup chain assumed for the present computer calculations, and Figure 2 shows the initial steps in the chain in more detail. As can be seen, neutron irradiation of Am^{241} produces two isomers of Am^{242} ,

Fig. 1. Buildup Path

Fig. 2. Am^{241} Irradiation Detail

an excited state and the ground-state atom. The literature before 1960 is somewhat confusing, since until that time the assumption had been made that the metastable nuclide was the 16-hr isomer, whereas the ground state was the long-lived isotope. In that year, however, Asaro, Perlman, Rassmussen and Thompson⁽²⁾ demonstrated that the reverse is true, and that the process proceeds as shown in Figure 2.

There is still some uncertainty in the cross sections to be used. Those shown in Figure 2 were employed in the present calculations, but there is reason to believe that some cross sections may be in error. This is particularly true of the value of 5550 b, used for the formation of Am^{243} by neutron capture by Am^{242m} . This value was originally determined⁽³⁾ by subtracting an experimentally determined value of 2500 b for the fission cross section of Am^{242m} from a total destruction cross section of 8000 b for the isotope. More recent work⁽⁴⁾ has indicated that the fission cross section is actually 6390 b, so, if the total destruction cross section is as was estimated in the earlier work, the capture cross section should be substantially less than shown in Figure 2. If this error exists, the Cm^{242} -production curves shown in this report will be changed very little, since Cm^{242} formation proceeds by the other branch of the chain. The curves of curium composition would be affected, however, since the Am^{243} produced by this capture eventually contributes substantially to the buildup of Cm^{244} and higher curium isotopes, as can be deduced from Figure 1. The capture cross section of Cm^{242} itself also seems to be somewhat uncertain.

Computations for this report were carried out with an IBM 1620(II) computer, using methods which have been previously described⁽⁵⁻⁷⁾. Further data on the cross sections, half-lives, etc., of the nuclides shown in Figure 1 and 2 are given in Appendix A.

Cm^{242} YIELDS

Figure 3 shows the calculated yields of Cm^{242} at "intermediate" fluxes (3×10^{13} to 7×10^{14} $\text{n/cm}^2/\text{sec}$), as does Figure 4 on a different time scale of particular use in relation to the problem of producing Cm^{242} as free as possible from heavier curium isotopes.

Figure 5 shows yields to be expected in very high flux reactors, such as the HFIR (High Flux Isotope Reactor) now under construction at Oak Ridge National Laboratory or A^2R^2 (Argonne Advanced Research Reactor) being planned for Argonne. These are yields calculated in the reactor. Some small additional yield will be obtained at the end of the irradiation as the Am^{242} present continues to decay during cooling.

As would be expected, the time needed to reach maximum production of Cm^{242} decreases markedly with increase in the irradiation flux. The maximum attainable yield, however, increases with flux to a certain point, then drops off at much the same rate as is shown in Figure 6. Several different factors are involved in establishing this pattern. At low fluxes, the time of attaining the maximum Cm^{242} yield (about two half-lives at 3×10^{13} $\text{n/cm}^2/\text{sec}$) allows for considerable decay of the Cm^{242} formed in the early stages of the irradiation. On the other hand, at very high

Fig. 3. Cm^{242} Yields at Intermediate Fluxes - Target: Am^{241}

Fig. 4. Early Cm^{242} Buildup at Intermediate Fluxes - Target: Am^{241}

Fig. 5. Cm^{242} Yields at High Fluxes - Target: Am^{241}

Fig. 6. Variation of Maximum Yields with Flux

fluxes, the extremely rapid fission destruction of the Am^{242} intermediate begins to dominate even the relatively short decay half-life of 16 hr to the daughter Cm^{242} . (The two removal processes proceed roughly at equal rates at a flux of $4 \times 10^{15} \text{ n/cm}^2/\text{sec}$, if we may assume a destruction cross section of 3000 b, i.e., $\sigma_D f = \lambda_{\text{Decay}}$, at this point.) At these very high

fluxes an even more important factor is probably the rapid depletion of the Am^{241} starting material supporting the production chain. At a flux of $1 \times 10^{16} \text{ n/cm}^2/\text{sec}$, only 3.5% of the initial Am^{241} remains when the Cm^{242} reaches its maximum yield, as compared with 55% remaining at the same point in a $3 \times 10^{13} \text{ n/cm}^2/\text{sec}$ flux.

