MAR 3 0 1960

IDO-16571
Chemistry - Radiation and Radiochemistry
TID-4500 (15th Ed.)

CALCULATION OF THE COMPOSITION OF REACTOR-IRRADIATED HEAVY NUCLIDES

R. P. Schuman and R. L. Tromp December 17, 1959

AEC RESEARCH AND DEVELOPMENT REPORT

PHILLIPS PETROLEUM CO.
ATOMIC ENERGY DIVISION
(UNDER CONTRACT NO. AT (10-1)-205)
IDAHO OPERATIONS OFFICE
U. S. ATOMIC ENERGY COMMISSION

DISCLAIMER

PRICE $1.50

Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in USA.

CALCULATION OF THE COMPOSITION OF REACTOR-IRRADIATED HEAVY NUCLIDES

by

R. P. Schuman and R. L. Tromp

A B S T R A C T

Calculations have been made of the compositions of reactor-irradiated Th-230, Th-232, U-233, U-235, U-236, U-238, Pu-239, Pu-242, and Cm-244. Pile fluxes with resonance flux per ln E interval equal to 1/12 (typical of MTR fuel) or 1/30 (typical of MTR beryllium pieces) of the thermal flux were used for the calculations; thermal fluxes of 5×10^{13}, 2×10^{14}, and 1×10^{15} n/cm^2 sec were assumed. Calculation methods, assumptions concerning cross sections, and possible methods of producing specific nuclides are discussed.

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

TABLE OF CONTENTS

LIST OF TABLES

LIST OF TABLES

LIST OF FIGURES

LIST OF FIGURES

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

CALCULATION OF THE COMPOSITION OF REACTOR-IRRADIATED HEAVY NUCLIDES

by

R. P. Schuman and R. L. Tromp

INTRODUCTION

Two neutron reactions are most probable for heavy nuclides in a thermal reactor: fission, especially with the even-odd and odd-odd nuclides; and capture, with all nuclides. Other reactions occurring with small yields, such as n,2n reactions with prompt fission neutrons, are also of interest to reactor operations. Because of the capture reactions, isotopic compositions of fuel and fertile material in a pile will change during operation, with corresponding changes in reactivity. Also, isotopes will be produced which may be valuable by-products or may, on the other hand, complicate the chemical reprocessing or the refabrication of the fuel.

Since the high burnup of fuel is essential to the low cost operation of most reactors contemplated today, and since isotopic capture products will accumulate in chemically reprocessed fuel, it is of interest to calculate the composition of the fuels and fertile materials after long irradiations. The calculations can serve as a guide in determining the value of fuels, how often their reprocessing is necessary from a nuclear standpoint, and how often they must be isotopically re-enriched. The calculations are also of importance in planning irradiations for the production of such heavy nuclides as U-232, Np-237, Pu-242 and the transplutonium elements.

Because the Nuclear Physics Branch at the MTR is interested in obtaining neutron cross section data on the heavy nuclides of interest to reactor operation, calculations have been made of the compositions of Th-230, Th-232, U-233, U-235, U-236, U-238, Pu-239, Pu-242, and Cm-244 irradiated at several different fluxes for different time periods. If a mixture of these isotopes is irradiated, the final composition can be found from these calculations by summing the final products from the several input isotopes. These calculations were made to serve as a guide for scheduling irradiations to produce samples for cross section measurements. The calculations will also be helpful in evaluating thermal Th-232 - U-233 breeder reactors and comparing U-233, U-235, and Pu-239 as enriched reactor fuels.

METHOD OF CALCULATION

The radioactive nuclides in a reactor may disappear both by radioactive decay and by neutron absorption. If the reactor operates at a constant flux, each nuclide may be considered as having an effective decay constant in the reactor which we will call the destruction constant "ℓ". The destruction constant of the ith nuclide is the sum of the radioactive decay constant, λ_i and the neutron reaction rate constant, $nv\sigma_i$, i.e.

$$\ell_i = \lambda_i + nv\sigma_i \qquad (1)$$

where ℓ_i = destruction constant of ith nuclide.

λ_i = radioactive decay constant of ith nuclide (the sum of partial decay constants for α and β decay, spontaneous fission, etc.).

nv = constant flux of reactor.

σ_i = total reaction cross section in the reactor flux (the sum of fission, radiative capture, n,2n, etc., cross sections).

Likewise, new nuclides will be produced in the reactor by radioactive decay and neutron reactions. There will be an effective production constant, which we have designated by p, for the production of a nuclide from each parent. The rate of production of a nuclide will be the sum of the products of the production constant from each parent and the amount of each parent. If a given nuclide is produced by radioactive decay (beta, alpha, or electron capture), the production constant is equal to the partial decay constant of the parent leading to that nuclide. If the nuclide is produced by a neutron reaction (radiative capture or n,2n), the production constant is equal to the product of pile flux and pile cross section for the reaction leading to the nuclide. The rate of change of a nuclide in a constant flux reactor is given by:

$$\frac{dN_i}{dt} = -\ell_i N_i + \sum_j p_j N_j \qquad (2)$$

where N_i = amount of ith nuclide

N_j = amount of a particular parent j

ℓ_i = destruction constant of the ith nuclide

p_j = production constant from parent j.

The equations for the rate of change of the amounts of nuclides in a constant flux reactor are entirely analogous to those for members of the natural radioactive decay chains except that the constants "ℓ" and "p" are substituted for the appropriate λ's. Thus in a reactor there are capture chains analogous to the natural radioactive decay chains, and the amounts of the chain members can be calculated using the Bateman equation[1,2] developed for the natural radioactive decay chains.

For the special case of production from only one starting nuclide and by only one path, the amount of the ith chain member is given by the Bateman equation:

$$N_i = (p_1 p_2 \cdots p_{i-1}) N_1^o \left[\frac{e^{-\ell_1 t}}{D_1} + \frac{e^{-\ell_2 t}}{D_2} + \cdots \frac{e^{-\ell_i t}}{D_i} \right] \quad (3)$$

where $D_1 = (\ell_2 - \ell_1)(\ell_3 - \ell_1)\cdots(\ell_i - \ell_1) = \prod_{j \neq 1} (\ell_j - \ell_1)$

$D_2 = (\ell_1 - \ell_2)(\ell_3 - \ell_2)\cdots(\ell_i - \ell_2) = \prod_{j \neq 2} (\ell_j - \ell_2)$

$D_i = (\ell_1 - \ell_i)(\ell_2 - \ell_i)\cdots(\ell_{i-1} - \ell_i) = \prod_{j \neq i} (\ell_j - \ell_i)$

N_1^o = Initial amount of starting nuclide.

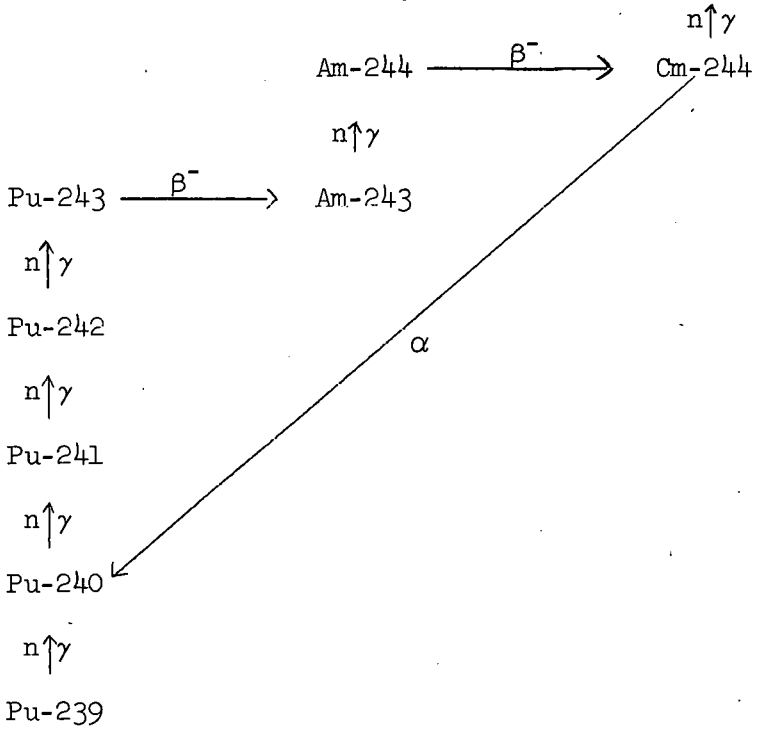
If production of a nuclide is possible by several paths, the amount of the nuclide is given by the sum of the Bateman equations, one for each path, with the production constants being those leading along the appropriate path. If more than one isotope is initially present, a summation is made for production from each initial nuclide.