Since Cm^{242} is by far the predominant curium isotope present in the early stages of an Am^{241} irradiation, the total curium production curve follows the same general pattern as is also shown in Figure 6.

CURIUM COMPOSITION

Figures 7-13 present the change with time in the reactor of the composition of the curium fractions present in Am^{241} samples irradiated in a series of fluxes ranging from 3×10^{13} to $1 \times 10^{16} \text{ n/cm}^2/\text{sec}$.

Fig. 7. Curium Fraction Composition - Am^{241} Target and Flux = $3 \times 10^{13} \text{ n/cm}^2/\text{sec}$

Fig. 8. Curium Fraction Composition - Am^{241} Target and Flux = $7 \times 10^{13} \text{ n/cm}^2/\text{sec}$

Fig. 9. Curium Fraction Composition - Am^{241}
Target and Flux = $3 \times 10^{14} \text{ n/cm}^2/\text{sec}$

Fig. 10. Curium Fraction Composition - Am^{241}
Target and Flux = $7 \times 10^{14} \text{ n/cm}^2/\text{sec}$

Fig. 11. Curium Fraction Composition - Am^{241}
Target and Flux = $2 \times 10^{15} \text{ n/cm}^2/\text{sec}$

Fig. 12. Curium Fraction Composition - Am^{241}
Target and Flux = $5 \times 10^{15} \text{ n/cm}^2/\text{sec}$

Fig. 13

Curium Fraction Composition -
 Am^{241} Target and Flux = $1 \times 10^{16} \text{ n/cm}^2/\text{sec}$

To produce highly isotopically pure Cm^{242} , is it better to irradiate a short time at a higher flux, or a longer time at a low flux? The figures as shown do not answer this question very clearly. Table I, however, presents calculated data for the very early irradiation stages at three different fluxes. Approximately the same amount of total curium is produced each time, but it is clear that better purity is obtained if the lower fluxes are used. If a specified quantity of Cm^{242} must be produced, this obviously should be accomplished by increasing the amount of Am^{241} target rather than by lengthening the irradiation time.

Table I

PRODUCTION OF "PURE" Cm^{242}

Flux ($\text{n/cm}^2/\text{sec}$)	Time		Cm^{242} Produced, * mg	$\text{Cm}^{243} + \text{Cm}^{244}$ (% of Cm fraction)
	sec $\times 10^{-4}$	days		
3×10^{13}	15	1.74	1.25	0.00514
7×10^{13}	10.5	1.33	1.27	0.0108
3×10^{14}	4	0.46	1.27	0.0133

*Per gram of Am^{241} target. 1 mg Cm^{242} = 3.32 Curies.

Table II gives curium composition data for use in relation to the high-purity problem for short irradiations at the two lowest fluxes considered in this report. Factors are given in Appendix A for application to these composition figures at zero time and after one and three years of cooling. These factors can be used if it is desired to know the relative number of alphas being emitted by the various isotopes, rather than their weight ratios alone.