The first step in calculating the composition of pile-irradiated material is to establish the capture chains. Although the chains may contain many branches, usually there are only one or two significant reactions for each chain member. Thus, in a reactor, Pu-239 can undergo neutron capture, undergo fission, undergo an n,2n reaction, or may undergo alpha decay; however, only the capture and fission reactions are important. The chains used for the calculations are given in Figures 1 through 4. In most cases the chains are quite simple with only occasional two way branching. Since the calculations are made for fairly high fluxes, and for irradiation periods of days to a few years, the approximation is made that long-lived nuclides, $t_{\frac{1}{2}} > 100$ years, are stable with respect to radioactive decay. Also the approximation is made that short-lived nuclides, $t_{\frac{1}{2}} < 1$ day, decay instantaneously, so that neutron capture by, or decay of a long-lived nuclide is

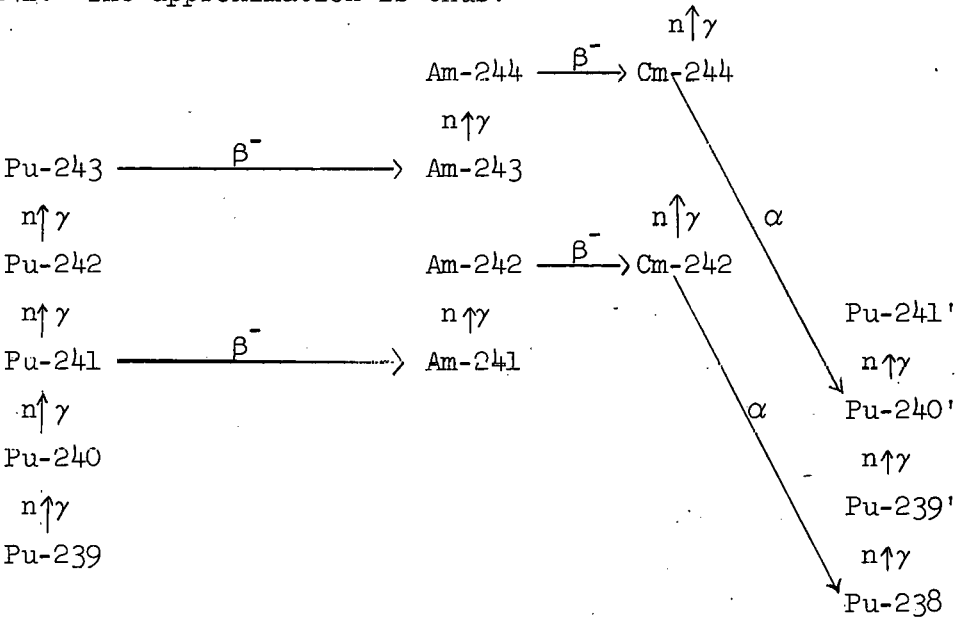
considered as leading directly to the next long-lived member of the chain. The approximation will not normally cause a significant error for times much longer than the half-life of the short lived nuclide, since nuclides with half-lives less than one day will usually decay before building up in quantities large enough to undergo significant neutron reactions. In any case, when one assumes the short-lived parent decays instantaneously, the actual amount of short-lived parent plus the amount of long-lived daughter is virtually equal to the abundance calculated for the daughter.

In order to simplify the calculations, a number of further approximations have been made which will normally not lead to serious errors. Inspection of Figures 1 through 4 shows many minor branches, such as the electron capture decay of Am-242m to Pu-242, neutron capture by U-232, n,2n reactions on U-233, U-235, U-236, Pu-239, Pu-242, and Cm-244, and others, which have been ignored since their contributions are very small. The assumption of constant flux irradiation is not correct for most operating reactors which have scheduled down periods. For nuclides with half lives much greater than the down period, the calculated compositions will be essentially correct if the average flux for the entire irradiation period is used for the calculations. A comparison of continuous vs cyclic irradiation of Th-232 has been made by R. G. Nisle[3].

An additional complication in the capture chains is that the alpha decay of capture products can lead to closed loops in which a nuclide may reappear as a result of four neutron captures, two beta decays, and an alpha decay. Usually the loops are unimportant and can be neglected in the calculations, but in the case of Pu-239, two capture products Cm-242 and Cm-244 have sufficiently short alpha half lives so that the loops will be important. Because of the loops, the concentrations of Pu-239, Pu-240, and Pu-241 in napkin rings are appreciable at high integrated fluxes even after the initial Pu-239 and the first generation Pu-240 and Pu-241 have been burned up. The closed loop which is due to Cm-244 α decay is:



This loop largely accounts for the presence of Pu-240 and Pu-241 in highly irradiated napkin rings. In order to make approximate calculations of the abundances of Pu-239, Pu-240, and Pu-241 in highly irradiated Pu-239, the closed loops have been approximated by assuming that recycle Pu-239, Pu-240, and Pu-241 are new members of a capture chain which ends at second generation Pu-241. The approximation is thus:



Since the amount of material recycled is fairly small due to losses by fission and to neutron capture of Cm-242 and Cm-244, the approximation will give satisfactory abundances for Pu-239, Pu-240, and Pu-241 for the times of interest. Since the "recycle chain" is cut off at Pu-241', the calculated abundances of Am-241, Cm-242, Pu-242, and Am-243 will be decidedly low at the very highest integrated fluxes. Calculated abundances that are expected to be highly in error have been omitted from the tables of results. In the other chains, the loops have been ignored since their effects are small. Because of the α decay of Pu-238, some U-234 and secondary U-235 capture product, which are not shown in the calculations, will be produced in very highly irradiated samples of U-235 and U-236. Actually there is a limit to the burnout of U-235 in U-235 → U-236 mixtures.

By using the approximation that the recycle members in a loop are new members of a chain, it is possible to calculate the compositions of the irradiated nuclides using the Bateman equations. Since in this case two chain members have the same destruction constants, the Bateman equation, as given in equation (3), becomes indeterminate. A satisfactory solution of the Bateman equation can be obtained by permitting the two destruction constants to differ by a small value ϵ, and then allowing ϵ to approach zero. The solution for the amount of the kth member of a chain in the case where two chain members, m and n, have the same destruction constant, $\ell_m = \ell_n$, is:

$$N_k = N_1^o(p_1 p_2 \cdots p_{k-1}) \left[\left(\sum_{j \neq m,n}^{k} \frac{e^{-\ell_j t}}{D_j} \right) + \frac{\left[t - \left(\sum_{j \neq m,n} \left(\frac{1}{\ell_j - \ell_m} \right) \right] e^{-\ell_m t}}{D_m} \right] \quad (4)$$

where $D_j = \prod_{i \neq j} (\ell_i - \ell_j)$

$D_m = \prod_{i \neq m,n} (\ell_i - \ell_m)$

A somewhat similar solution is obtained if there are two pairs of nuclides with identical destruction constants, as there will be for the calculation of recycle Pu-241.

A number of calculations have been made with a desk calculator. These were made as check calculations for the IBM-650, and for some of the short chains, were the only calculations made. Most of the compositions were calculated using the IBM-650 and a radioactive decay program set up by

Adrian Grimaud of the Theoretical Physics and Applied Mathematics Branch of MTR Technical.[4] The program is such that the machine cannot calculate abundances if two destruction constants are the same; therefore, for the loops, an additional approximation was made (for the machine calculations) that the recycle nuclide had destruction constants ~2% lower than the original. For our case, where the recycle nuclides have much greater destruction constants than several other loop members, the assumption makes only ~2% (or less) error in the calculated abundances, which is insignificant in comparison with other uncertainties.

The input data cards for the IBM-650 supplied the cross sections and decay constants of the nuclides and the chain member to which they lead. The machine then determined the possible paths, calculated the abundances of the nuclides along each path, then added the production from all the possible paths to give the total production for the desired fluxes and times. Since the machine was using floating point arithmetic which can carry eight decimal places, the accuracy of the results (which are sums and differences of large numbers) is limited to around one part in 10^7 of the maximum abundance until after the maximum abundance is reached. Therefore the very small yields of the heaviest nuclides from low integrated flux irradiations could not be calculated by the program. Early values, which are in error due to the limited number of decimal places carried by the machine, have been omitted from the tables of results. Calculations made with a desk calculator were carried out to ten decimal places.

CROSS SECTIONS

The calculations of the production of heavy nuclides in a pile depend upon the radioactive decay constants and the neutron cross sections. The decay constants are well known in the range where they are important. The values used for the calculations are given in Table 1.[5]

The largest uncertainties in the calculations arise from the assumed cross sections. The neutron cross sections of the elements vary greatly with neutron energy, and the energy spectrum of neutrons varies from reactor to reactor and from place to place in one reactor. To calculate the composition of irradiated material, ideally one should know the neutron energy spectrum as a function of time, and the cross section of each nuclide as a function of neutron energy. Further, self-shielding and flux depression corrections

should be made. Thus the pile destruction and production constants of a particular nuclide will depend upon pile spectrum and sample thickness, and will change with time as the pile spectrum shifts, the flux density varies, and the self-shielding changes. Thus the abundances calculated in this report may be quite seriously in error due to erroneous assumptions as to neutron spectrum and self-shielding. Another complication in comparing calculated and observed results is the large possible error in the integrated flux, "nvt", of the irradiation. Since the "nvt" of irradiations are often poorly known, and compositions vary rapidly with "nvt", it is probably best to use one component (usually the initial component) as an internal monitor; thus for irradiations up to $\sim 6 \times 10^{21}$ n/cm^2, U-235 or Pu-239 burnout is a fairly good flux monitor for enriched fuel or napkin rings.

For our calculations, we have assumed very thin samples so that self shielding and flux depression are negligible. The pile flux is considered as being composed of three energy groups; thermal (Maxwellian) neutrons, resonance (1/E) neutrons, and unmoderated fission neutrons.[6] For some of the nuclides, Th-232, U-233, U-235, U-236, and Pu-239, a pile flux was assumed in which the resonance flux per ln E interval is 1/12 of the thermal flux and the fast flux (unmoderated fission neutron flux) is equal to the thermal flux; this roughly approximates the flux seen by MTR fuel.[7] For U-238, Pu-239, Pu-242, and Cm-244, a pile flux was assumed in which the resonance flux per ln E interval is 1/30 of the thermal flux and the fast flux is equal to 1/3 of the thermal flux; this roughly approximates the flux seen by the plutonium napkin rings irradiated in the beryllium region of the MTR.

When resonance integral data were available, the pile cross sections used for the calculations were calculated from resonance integrals and thermal (Maxwellian) cross sections.[6-14,17] When resonance integral data were unavailable, pile cross sections were taken,[8,15,16] and assumed to be the cross section for a pile flux with resonance flux equal to 1/30 thermal flux. Some literature values of the cross sections are given in Table II. In certain cases, cross section values had to be estimated. Most of the estimated values were those which gave a reasonable fit between the calculated and observed compositions of the three KAPL napkin rings (irradiated Pu-239).[16] The values are really only "educated guesses", since the measured abundances of the napkin ring components were only approximate, and

the amount of data was insufficient to determine accurate cross section values. The measured abundances show that certain pile cross sections apparently varied considerably from napkin ring to napkin ring, probably due to the varying resonance to thermal flux ratio. The actual pile cross sections that were used for the calculations are given in Table III for the case of resonance flux per ln E interval equal to 1/12 thermal flux, and in Table IV for the case of resonance flux per ln E interval equal to 1/30 thermal flux. Stoughton and Halperin have recently published a compilation of effective pile cross sections for $nv_{res}/nv_{th} = 1/12$[18] for most of the nuclides considered for the calculations up to and including Pu-242; their compilation was not available when these calculations were made. With the exception of Pa-233, their cross sections usually agree well within the quoted limit of error with those used in our calculations. Most of the cross sections for nuclides above Pu-242 are poorly known.

DISCUSSION OF RESULTS

I. Irradiation of Ionium

Protactinium-231 is a naturally occurring decay product of U-235, and can be isolated from uranium ore residues. However, because it is very rare (0.33 g Pa-231 per metric ton uranium at equilibrium), and difficult to isolate chemically, the alternate production of Pa-231 by pile irradiation of ionium concentrates (mixture of Th-230 from U-238 decay and Th-232) is quite attractive. In order to evaluate pile irradiation of Th-230 (+ Th-232) as a method of producing gram quantities of Pa-231, hand calculations of the composition of a pile irradiated ionium-thorium mixture has been made, see Table V and Figure 5. The calculations show that Pa-231 can be produced in fairly high concentrations in the ionium, but that, due to Th-232 in the sample, large amounts of Pa-233 will also be produced so that the irradiated ionium will have to be cooled for a long time before it is chemically processed. The irradiated ionium will also contain a considerable fission product activity due to U-233 fission.

II. Irradiation of Th-232 and U-233

There are two power reactor cycles that hold the best promise of cheap nuclear power: the Th-232 - U-233 breeder reactor, which can be either a thermal or fast reactor, and the U-238 - Pu-239 breeder reactor, which must be a fast reactor. If efficient power-breeder reactors are developed, the

earth's thorium and uranium resources form an almost limitless source of energy, comparable in magnitude to the fusion energy content of the deuterium in the oceans.

In many respects, the thermal Th-232 - U-233 breeder reactor holds the best promise of producing cheap power, since it would use the more abundant thorium as its energy source, would require a smaller inventory of fissionable material, and would be similar to the well-tested enriched U-235 thermal reactors presently in operation. However, a very well designed reactor will be required if it is to breed using Th-232 and U-233. The value of thermal η for U-233 of 2.28 neutrons produced/neutron absorbed,[8] leaves relatively few neutrons, 0.28 n/neutron absorbed, for parasitic capture, leakage, and breeding gain. Also, an abundant nuclide in the Th-232 - U-233 cycle is 27.4 day Pa-233, which has a moderately high parasitic capture cross section, and which will build up in large amounts in a high flux reactor. Neutron capture by Pa-233 will use up neutrons and also decrease the U-233 production. One advantage of U-233 over U-235 as a reactor fuel is that the capture product of U-233 is fertile U-234 which forms fissionable U-235 upon neutron capture.

Another problem in the operation of a Th-232 - U-233 breeder is that U-232 is produced which will build up in the uranium, and Th-228, which is formed through U-232 α decay, will build up in the thorium. These fairly short lived alpha emitters, through subsequent decay, result in large sources of gamma emitters and thus increase the cost of fuel handling.

Calculations have been made of the compositions of thin Th-232 and U-233 irradiated in a pile flux with resonance flux per ln E interval equal to 1/12 thermal flux, see Tables VI and VII and Figures 6 and 7. The calculations demonstrate U-232 production due to the fast Th-232 (n,2n) reaction, and also the very high buildup of Pa-233, especially at high fluxes. The Pa-233 is probably even a bigger unknown in evaluating the Th-232 - U-233 cycle than η for U-233, and Pa-233 buildup may very likely limit the flux at which the reactor can operate. Better cross section data on Pa-233 are needed for evaluating Th-232 - U-233 breeder reactors. In an actual reactor, the thorium breeding blanket elements will be thick, particularly for resonance neutrons, so the effective pile cross section of Th-232 will be smaller than that used in these calculations.[14,18] The main result of this "cross section change" will be to decrease the ratio of the products (except

n,2n products) to thorium by approximately the ratio of effective thick Th-232 pile cross section to the thin Th-232 cross section used in the calculations. A comparison of cyclic and continuous irradiation of Th-232 has been made by Nisle.[3]

Even though a Th-232 - U-233 reactor does not breed or break even, it will greatly extend the fission energy resources. The U-233 produced from Th-232 will be an excellent enriched reactor fuel. The U-233 will be more difficult to fabricate than U-235 due to the much more intense alpha and gamma irradiation, but with respect to neutron economy, it will be better fuel since U-233 has a higher η than U-235 and also the second order capture product of U-233 is fissionable U-235, while the second order capture product of U-235 is the relatively short lived U-237 which beta decays to essentially nonfissionable Np-237.

III. Irradiation of U-235 and U-236

Since there is a great deal of experience in operating enriched uranium reactors, our main interest in calculating the composition of irradiated U-235 and U-236 was to evaluate the possiblity of producing valuable by-products like Np-237 and plutonium rich in Pu-238. The effect of the capture products on the enriched uranium fuel reprocessing has been discussed elsewhere.[19]

Although the thermal cross sections and resonance integrals of the nuclides in the chain are better known than those in the other chains, the calculations still only approximate actual compositions, since the assumed constant pile flux only approximates the actual pile fluxes. The results of the calculations for a resonance flux per ln E interval equal to 1/12 the thermal flux are given in Tables VIII and IX and Figures 8 and 9. The calculations show that irradiated enriched reactor fuel, such as MTR fuel, is an excellent source of Np-237, particularly if the enriched uranium had been previously irradiated so that it initially contained U-236. The Np-237 production is highly dependent upon the resonance flux since U-236 has a low thermal capture cross section and a high resonance capture integral.