Table II
COMPOSITION OF CURIUM FRACTION
IN IRRADIATED Am^{241}

Time		Curium Fraction, %				Total Curium Yield, * mg
sec x 10 ⁻⁵	days	Cm ²⁴²	Cm ²⁴³	Cm ²⁴⁴	Cm ²⁴⁵	
Flux = 3 x 10 ¹³ n/cm ² /sec						
1	1.15	99.997	0.00327	3.07 x 10 ⁻⁵	-	0.65
2	2.30	99.993	0.00698	7.81 x 10 ⁻⁵	-	1.92
4	4.61	99.985	0.0152	2.48 x 10 ⁻⁴	-	4.87
8	9.22	99.962	0.0329	8.88 x 10 ⁻⁴	-	10.8
14	16.1	99.938	0.0595	0.00253	1.02 x 10 ⁻⁶	13.7
20	23.1	99.908	0.0866	0.00492	2.52 x 10 ⁻⁶	30.4
25	28.8	99.883	0.109	0.00749	4.93 x 10 ⁻⁶	34.4
30	34.6	99.857	0.132	0.0106	8.32 x 10 ⁻⁶	40.9
35	42.4	99.831	0.155	0.0140	1.30 x 10 ⁻⁵	47.1
Flux = 7 x 10 ¹³ n/cm ² /sec						
1	1.15	99.992	0.00761	1.59 x 10 ⁻⁴	-	1.51
2	2.30	99.983	0.0163	4.21 x 10 ⁻⁴	-	4.46
4	4.61	99.963	0.0355	0.00131	-	11.2
8	9.22	99.920	0.0758	0.00443	2.01 x 10 ⁻⁶	24.8
14	16.1	99.850	0.138	0.0122	1.04 x 10 ⁻⁵	44.3
20	23.1	99.779	0.198	0.0228	2.79 x 10 ⁻⁵	62.4
25	28.8	99.718	0.249	0.0335	5.15 x 10 ⁻⁵	77.1
30	34.6	99.654	0.300	0.0457	8.40 x 10 ⁻⁵	90.9
35	42.4	99.588	0.352	0.0597	1.26 x 10 ⁻⁴	103.4

*Per gram of Am^{241} target.

SUPPLEMENTARY IRRADIATION DATA

In a previous publication,⁽⁷⁾ computer programs were used to calculate the change with irradiation time of certain parameters of practical interest for the targets Pu^{242} , Am^{243} , Cm^{244} , and Cf^{252} . The variables studied included the change with time at various flux levels of total emitted alpha particles, total emitted neutrons from spontaneous fission, milliliters of helium accumulation from alpha decay, and heat evolved in the sample per gram of target, as well as the rate at which the various materials were converted to fission products. Similar data for Am^{241} are given in Figures 14-20.

Fig. 14

Alpha Activity at Intermediate Fluxes - Target: 1 Gram Am^{241}

Fig. 15
Alpha Activity at High Fluxes - Target: 1 Gram Am^{241}

Fig. 16. Cumulative Helium from Alpha Activity - Target: 1 Gram Am^{241}

Fig. 17. Total Neutron Emission from Spontaneous Fission - Target: 1 Gram Am^{241}

Fig. 18. Percent Original Atoms Converted to Fission Products - Target: Am^{241}

Fig. 19. Heat Production at Intermediate Fluxes - Target: 1 Gram Am^{241}

Fig. 20

Heat Production at High Fluxes -
Target: 1 Gram Am²⁴¹

As with total yield, the alpha-emission rate (see Figures 14 and 15) reaches a maximum at a flux of about 7×10^{14} n/cm²/sec, as would be expected. Cm²⁴² dominates the alpha-emission picture; after peak production is reached, the total number of alphas present drops rapidly as the isotope is destroyed by reactor processes or decay. The effect of the "feedback" chain:

is not included in the present calculations. This chain would have some effect on the alpha emission, heat evolution, and accumulated helium curves due to the relatively short half-life of Pu²³⁸ (89.6 yr) and the high fission-ability of Pu²³⁹ and Pu²⁴¹.

Helium accumulation in an Am²⁴¹ sample (see Figure 16) again probably reaches a maximum at the 7×10^{14} -n/cm²/sec flux level, although complete data were not calculated for this particular flux. As can be seen from the curves, substantial amounts of helium accumulate during a long irradiation at the lower fluxes, and adequate provisions must be made in designing the sample container to handle the resulting pressure buildup at reactor temperatures. Accumulation of fission product gas will also contribute substantially to this problem.