IV. Irradiation of U-238

Since a thermal U-238 - Pu-239 breeder reactor is impossible, the thermal pile irradiation of U-238 is of interest as an extender for U-235 or Pu-239 fuel, as a producer of plutonium, and as a source of certain heavy elements as Np-237 and possibly transplutonium isotopes.

The results of the U-238 calculations are given in Table X and Figure 10. The calculations were made for a resonance to thermal flux ratio of 1/30, a fast flux equal to 1/3 of the thermal flux, and thin U-238 samples. The actual composition of irradiated U-238 is highly dependent upon the resonance flux since U-238 has a low thermal cross section and a high resonance capture integral. The composition will also depend upon the thickness of the U-238 since U-238 samples, like Th-232 and U-236 samples, will show considerable self protection especially for resonance neutrons.[14,18] Therefore, the effective pile cross section of typical U-238 breeding blanket samples will be less than that used in the calculation; however, the yield of the capture products will be nearly those calculated times the ratio of effective thick U-238 cross section to thin U-238 cross section.

V. Irradiation of Pu-239, Pu-242, and Cm-244

The long term pile irradiation of plutonium, as plutonium-aluminum alloy napkin rings, has been the source of most of the transcurium elements used for nuclear and chemical studies. At present a larger scale plutonium irradiation is getting underway which will produce samples of Am, Cm and transcurium elements sufficiently large to permit more accurate cross section measurements. This program will consist of irradiating Pu-239 as plutonium aluminum alloy until the remaining plutonium is largely Pu-242. The material will then be chemically processed and the resulting plutonium (mainly Pu-242) and perhaps also curium (mainly Cm-244) further irradiated. Because of our interest in the irradiation of Pu-239, Pu-242 and Cm-244 for the production of heavy element samples for MTR chopper cross section measurements, as well as the interest in Pu-239 as a reactor fuel, calculations have been made of the composition of Pu-239, Pu-242, and Cm-244 irradiated to a very high integrated flux. The results of the calculations, assuming a resonance flux per ln E interval of 1/30 the thermal flux, are given in Table XI and Figure 11 for Pu-239, in Table XII and Figure 12 for Pu-242, and in Table XIII and Figure 13 for Cm-244. The composition of Pu-239 irradiated in a flux with $nv_{res}/nv_{th} = 1/12$ has also been calculated, see Table XIb and Figure 11b. Other calculations of the composition of irradiated plutonium and curium have been made by UCRL, ANL,[15] and Hanford.[20] The cross sections (especially those of the heaviest nuclides) used for these calculations often differ quite extensively from those we have used.

Since many of the cross sections of the heaviest nuclides are very poorly known or unknown, we have selected values for certain cross sections in order to obtain a fair fit between the calculated yields and the observed KAPL napkin ring yields. Table XIV compares the calculated and observed yields for three KAPL and three ANL napkin rings irradiated in the MTR.[15,16] The "nvt" for comparison was chosen to give the best agreement between observed and calculated yields and is appreciably different from the "nvt" reported for the irradiation. For the lower "nvt" values, the burnup of Pu-239 was used to estimate the "nvt" for comparison. For the higher "nvt" values, the yields of Cm-244 and Pu-242 were used to estimate the "nvt" for comparison. As shown in Table XIV, fair agreements between calculated and observed yields are obtained. Unfortunately, the observed yields are only approximate, and no attempt has been made to obtain the best cross section values, so the estimated cross section values used for the calculations should be considered only as educated guesses.

Also, since many pile cross sections, such as the Pu-240 and Pu-242 capture cross sections, have very high resonance integrals, the yields of the nuclides will depend strongly upon the resonance flux contribution. The mass spectrometrically determined ratio of Cm-245/Cm-244 shows that the cross section ratio of Cm-245 to Cm-244 varied considerably from napkin ring to napkin ring. Most likely Cm-244 (possibly also Cm-245) has a very large resonance integral compared to its thermal cross section. The comparison of plutonium irradiated with nv_{res}/nv_{th} of 1/12 to $nv_{res}/nv_{th} = 1/30$ shows the quite large dependance on resonance flux, even when both curves are normalized to the same burnup of Pu-239, see Figure XIc.

Because the IBM-650 carries only eight significant figures when using floating point arithmetic, and the calculated yields are determined as the difference of large numbers, the limit of accuracy of the calculated yields is roughly 10^{-7} of the maximum abundance (it may be somewhat more or less depending upon the actual coefficients in the Bateman equation). Therefore, the yields of the heaviest nuclides can not be calculated for the shorter irradiations. Another error in the calculated yields results from cutting off the Cm-242 - Pu-238 and Cm-244 - Pu-240 cycles at Pu-241, so that the calculated abundances of Pu-242, Am-241 and Am-243 are low for the highest "nvt" values.

PRODUCTION OF HEAVY NUCLIDES

It is of interest to look at possible methods of producing samples of nuclides for cross section measurements. The nuclides for which cross section measurements will be most valuable are those which are found in appreciable amounts in reactor fuel or fertile material, and which can be produced in high purity and in gram quantities for chopper measurements. Only methods which do not require isotope separation will be considered here. Many nuclides, such as Th-230, U-234, U-236, Pu-240, Pu-241, etc., will probably be best produced by isotope separation. The production methods are summarized below:

Pa-231: Gram samples of Pa-231 can be produced by pile irradiation of ionium concentrates. A long decay period before the sample is processed will be necessary to allow Pa-233 to decay.

Pa-233: Gram samples of Pa-233 can be produced by irradiating Th-232, then chemically separating and purifying Pa. The sample will contain very small amounts of Pa-231 and U-233 growing in from the 27.4 day beta decay of Pa-233. The Pa-233 has a specific activity of $\sim$23,000 curies/gram of $\sim$400 kev gamma. Capture cross section vs energy data on Pa-233 are needed to evaluate Th-232 - U-233 breeder reactors.

U-232: Sufficient U-232 for chopper cross section measurements can be obtained by irradiating gram quantities of Pa-231, and then chemically separating and purifying U. Repeated short irradiations and chemical processing will be required to produce U-232 with a minimum U-233 content.

U-233: The beta decay of the Pa-233 samples will produce very high purity U-233.

U-236: Isotopically enriched U-236 can be further purified from U-235 by selectively burning out the U-235 by pile irradiation (preferably with a highly thermal flux). The α decay of Pu-238 to U-234 and subsequent neutron capture to U-235, as well as the fast neutron n,2n reaction on U-236, will limit the burnout of U-235; however, if the highly irradiated uranium is purified from Np and Pu, then reirradiated for a short time (so that Pu-238 does not have a chance to build up) the U-235 content can be reduced to a minimum.

U-237: Uranium containing ~1% U-237 can be produced by the very high flux irradiation of U-236 followed by chemical separation and purification of U. If the U-236 is sufficiently free of U-235, it should be possible to obtain the fission cross section of U-237. If the original U-236 is sufficiently free of U-238, the buildup of U-238 in the sample will give a capture cross section for U-237.

Pu-238: The alpha decay of Cm-242 will give very pure samples of Pu-238. Sufficient Pu-238 for chopper measurements can be made in this manner.

Pu-240: The alpha decay of Cm-244 will give very pure samples of Pu-240. The plutonium will contain less fissionable Pu-239 and Pu-241 than the presently available isotopically enriched Pu-240 and so will be valuable for fission cross section measurements.

Pu-242: Highly irradiated plutonium will ultimately become essentially pure (~99%) Pu-242. Napkin ring Pu-242 has been used by ANL for chopper cross section measurements,[21] and larger and purer samples should become available from the large scale irradiation of Pu. The Pu-242 samples will be the best samples for chemical studies of plutonium since they contain only about 10% of the specific alpha activity of Pu-239.

Pu-244: Pu-244 is produced by the minor Pu-243 capture branch in the Pu-239 and Pu-242 capture chains. A small concentration of Pu-244 will build up in residual Pu-242, and will be sufficient for resonance integral measurements. If a very high flux reactor becomes available, $\phi \gg 10^{15}$ n/cm^2 sec, quite high concentrations of Pu-244 could be produced by the long term irradiation of Pu-239 or better of Pu-242. Because of its very long half-life, pure Pu-244 would be very valuable for heat capacity measurements on plutonium compounds.