As with the other very-heavy-element targets studied, the neutron-emission curves for Am²⁴¹ (see Figure 17) first show a small peak followed by a much larger one later. The second peak, for all targets, is due to Cf²⁵² buildup, but in the case of an Am²⁴¹ target, the initial small peak is due to Cm²⁴² rather than to Cm²⁴⁴.

The fission product conversion rate (see Figure 18) is closely related to the heat-evolution data (see Figure 19 and 20), since heat evolution in the Am^{241} targets arises primarily from induced fission of the two Am^{242} isomers. (The heat curves include the contributions from induced and spontaneous fission and from natural decay of the actinides, but does not include heat from fission product decay.) Heat evolution from Am^{241} targets in very high fluxes reaches rather startling levels. A one-gram sample in a flux of $1 \times 10^{16} \text{ n/cm}^2/\text{sec}$ rapidly attains a maximum heat release of $2.74 \times 10^5 \text{ W}$. (A gram of pure U^{235} in the same flux would evolve $4.95 \times 10^5 \text{ W}$, assuming a fission cross section of 582 b.) The spectacular effect of flux increase on heat evolution in Am^{241} targets is demonstrated in Figure 21.

Fig. 21

Maximum Heat at Various
Fluxes - Target: 1 Gram Am^{241}

PRODUCTION OF VERY HEAVY ELEMENTS FROM Am^{241}

In an earlier publication⁽⁷⁾ the statement was made that Am^{241} was not a very satisfactory starting material for irradiations designed for the production of the heaviest nuclides (Cm^{244} and heavier). Gram for gram, as compared with Pu^{242} or Am^{243} as targets, this statement is certainly true, but the considerations listed below might justify reconsideration of Am^{241} as starting material for an actual production process:

- (1) As plutonium-based power reactors become more common, relatively large amounts of Am^{241} should become available as a byproduct of plutonium fuel reprocessing.

(2) Am^{241} can be obtained from reactor plutonium by a relatively simple "milking" operation, whereas other heavy nuclide targets, such as Pu^{242} , Am^{243} and Cm^{244} , require a complicated and expensive preliminary preparation involving a long irradiation of a large amount of Pu^{239} (the bulk of which is destroyed), followed by an industrial-scale remote-processing step.

(3) As shown in the calculations of this report, quite high total yields of curium can be obtained from an Am^{241} target irradiated in a properly selected neutron flux.

(4) The decay product of Cm^{242} , 89.6-yr Pu^{238} , is an isotope of much interest as an isotopic power source, and, as such, would be a most valuable byproduct.

The production scheme as envisaged would involve irradiation of Am^{241} in fluxes of the order of $5-7 \times 10^{14}$ n/cm²/sec to the point of maximum curium production, processing the irradiated material to isolate the curium fraction, storage of this fraction until the Cm^{242} had essentially completely decayed to Pu^{238} , and reprocessing to separate the plutonium and curium (now primarily Cm^{244}). A more detailed outline of this proposal is given in Appendix B.

REFERENCES

1. J. H. Patterson, A. L. Turkevich, and Ernest Franzgrote, J. Geophys. Research, to be published. Also see A. Turkevich, Science 134, 672 (1961).
2. F. Asaro, I. Perlman, J. O. Rassmussen, and S. G. Thompson, Phys. Rev. 120, 394 (1960).
3. G. C. Hanna, B. G. Harvey, N. Moss, and P. R. Tunncliffe, Phys. Rev. 81, 893 (1951).
4. E. K. Hulet, R. W. Hoff, H. R. Bowman, and M. C. Michel, Phys. Rev. 107, 1294 (1957).
5. J. Milsted, P. R. Fields, and D. N. Metta, ANL-6756 (Aug. 1963).
6. J. Milsted, P. R. Fields, and D. N. Metta, Revision of Ref. 5 to be published.
7. D. C. Stewart, R. W. Anderson, and J. Milsted, ANL-6932 (Oct. 1964).

APPENDIX A

Information Relative to the Production of Curium from Am²⁴¹A. Basic Data

Nuclide	Half-life	Primary Decay	Specific Activity, d/sec/ μ g	C/g	Decay Heat, W/g	Cross Sections		
						Capture		Fission
						b	cm ² /g	b
Pu ²³⁸	89.6 yr	α	6.22×10^5	16.8	0.56	489	1.25	16.8
Pu ²³⁹	24,360 yr	α	2,270	0.061	0.0019	340	0.86	810
Pu ²⁴⁰	6,580 yr	α	8,380	0.23	0.007	530	1.34	
Pu ²⁴¹	13 yr	β	4.23×10^6	114	0.014	350	0.88	1,100
Pu ²⁴²	3.79×10^5 yr	α	144	0.0039	0.00012	50	0.125	
Am ²⁴¹	458 yr	α	1.20×10^5	3.24	0.11	50 (to Am ^{242m}) 620 (to Am ²⁴²)	0.13 1.56	3.13
Am ^{242m}	152 yr	α	3.59×10^5	9.7	0.035	5,500	13.8	6,390
Am ²⁴²	16.01 hr	α , EC	2.99×10^{10}	8.1×10^5	2,800			3,000
Am ²⁴³	7,600 yr	α	7,150	0.193	0.0062	150	0.37	
Am ²⁴⁴	26 min	β	1.1×10^{12}	3×10^7	2.6×10^5			2,000
Cm ²⁴²	162.7 d	α	1.23×10^8	3,320	120	30	0.075	
Cm ²⁴³	35 yr	α	1.56×10^6	42	1.5	250	0.62	590
Cm ²⁴⁴	17.9 yr	α	3.03×10^6	82	2.9	35	0.086	
Cm ²⁴⁵	14,000 yr	α	3,850	0.10	0.0035	300	0.74	1,850
Cm ²⁴⁶	6,600 yr	α	8,170	0.22	0.0071	7	0.017	
Cm ²⁴⁷	$>4 \times 10^7$ yr	α	<1.34	-	-	50	0.12	200
Cm ²⁴⁸	4.7×10^5 yr	α	113	0.0031	-	8	0.020	
Cm ²⁴⁹	64 min	β	4.4×10^{11}	1.2×10^7	6.2×10^4	-		

For the most part, data in this table were abstracted from Reference (7).

B. Alpha Ratio Factors

Multiplication of the appropriate weight ratios from Table II or Figures 7-13 by the factors in the "zero time" column below gives the alpha-activity ratio between Cm²⁴² and the indicated associated curium isotope at the time the material is removed from the reactor. Multiplications of the same weight ratios by the figures in the other two columns give the alpha-emission ratios after one- and three-year cooling, respectively. Thus, for the Cm²⁴²/Cm²⁴³ alpha-emission ratio in the curium fraction of Am²⁴¹ irradiated 2×10^6 sec in a flux of 3×10^{13} n/cm²/sec (see Table II):

$$\frac{99.908}{0.0866} \times 78.9 = 90,000 \text{ at zero time}$$

$$\frac{99.908}{0.0866} \times 17.0 = 19,600 \text{ after 1-yr cooling}$$

$$\frac{99.908}{0.0866} \times 0.796 = 918 \text{ after 3-yr cooling.}$$

Nuclides	Alpha Ratio Factor		
	At Zero Time	After 1-yr Cooling	After 3-yr Cooling
$\text{Cm}^{242}/\text{Cm}^{243}$	78.9	17.0	0.796
$\text{Cm}^{242}/\text{Cm}^{244}$	40.6	8.93	0.433
$\text{Cm}^{242}/\text{Cm}^{245}$	32,000	6780	304
$\text{Cm}^{242}/\text{Cm}^{246}$	15,100	3300	148

C. Gamma Hazard

The following data are taken from the review by E. K. Hyde (UCRL-9148, Jan 1961).