Am-243: The high flux irradiation of Pu-239 or Pu-242 will produce high purity Am-243. Napkin ring Am-243 has been used by ANL for chopper cross section measurements.[21] Am-243 samples will be best for americium chemistry studies.

Cm-242: Quite pure Cm-242 can be produced by the fairly short irradiation of Am-241. The curium could be used for cross section measurements, or allowed to decay to produce pure Pu-238.

Cm-243: The residual curium after the Cm-242 in the Cm-242 samples have decayed will be quite rich in Cm-243. Sufficient Cm-243 may be produced for chopper or crystal spectrometer fission cross section measurements.

Cm-244: Cm-244 is the most abundant curium isotope produced in napkin rings. Purer Cm-244 for cross section measurements could be produced by the short term irradiation of pure Am-243.

Cm-245: Because of its high cross section, Cm-245 builds up to only a small extent in napkin ring Pu. Very small samples of pure Cm-245 could be produced by irradiating Pu rich in Pu-244, then immediately separating the plutonium from Cm and Am. The Pu-245 in the resulting Pu will then beta decay through Am-245 to pure Cm-245. Cf-249 alpha decay would also produce very small samples of pure Cm-245. The pure Cm-245 samples could be used for thermal cross section and resonance fission integral measurements.

Cm-246: Cm-246 will build up in very highly irradiated Pu-239, Pu-242, or Cm-244, but will be of low purity. Cm-246, contaminated with Cm-248, will be produced by Cf decay (preferably californium as rich as possible in Cf-250), and could be used to obtain a better value for the Cm-246 capture cross section.

Cm-247: Cm-247 will build up to a low abundance in very highly irradiated Pu-239, Pu-242, or Cm-244. Perhaps, if a very high flux ($\phi \gg 10^{15}$ n/cm^2 sec) reactor were available, it could be built up to a greater abundance than Cm-245 and a pile σ_f and certain decay properties of Cm-247 could be determined. If pure Cm-247 could be obtained, say by isotope enrichment, it would be extremely valuable for chemical studies and heat capacity measurements because of its long half-life.

Cm-248: A moderate concentration of Cm-248 could be produced in the residual curium if Pu-242 or Cm-244 were irradiated for a long time in a very high flux ($\phi \gg 10^{15}$ n/cm^2 sec) reactor. Very small samples of fairly pure Cm-248 can be produced by californium (preferably rich in Cf-252) decay. Such curium would be useful for thermal cross section, resonance integral, and decay property studies. Also Cm-249 could be produced for decay studies. Curium rich in Cm-248 will probably be the best curium available for chemical studies.

Bk-249: The large scale irradiation of Pu will ultimately produce milligram amounts of Bk-249 for chemical studies, cross section measurements, and studies of Bk-250 produced by neutron capture.

Cf-249: The beta decay of Bk-249 will produce pure Cf-249 for thermal and resonance cross section measurements. Because of its moderately long half-life and ease of production, Cf-249 samples will be the best californium for chemical studies.

Cf-250: Cf-250 is a constituent of napkin ring californium. Purer Cf-250, produced by irradiation of Bk-249, might be used for thermal and resonance cross section measurements.

Cf-251: Cf-251 is a minor constituent of napkin ring californium. It can not be produced in any degree of purity by pile irradiation. Perhaps better cross section data can be obtained from the low purity samples. Very small samples of quite pure Cf-251 could be produced by the decay of cyclotron produced Es-251.

Cf-252: Fairly pure Cf-252 can be obtained by the long term irradiation of Pu-239, Pu-242, and Cm-244. A determination of the Cf-252 resonance capture integral would be valuable. Cf-252 is very valuable for studying spontaneous fission. The chief interest in Cf-252 production at UCRL is in its use as a target for production of heavy elements by heavy ion bombardment with the Heavy Ion Linear Accelerator.

Transcalifornium Elements: Isotopes of einsteinium and fermium are also produced in long term irradiated Pu-239, Pu-242, and Cm-244. When larger samples become available, better cross section data on Es-253, Es-254, Es-255, Fm-254, and Fm-255 can be obtained. Both 20d E-253 and 480d E-254 will be valuable target materials for the production of new heavy nuclides.

REFERENCES

1. H. Bateman, "Solution of a System of Differential Equations Occurring in the Theory of Radioactive Transformations", Proc. Cambridge Phil. Soc. 15, 423 (1910).

2. W. Rubinson, "The Equations of Radioactive Transformation in a Neutron Flux", J. Chem. Phys. 17, 542 (1949).

3. R. G. Nisle, "Calculation of Isotope Production by the Cyclic Irradiation of Thorium", IDO-16504 March 26, 1959.

4. A. V. Grimaud, "Radioactive Decay Utility Program", IBM-650 Library Routine No. 03-019.

5. D. Strominger, J. M. Hollander, and G. T. Seaborg, "Table of Isotopes", Revs. Modern Phys., 30, 585 (1958).

6. D. J. Hughes, "Pile Neutron Research" Addison-Wesley Publishing Co., Inc., Cambridge, Mass., (1953).

7. J. Halperin, J. O. Blomeke, and D. A. Mrkvicka, "Effective Reactor Cross Sections in MTR Fuel Assemblies," Nuclear Science and Engineering 3, 395 (1958).

8. D. J. Hughes and R. B. Schwartz, "Neutron Cross Sections", BNL 325 July 1, 1958.

9. J. P. Butler, M. Lounsbury, and J. Merritt, "Determination of the Neutron Capture Cross Sections of Pu-242, Pu-238 and Am-243", CRC-628 Jan. 1956 and "The Neutron Capture Cross Sections of Pu-238, Pu-242 and Am-243 in the Thermal and Epicadmium Regions", Can. J. Phys. 35, 147 (1957).

10. T. A. Eastwood, A. P. Baerg, C. B. Bigham, F. Brown, M. J. Cabell, W. E. Grummitt, J. C. Roy, L. P. Roy, and R. P. Schuman, "Radiochemical Methods Applied to the Determination of Cross Sections of Reactor Interest", Progress in Nuclear Energy, Ser. 1., Physics and Mathematics, Vol. 3, p. 55 Pergammon Press, Ltd. (London) (1959).

11. R. R. Smith, T. O. Passell, S. D. Reeder, N. P. Alley, and R. L. Heath, "Neutron Absorption Cross Section of Pa-233", IDO-16226 Oct. 27, 1955.

12. J. A. Harvey and J. E. Sanders, "Summary of Data on the Cross Sections and Neutron Yields of U-233, U-235, and Pu-239", Progress in Nuclear Energy, Ser. 1., Physics and Mathematics, Vol. 1, p. 1, McGraw Hill Book Co., Inc., (New York) (1956).

13. J. A. Harvey and R. B. Schwartz, "Thermal and Resonance Neutron Cross Sections of Heavy Nuclei", Progress in Nuclear Energy, Ser. 1., Physics and Mathematics, Vol. II, p. 51, Pergammon Press, Ltd. (London) (1958).

14. R. L. Macklin and H. S. Pomerance, "Resonance Capture Integrals" Progress in Nuclear Energy, Ser. 1., Physics and Mathematics, Vol. I, p. 179, McGraw Hill Book Co., Inc., (New York) (1956).

15. W. C. Bentley, H. Diamond, P. R. Fields, A. M. Friedman, J. E. Gindler, D. C. Hess, J. R. Huizenga, M. G. Inghram, A. H. Jaffey, L. B. Magnusson, W. M. Manning, J. F. Mech, G. L. Pyle, R. Sjoblom, C. M. Stevens, and M. H. Studier, "The Formation of Higher Isotopes and Higher Elements by Reactor Irradiation of Pu-239; some Nuclear Properties of the Heavier Isotopes". Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955. Paper P/809; Vol. 7, p. 261. United Nations (1956).

16. J. P. Butler, T. A. Eastwood, H. G. Jackson, T. L. Collins, M. E. Jones, F. M. Rourke, and R. P. Schuman, "Review of the Joint AECL-KAPL Studies of the Transuranium Elements", KAPL-1781, July 2, 1957.

17. R. G. Fluharty, M. S. Moore and J. E. Evans, "The Total and Fission Cross Sections of Uranium-233 below 1 Kev", Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, Sept. 1958. Paper P/645; Vol. 15, p. 111. United Nations (1958).

18. R. W. Stoughton and J. Halperin, "Heavy Nuclide Cross Sections of Particular Interest to Thermal Reactor Operations: Conventions, Measurements, and Preferred Values", Nuclear Science and Engineering 6, 100 (1959).

19. E. D. Arnold, "Effect of Uranium Recycle on Transuranic Element Buildup". Nuclear Science and Engineering, 3, 707 (1958).

20. F. P. Brauer and H. H. Burley, "Trans-Plutonium Isotope Buildup by Neutron Irradiation of Plutonium", HW-58596, Dec. 15, 1958.

21. R. E. Cote', L. M. Bollinger, R. F. Barnes, and H. Diamond, "Slow-Neutron Cross Section of Pu-240, Pu-242, and Am-243", Physical Review 114, 505 (1959).

22. J. A. Phillips, "The n,2n Cross Section of Th-232 for Fission Neutrons", AERE-R/R-2366 August 1957.

TABLE I

Decay Constants of Heavy Nuclides
Used for Calculations

Nuclide	Half-Life	Decay Process	Decay Constant assumed for calculation sec^{-1}	To
Th-230	8.1×10^4 yr	α	0	
Th-231	25.6 hr	β^-	(a)	Pa-231
Th-232	1.42×10^{10} yr	α	0	
Th-233	23 min	β^-	(a)	Pa-233
Pa-231	3.43×10^4 yr	α	0	
Pa-232	1.32 day	β^-	(a)	U-232
Pa-233	27.4 day	β^-	2.93×10^{-7}	U-233
Pa-234	6.66 hr	β^-	(a)	U-234
Pa-234m	1.16 min	β^-	(a)	U-234
U-232	74 yr	α	3.05×10^{-10}	Ignore
U-233	1.62×10^5 yr	α	0	
U-234	2.50×10^5 yr	α	0	
U-235	7.1×10^8 yr	α	0	
U-236	2.42×10^7 yr	α	0	
U-237	6.75 day	β^-	1.19×10^{-6}	Np-237
U-238	4.50×10^9 yr	α	0	
U-239	23.5 min	β^-	(a)	Np-239
Np-237	2.20×10^6 yr	α	0	
Np-238	2.10 day	β^-	3.82×10^{-6}	Pu-238
Np-239	2.34 day	β^-	3.43×10^{-6}	Pu-239
Np-240	7.3 min	β^-	(a)	Pu-240
Pu-238	88 yr	α	2.5×10^{-10}	Ignore
Pu-239	2.44×10^4 yr	α	0	
Pu-240	6580 yr	α	0	
Pu-241	13.0 yr	β^-	1.69×10^{-9}	Am-241
Pu-242	3.79×10^5 yr	α	0	
Pu-243	5.0 hr	β^-	3.85×10^{-5}	Am-243
Pu-244	7.5×10^7 yr	α	0	

TABLE I (Cont)

Decay Constants of Heavy Nuclides
Used for Calculations

Nuclide	Half-Life	Decay Process	Decay Constant assumed for calculation sec^{-1}	To
Am-241	458 yr	α	0	
Am-242	100 yr	90% β^-, E.C.	2.2×10^{-10}	Ignore
Am-242m	16.0 hr	80% β^-, E.C.	(a)	0.8 to Cm-242 0.2 Ignore
Am-243	7950 yr	α	0	
Am-244	26 min	β^-	(a)	Cm-244
Cm-242	162.5 d	α	4.94×10^{-8}	Pu-238
Cm-243	35 yr	α	6.28×10^{-10}	Ignore
Cm-244	18.4 yr	α	1.19×10^{-9}	Pu-240
Cm-245	8000 yr	α	0	
Cm-246	6600 yr	α	0	
Cm-247	$> 4 \times 10^7$ yr	α	0	
Cm-248	4.2×10^5 yr	89% α, S.F.	0	
Cm-249	64 min	β^-	(a)	Bk-249
Bk-249	314 d	β^-	2.55×10^{-8}	Cf-249
Bk-250	3.13 hr	β^-	(a)	Cf-250
Cf-249	360 yr	α	0	
Cf-250	10.9 yr	α	2.02×10^{-9}	Ignore
Cf-251	~800 yr	α	0	
Cf-252	2.2 yr	97% α, S.F.	9.98×10^{-9}	Ignore

(a) Considered as decaying immediately to daughter.

TABLE II

Thermal Cross Sections and Resonance Integrals of Heavy Nuclides

Nuclide	Process	Thermal (Maxwellian at 20.4°C) Cross Section - barns	Resonance Integral excluding 1/v contribution - barns
Th-230	capture	27*[8]	
Th-232	capture	7.4[8]	82[13,14]
	n,2n	0.0125**[22]	
Pa-231	capture	200[8]	
Pa-233	capture to UX_2	43[11], 20[9]	470[9], 400[11]
	capture to UZ	25[11], 25*[9]	270[11]
U-232	capture	300*[8]	
	fission	80*[8]	
U-233	capture	61[12]	115[17]
	fission	530[12]	812[17]
U-234	capture	97[8]	710[13]
U-235	capture	104[12]	50[7]
	fission	576[12]	270[7,14]
U-236	capture	6[8,10]	310 (calc.), 340[13], 257[10]
U-238	capture	2.71[8]	
	n,2n	0.0145**(calc.)	280[7,13], 290[14]
Np-237	capture	170[8]	600[7]
Np-238	fission	1600*[8]	
Np-239	capture	60*[8]	

TABLE II (Cont)

Thermal Cross Sections and Resonance Integrals of Heavy Nuclides

Nuclide	Process	Thermal (Maxwellian at 20.4°C) Cross Section - barns	Resonance Integral excluding 1/v contribution - barns
Pu-238	capture	$454^{(9)}$ $403^{(9)}$	$950^{(9)}$ $3260^{(9)}$
	fission	$17^{(10)}$	$25^{(10)}$
Pu-239	capture	$338^{(12)}$	$1600^{(7)}$
	fission	$770^{(12)}$	$2000^{(7)}$
Pu-240	capture	$285^{(8)}$	$8400^{(7,13)}$
Pu-241	capture	$350^{(13,15)}$	1200(est.)
	fission	$1060^{(13)}$	1800(est.)
Pu-242	capture	$23^{(7,9)}$ $19^{(9)}$	$870^{(7)(9)}$ $1275^{(9)}$
Pu-243	capture	$170^{*(8,15)}$	
Pu-244	capture	$2.1^{*(16)}$ $1.4^{*(15)}$	
Am-241	capture to 16hr Am-242	$750^{*(8)}$	
	capture to 100yr Am-242	$50^{*(8)}$	
	fission	$3.1^{*(8)}$	
Am-242m	fission	$2500^{*(8)}$	
Am-242	fission	$6400^{*(8)}$	
	capture	$1600^{*(8)}$	
Am-243	capture	$82^{(9)}$ $74^{(8,9)}$	$1580^{(9)}$ $2290^{(9)}$
Cm-242	capture	$20^{*(8)}$	
Cm-243	capture	$250^{*(8)}$	
	fission	$700^{*(8)}$	

TABLE II (Cont)

Thermal Cross Sections and Resonance Integrals of Heavy Nuclides

Nuclide	Process	Thermal (Maxwellian at 20.4°C) Cross Section - barns	Resonance Integral excluding 1/v contribution - barns
Cm-244	capture	$15^{*(8)}_{20^{*(15)}}$	
Cm-245	capture fission	$200^{*(15)}$ $1800^{*(15)}$	
Cm-246	capture	$15^{*(15)}$	
Cm-247	capture	$180^{*(15)}$	
Cm-248	capture	$6^{*(16)}_{4^{*(15)}}$	
Bk-249	capture	$500^{*(8)}_{1100^{*(15)}}$	
Cf-249	capture fission	$270^{*(15)}$ $600^{*(15)}$	
Cf-250	capture	$1500^{*(15)}$	
Cf-251	capture	$3000^{*(15)}$	
Cf-252	capture	$30^{*(8)}_{25^{*(15)}}$	

* Pile cross sections
** Cross Section for unmoderated fission neutrons

TABLE III

Pile Cross Sections Used for Calculations where Resonance Flux = 1/12 Thermal Flux