Am^{241}

Gamma energy, keV	Absolute intensity (photons/alpha)
26.36	0.025
33.20	0.0011
43.46	0.007
59.56 ("60 keV")	0.359
(99.0 → 370)	(Number of others in very low abundance)

It will be seen that the "60 keV" photon dominates the gamma-emission picture for Am^{241} . One gram of the material is a 1.16-Curie (4.3×10^{10} photons/sec) source of these gammas. In the table below, mass absorption coefficients without coherent scattering (R. T. McGinnies, U. S. National Bureau Standards. Circular 583, Suppl., Oct 30, 1959) have been used to calculate tenth-value layers (thickness to reduce dose by a factor of ten) for 60-keV photons.

Material	Tenth-value Layer	Material	Tenth-value Layer
Air	10.9 m	Iron	2.64 mm
Water	11.8 cm	Lead	0.43 mm
Concrete*	3.7 cm	Tungsten	0.36 mm
Aluminum	3.5 cm	Platinum	0.28 mm
Silicon	3.4 cm	Uranium	0.14 mm

*Specific Gravity = 2.35

It will be seen that heavy metals are very effective in shielding against the 60-keV photon. The possibility that small amounts of long-lived gamma-emitting rare earth fission products may be present in the americium to complicate the shielding picture should not be overlooked, however.

Cm²⁴²

The most abundant gamma transition in Cm²⁴² decay is at 44.11 keV (3.9×10^{-4} photons per alpha). Other low-energy gammas and X rays also occur in very small abundance.

D. Neutron Hazard

Many of the heavier nuclides emit neutrons because of spontaneous fission decay, but most of those considered in this report are relatively stable in this respect. The more intense neutron emitters are the following:(7)

Nuclide	Spontaneous Fission Half-life, yr	Neutrons/sec/g
Pu ²⁴²	7.1×10^{10}	2900
Cm ²⁴²	7.2×10^6	2.9×10^7
Cm ²⁴⁴	1.35×10^7	1.52×10^7
Cm ²⁴⁶	1.66×10^7	1.23×10^7
Cm ²⁴⁸	4.6×10^6	4.4×10^7
Cf ²⁵²	85.5	2.33×10^{12}

Values in the last column were obtained by use of a ν value of 3.8 neutrons per fission (based on Cf²⁵²) throughout, and are thus probably somewhat high for the lighter isotopes. Buildup of neutron emission in irradiated Am²⁴¹ samples is graphed in Figure 17. A more complete and exact listing has been given in ANL-5800, 2nd Ed. (July 1963), p. 715.

A second, possibly more important, source of neutrons is by (α, n) reaction with light elements associated with large quantities of alpha

emitters. Both the neutron yield and the maximum neutron energy are dependent on the energy of the alphas in the source, and there is not complete agreement on data as reported by various investigators. The following table presents some selected values:

Source	Neutrons/sec per 10^6 Alphas	Neutrons/sec per g α Emitter	Maximum Neutron Energy, MeV	Ref.
Pu ²³⁹ -Be	60	1.36×10^5		(a)
Am ²⁴¹ -Be	74	8.88×10^6		(a)
Cm ²⁴² -Be	112	1.38×10^{10}		(a)
Cm ²⁴² -Be	189	2.35×10^{10}	11.5	(b)
Cm ²⁴² -C	0.27	3.36×10^7	8.0	(b)
Cm ²⁴² -O	0.16	2.01×10^7	6.3	(b)
Cm ²⁴² -Al	1.9	2.35×10^8	3.1	(b)
Po ²¹⁰ -Be	80			(c)
Po ²¹⁰ -Li	2.6			(c)
Po ²¹⁰ -B	24			(c)
Po ²¹⁰ -C	0.11			(c)
Po ²¹⁰ -F	12			(c)
Po ²¹⁰ -Al	0.74			(c)

(a) O. J. C. Runnalls and R. R. Boucher, Can. J. Phys. 34, 949 (1956).