Nuclide	Process	Cross Section used for calculations barns	To	Cross Section Recommended by Stoughton and Halperin[18] barns
Th-230	capture	27	Th-231	---
Th-232	capture	14.5	Th-233	14.5 ± 1.5
	n,2n	0.0125	Th-231	---
Pa-231	capture	200	Pa-232	---
Pa-233	capture	90	Pa-234 Pa-234m	170 ± 40
U-232	capture	300	U-233	---
	fission	80	---	---
U-233	capture	73	U-234	70 ± 20
	fission	617	---	610 ± 30
U-234	capture	160	U-235	150 ± 20
U-235	capture	112	U-236	125 ± 20
	fission	620	---	595 ± 30
U-236	capture	32	U-237	40 ± 10
U-237	fission	200*	---	---
U-238	capture	26	U-239	26 ± 2
Np-237	capture	226	Np-238	
Np-238	fission	1900	---	---
Np-239	capture	70	Np 240	80 ± 20
Pu-238	capture	550	Pu-239	---
	fission	20	---	---
Pu-239	capture	485	Pu-240	480 ± 50
	fission	965	---	980 ± 60
Pu-240	capture	995	Pu-241	1100 ± 200
Pu-241	capture	475	Pu-242	500 ± 100
	fission	1285	---	1250 ± 100
Pu-242	capture	96	Pu-243	130 ± 30
Am-243	capture	220	Am-244	---
Cm-244	capture	35	Cm-245	---
Cm-245	fission	2250	end	---
	capture			

* Estimated

TABLE IV

Cross Sections of Heavy Nuclides
Used for Calculations
For Resonance Flux = 1/30 Thermal Flux

Nuclide	Cross Section barns	To	Cross Section barns	To
U-237	200	ignore		
U-238	12.1	U-239	0.0035	U-237
Np-237	192	Np-238		
Np-238	1600	fission		
Np-239	60	Np-240		
Pu-238	502	Pu-239	18	fission
Pu-239	380	Pu-240	860	fission
Pu-240	590	Pu-241		
Pu-241	400	Pu-242	1150	fission
Pu-242	57	Pu-243		
Pu-243	180	Pu-244		
Pu-244	1.8	ignore		
Am-241	750	Am-242m	50	Am-242
Am-242	6400	fission	1600	ignore
Am-243	170	Am-244		
Cm-242	15	Cm-243		
Cm-243	250	ignore	700	fission
Cm-244	20	Cm-245		
Cm-245	450	Cm-246	1400	fission
Cm-246	13	Cm-247		
Cm-247	150*	Cm-248	1000(b)	fission
Cm-248	6	Cm-249		
Bk-249	950	Bk-250		
Cf-249	270	Cf-250	600	fission
Cf-250	1200*	Cf-251		
Cf-251	1500*	Cf-252	1800	fission
Cf-252	30	ignore		
recycle Pu-238	500	recycle Pu-239		
recycle Pu-239	360	recycle Pu-240	840	fission
recycle Pu-240	570	recycle Pu-241		
recycle Pu-241	1500	fission or ignore		

* Estimated

TABLE V

Composition of Irradiated 9% Th-230 - 91% Th-232 Mixture
in Atoms per Million Initial Th-230 + Th-232 Atoms

Time days	nvt_2 n/cm^2	Th-230	Pa-231	U-232	Th-232	Pa-233	U-233	Fissions
0	0	90,000	0	0	910,000	0	0	0
For Flux = 3×10^{13} n/cm^2 sec								
1.16 d	3×10^{18}	89,993	7.3	0	909,961	38.2	0.56	0
2.32 d	6×10^{18}	89,985	14.6	0	909,922	75.3	2.22	0
3.47 d	9×10^{18}	89,978	21.8	0	909,884	111.3	4.95	0.01
6.95 d	1.8×10^{19}	89,956	43.6	0.1	909,767	213.2	19.21	0.15
11.6 d	3×10^{19}	89,927	72.6	0.2	909,612	335.6	51.3	0.29
23.2 d	6×10^{19}	89,854	145	0.9	909,225	585.4	185.9	2.14
34.7 d	9×10^{19}	89,782	216	1.9	908,838	771.2	381	6.72
69.5 d	1.8×10^{20}	89,564	429	7.6	907,677	1,088	1,176	44.1
116 d	3×10^{20}	89,274	705	20.8	906,132	1,242	2,426	161.9
232 d	6×10^{20}	88,554	1,363	77.4	902,280	1,301	5,457	814
347 d	9×10^{20}	87,839	1,977	163.9	898,444	1,299	8,019	1,925
695 d	1.8×10^{21}	85,731	3,580	549	887,035	1,283	13,356	7,289
For Flux = 1×10^{14} n/cm^2 sec								
1.16 d	1×10^{19}	89,976	24.3	0	909,871	127.3	1.9	0
2.32 d	2×10^{19}	89,951	48.5	0.1	909,742	250.8	7.4	0.03
3.47 d	3×10^{19}	89,927	72.6	0.2	909,612	370.6	16.4	0.09
6.95 d	6×10^{19}	89,854	144.8	0.8	909,225	709.2	63.4	0.71
11.6 d	1×10^{20}	89,757	240.3	2.4	908,709	1,116	168.1	3.16

TABLE V (Cont)

Composition of Irradiated 9% Th-230 - 91% Th-232 Mixture
in Atoms per Million Initial Th-230 + Th-232 Atoms

Time days	nvt_2 n/cm^2	Th-230	Pa-231	U-232	Th-232	Pa-233	U-233	Fissions
23.2 d	2×10^{20}	89,515	475.1	9.3	907,419	1,940	599	23.2
34.7 d	3×10^{20}	89,274	704.7	20.6	906,132	2,550	1,207	71.9
69.5 d	6×10^{20}	88,554	1,363	77.5	902,280	3,576	3,537	455
116 d	1×10^{21}	87,603	2,172	199	897,169	4,051	6,794	1,587
232 d	2×10^{21}	85,269	3,892	654	884,520	4,196	12,867	7,083
347 d	3×10^{21}	82,997	5,245	1,217	872,048	4,147	16,244	15,038
695 d	6×10^{21}	76,540	7,715	2,837	835,679	3,974	19,567	44,218
For Flux = 3×10^{14} n/cm^2 sec								
1.16 d	3×10^{19}	89,927	72.6	0.2	909,612	382	5.6	0.03
2.32 d	6×10^{19}	89,854	145	0.8	909,225	751	22.0	0.24
3.47 d	9×10^{19}	89,782	216	1.8	908,838	1,109	48.6	0.81
6.95 d	1.8×10^{20}	89,564	429	7.5	907,677	2,118	185	6.23
11.6	3×10^{20}	89,274	705	20.5	906,132	3,321	482	27.4
23.2	6×10^{20}	88,554	1,363	77.4	902,280	5,736	1,642	194.6
34.7	9×10^{20}	87,839	1,977	164	898,444	7,487	3,161	584
69.5	1.8×10^{21}	85,731	3,580	551	887,035	10,319	8,123	3,350
116	3×10^{21},	82,997	5,245	1,219	872,048	11,483	13,277	10,387
232	6×10^{21}	76,540	7,715	2,845	835,679	11,506	18,436	36,480
347	9×10^{21}	70,584	8,694	3,904	800,827	11,049	19,247	65,484
695	1.8×10^{22}	55,357	8,256	4,465	704,751	9,724	17,853	147,650

TABLE VI

Composition of Irradiated Th-232 in Atoms per Million Initial Th-232 Atoms
Resonance Flux per ln E Interval Equal to 1/12 Thermal Flux