(b) Calculated from E. D. Arnold, ORNL-3576 (April 1964) p. 36.

(c) J. H. Roberts, MDDC 731 (1944). Quoted in H. L. Anderson, Nat. Res. Counc. Nuc. Sci. Series Prelim. Rpt. #3 (Dec 1948).

E. Heat during Irradiation

Calculations have been made of the heat release of 8-in.-long x 0.5-in.-OD aluminum capsules containing 3 g AmO₂ plus 60 g of aluminum powder (USAEC Report ORNL-3452, Sept 20, 1963, p. 143). In a flux of 3×10^{14} n/cm²/sec the surface heat was computed at 438,000 BTU/hr/ft², the maximum surface temperature at 192°F, and the centerline temperature at 682°F in a helium-filled capsule, but 789°F with an air gap. Maximum internal pressures were 500 psi, assuming 100% release of fission gases.

F. Sample Encapsulation

Most Am^{241} samples are irradiated as AmO_2 -Al powder mixtures pelletized under high pressure, generally in aluminum containers. Pertinent information is as follows:

Material	Density, g/cm ³	Cross Section		Comment
		b	cm ² /g	
AmO_2	10.0 $\pm$ 10%	-	-	ORNL-3627 (Oct 1964) p. 124.
AmO_2	11.76	-	-	Calculated by W. T. Carnall from cell constants presented by Asprey et al., J. Amer. Chem. Soc. 77, 1707 (1955).
CmO_2	11.82	-	-	
EC grade Al	2.70	0.241	0.005	Al > 99.45%.
"2S" Al	2.71	0.271	0.006	Al > 99%; Cu + Si to 1%; Fe to 0.2%.
"18-8" stainless steel	7.91	2.94	0.0312	AISI 304. 18-20% Cr; 8-10% Ni; Mn, 2% max; Fe 70-74%.

Oak Ridge National Laboratory [ORNL-3558 (Aug 31, 1963), p. 63] reports that 89-90% of theoretical density is obtained when 14 v/o PuO_2 -aluminum powder mixtures are submitted to pressures of 20 tons/in.². Lawrence Radiation Laboratory (M. S. Coops, Livermore, personal communication) indicates that they obtain about 87-89% of theoretical density in americium oxide-aluminum powder pressed pellets. Recent preparations at Argonne, at 17-tons/in.² pressure, gave an average density (1.83 g/cc) equivalent to 76% of theoretical density for a series of Al- AmO_2 mixtures with aluminum to oxide weight ratios ranging from 8 to 150. Twelve Be- AmO_2 mixtures (weight ratio of 2.6 to 1) gave an average density of 1.85 g/cm², or 78.5% of theoretical.

G. Shielding of Irradiated Americium-241 and Pure Cm^{242}

W. G. Ruehle Jr. has developed a method for estimating shielding required for Am^{241} irradiated in fluxes ranging from 1×10^{12} to 5×10^{14} n/cm²/sec (GEAP-0974, Feb 25, 1957). Application of this method to 0.5 g of Am^{241} irradiated for 6 mo at a flux of 7×10^{13} n/cm²/sec gives a dose of 100 mr/hr through 6 in. of lead at one-foot distance after 1 day of cooling.

E. D. Arnold (ORNL-3576, April 1964, pp. 176-180) gives graphs of dose rate versus distance from the source for large, unshielded, pure Cm^{242} sources, as well as dose rates versus shield thickness for a number of sources shielded with iron, lead, uranium, and water.

APPENDIX B

Production of Very Heavy Elements

The steps required to produce gram amounts of Am^{243} and Cm^{244} from Pu^{239} versus Am^{241} targets are compared in outline below. Processing yields of 100% are assumed throughout. Yield data are from references (5) and (6) or the present report. Final irradiation steps were calculated for a flux of 2×10^{15} n/cm²/sec since more complete data were available for that level. More detailed analysis might show other fluxes to be more desirable, based on the monetary value assigned to the various products and byproducts.

A. Pu^{239} Starting Material

Process Step	Products and Salvage	Comments
1. Encapsulation of 2.71 kg Pu^{239}		Glovebox operation. Book value of Pu^{239} = \$43/g.*
2. Irradiation for 15 mo at 3×10^{14} n/cm ² /sec		Lower-flux irradiation because of heat-release problem.
3. First chemical processing	Pu^{242} 100 g Am^{243} 25.8 g Cm^{244} 22.1 g Cm^{245} 0.33 g Cm^{246} 0.35 g	Remote operation on production plant scale. Am-Cm recovery a separate step involving elimination of 2.0 kg of fission products.
4. Re-encapsulation of Pu^{242} as oxide		Remote or semiremote operation.
5. Irradiation of Pu^{242} for 2 mo at 2×10^{15} n/cm ² /sec		Irradiation time arbitrarily selected. Target could be left to go to higher burnup, if desired.
6. Second chemical processing	Pu^{242} 60.6 g (salvage) Am^{243} 18.8 g Cm^{244} 17.2 g Cm^{245} 0.26 g Cm^{246} 0.30 g	Remote operations. Fission product content relatively low (about 3 g).

*Federal Register, 28FR, 5314, May 29, 1963.

B. Am^{241} Starting Material

Process Step	Products and Salvage	Comments
1. "Milking" of 100 g Am^{241} from aged, high-burnup plutonium		Semiremote or remote operation. Present book value of $\text{Am}^{241} = \$1500/\text{g}^*$
2. Encapsulation of Am^{241} as oxide		Semiremote or remote operation.
3. Irradiation for 2 mo at $2 \times 10^{15} \text{ n/cm}^2/\text{sec}$		Would involve severe heat-release problems in early stages.
4. (Optional.) Preliminary processing to remove Pu^{242} to avoid its later excessive contamination with Pu^{238} .	Pu^{242} 6.49 g	Remote operation (about 50% converted to fission products). Material left in form to expedite final processing.
5. Four years cooling to permit Cm^{242} to decay to < 1% of original amount (7+ half-lives).		Special long-term storage facilities required for "steady-state" operation.
6. Completion of chemical processing	Pu^{238} 32.8 g Am^{241} 0.12 g (salvage) Am^{243} 2.80 g Cm^{244} 5.56 g Cm^{245} 0.09 g Cm^{246} 0.11 g	Remote operation. Fission product activity comparatively low due to long cooling time.

*Chem. & Eng. News, August 19, 1963. p. 26.

Under present circumstances, this comparison would indicate that Pu^{239} is the superior starting material for heavy isotope production in spite of the fact that its use necessitates two encapsulation-irradiation-processing cycles (one involving a very long irradiation and a major processing operation) rather than one, as with the Am^{241} targets. This situation might change, however, if the "book value" of Am^{241} eventually approaches that of Pu^{239} due to its availability as a relatively limited-use byproduct of large-scale reactor fuel-processing operations. The extent of the demand for Pu^{238} or for Cm^{242} itself as heat sources also obviously would have to be considered.

Under the conditions shown, about 8.6% of the Am^{241} target is converted to Am^{243} or heavier nuclides, while 3.9% of the Pu^{239} starting material is similarly transmuted. Overall production of useful heavy nuclides of atomic weight 238 or greater is 48% for the Am^{241} case and 5.4% for Pu^{239} . However, if "burnt-out" reactor plutonium containing higher plutonium isotopes can be used as a starting material rather than pure Pu^{239} , this picture is markedly altered. The use of such an enriched plutonium would probably still require a double irradiation-processing schedule if microgram quantities of elements above berkelium were among the desired products.