Time days	nvt_2 n/cm^2	Th-232	Pa-231	Pa-233	U-232	U-233	U-234	U-235	U-236	U-237	Np-237
0	0	1,000,000	0	0	0	0	0	0	0	0	0
For Thermal Flux = 5×10^{13} n/cm^2 sec											
11.6	5×10^{19}	999,275	0.62	627	0.003	95.3	1.60	0.003	0	0	0
23.2	1×10^{20}										
46.3	2×10^{20}	997,102	2.45	1,693	0.048	1,128	24.0	0.251	0.002	0	0
81.1	3.5×10^{20}	994,934	4.22	2,126	0.143	2,641	69.6	1.26	0.012	0	0
116	5×10^{20}	992,770	5.93	2,301	0.283	4,242	135	3.43	0.050	0	0
232	1×10^{21}	985,592	11.3	2,401	1.03	8,900	463	22.3	0.674	0.0008	0.004
463	2×10^{21}	971,392	20.3	2,373	3.39	14,570	1,413	121	7.85	0.0098	0.094
811	3.5×10^{21}	950,475	30.6	2,322	7.87	18,006	3,017	334	47.8	0.062	1.05
1160	5×10^{21}	930,008	37.9	2,272	12.3	18,950	4,461	695	134	0.175	4.26
2320	1×10^{22}	864,914	49.2	2,113	22.3	18,306	7,390	1,476	705	0.932	43.2
For Thermal Flux = 2×10^{14} n/cm^2 sec											
1.16	2×10^{19}	999,710	0.25	285	0	4.19	0.26	0	0	0	0
2.32	4×10^{19}	999,420	0.50	562	0.002	16.5	1.04	0.003	0	0	0
4.63	8×10^{19}	998,840	0.99	1,090	0.008	64.0	4.12	0.018	0	0	0
8.11	1.4×10^{20}	997,970	1.72	1,822	0.024	188	12.5	0.092	0	0	0
11.6	2×10^{20}	997,102	2.45	2,489	0.048	366	25.2	0.262	0.002	0	0
23.2	4×10^{20}	994,212	4.79	4,305	0.185	1,267	96.7	1.96	0.022	0	0
46.3	8×10^{20}	988,457	9.19	6,591	0.684	3,840	357	13.7	0.323	0.0009	0.001
81.1	1.4×10^{21}	979,888	15.1	8,156	1.86	7,974	960	60.4	2.58	0.0092	0.014
116	2×10^{21}	971,392	20.3	8,724	3.40	11,395	1,718	144	9.16	0.0365	0.083
232	4×10^{21}	943,603	33.3	8,863	9.45	16,993	4,484	611	87.4	0.403	1.89
463	8×10^{21}	890,386	46.4	8,381	19.5	18,031	8,529	1,597	557	2.76	25.7
811	1.4×10^{22}	816,135	50.9	7,682	25.6	16,658	10,893	2,304	1,702	8.68	129
1160	2×10^{22}	748,077	49.2	7,041	26.1	15,271	11,169	2,442	2,874	14.8	277

TABLE VI (Cont)

Composition of Irradiated Th-232 in Atoms per Million Initial Th-232 Atoms
Resonance Flux per ln E Interval Equal to 1/12 Thermal Flux

Time days	nvt_2 n/cm^2	Th-232	Pa-231	Pa-233	U-232	U-233	U-234	U-235	U-236	U-237	Np-237
For Thermal Flux = 1 x 10^{15} n/cm^2 sec											
11.6	1 x 10^{21}	985,592	11.3	11,954	1.03	1,493	515	26.8	0.78	0.0043	0.001
23.2	2 x 10^{21}	971,392	20.3		3.39						
46.3	4 x 10^{21}	943,603	33.3	28,627	9.46	9,287	6,241	806	108	1.50	1.26
81.1	7 x 10^{21}	903,402	44.3	32,854	17.5	13,011	12,626	2,151	573	10.0	14.9
116	1 x 10^{22}	864,914	49.2	33,180	22.6	13,968	17,308	3,331	1,411	27.4	56.9
232	2 x 10^{22}	748,077	49.2	29,418	26.2	12,751	22,255	4,831	5,218	114	406
463	4 x 10^{22}	559,618	37.7	22,021	20.6	9,552	18,318	4,080	10,232	233	1,127
811	7 x 10^{22}	362,085	24.4	14,248	13.3	6,180	11,914	2,657	10,856	251	1,337
1160	1 x 10^{23}	234,277	15.8	9,219	8.63	3,999	7,709	1,719	8,651	201	1,100
2320	2 x 10^{23}	54,886	3.70	2,160	2.02	937	1,806	403	2,483	57.7	324

TABLE VII

Composition of Irradiated U-233 in Atoms per Million Initial U-233 Atoms
Resonance Flux per ln E Interval Equal to 1/12 Thermal Flux

nvt n/cm^2	U-233	U-234	U-235	U-236	U-237 + Np-237*
1 x 10^{19}	993,125	727	0.58	0	0
2 x 10^{19}	986,295	1,448	2.31	0.002	0
3.5 x 10^{19}	976,139	2,517	7.01	0.010	0
4 x 10^{19}	972,777	2,871	9.16	0.015	0
5 x 10^{19}	966,088	3,573	14.2	0.026	0
7 x 10^{19}	952,848	4,961	27.6	0.073	0
8 x 10^{19}	946,296	5,645	35.8	0.108	0
1 x 10^{20}	933,327	6,997	55.4	0.211	0
1.4 x 10^{20}	907,919	9,632	106	0.565	0
2 x 10^{20}	871,099	13,417	210	1.61	0
3.5 x 10^{20}	785,449	22,050	595	8.31	0.023
4 x 10^{20}	758,813	24,681	758	11.9	0.039
5 x 10^{20}	708,220	29,599	1,124	22.3	0.091
7 x 10^{20}	616,930	38,169	1,987	56.6	0.325
8 x 10^{20}	575,797	41,879	2,465	81.3	0.534
1 x 10^{21}	501,576	48,286	3,477	147	1.22
1.4 x 10^{21}	380,602	57,672	5,565	347	3.96
2 x 10^{21}	251,579	65,365	8,429	809	13.7
3.5 x 10^{21}	89,367	66,367	12,624	2,561	78.4
4 x 10^{21}	63,292	63,910	13,118	3,237	114
5 x 10^{21}	31,746	57,516	13,165	4,594	204
8 x 10^{21}	4,006	37,744	10,002	7,939	562
1 x 10^{22}	1,008	27,670	7,572	9,344	808
2 x 10^{22}	1.02	5,614	1,570	10,326	1,442
4 x 10^{22}	0	229	64.0	6,113	1,011
7 x 10^{22}	0	1.88	0.53	2,362	397
1 x 10^{23}	0	0	0	904	152

* Values of U-237 + Np-237 vary slightly with flux, other values depend only on "nvt".

TABLE VIIb

U-237 and Np-237 in Irradiated U-233
in Atoms per 10^6 Initial U-233 Atoms

Flux 5 x 10^{13} n/cm^2 sec				Flux 1 x 10^{15} n/cm^2 sec			
Time days	"nvt" n/cm^2	U-237 ppm	Np-237 ppm	Time days	"nvt" n/cm^2	U-237 ppm	Np-237 ppm
81.1	3.5 x 10^{20}	0.008	0.016	11.6	1 x 10^{21}	0.975	0.247
116	5 x 10^{20}	0.024	0.067	23.2	2 x 10^{21}	9.04	4.80
232	1 x 10^{21}	0.176	1.04	46.3	4 x 10^{21}	53.8	61.5
463	2 x 10^{21}	1.03	12.7	81.1	7 x 10^{21}	142	300
811	3.5 x 10^{21}	3.34	75.1	116	1 x 10^{22}	204	617
1160	5 x 10^{21}	6.05	198	232	2 x 10^{22}	240	1202
2320	1 x 10^{22}	12.4	796	463	4 x 10^{22}	144	867
				811	7 x 10^{22}	55.6	341
				1160	1 x 10^{23}	21.3	131

TABLE VIII

Composition of Irradiated U-235 in Atoms per Million Initial U-235 Atoms
Resonance Flux per ln E Interval Equal to 1/12 Thermal Flux

Time days	nvt n/cm^2	U-235	U-236	U-237	Np-237	Np-238	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
0	0	1,000,000	0	0	0	0	0	0	0	0	0
For Thermal Flux = 5×10^{13} n/cm^2 sec											
11.6	5×10^{19}	964,062	5,494	3.08	1.33	0.002	0.004	0	0	0	0
23.2	1×10^{20}	929,415	10,782	9.01	8.35	0.018	0.034	0.002	0	0	0
46.3	2×10^{20}	863,812	20,769	22.2	44.9	0.112	0.463	0.012	0.002	0	0
81.1	3.5×10^{20}	773,987	34,380	40.9	155	0.414	3.17	0.127	0.005	0	0
116	5×10^{20}	693,503	46,500	57.5	324	0.887	9.95	0.568	0.026	0.003	0
232	1×10^{21}	480,946	78,010	101	1,202	3.39	77.1	8.33	0.721	0.099	0.006
463	2×10^{21}	231,309	113,071	149	3,717	10.6	463	85.6	14.1	3.39	0.533
811	3.5×10^{21}	77,150	130,703	174	7,657	22.0	1,520	393	101	35.5	11.1
1160	5×10^{21}	25,733	132,226	176	10,791	31.1	2,747	832	273	115	57.6
2320	1×10^{22}	662	116,078	155	15,223	43.9	5,391	1,960	875	457	617
For Thermal Flux = 2×10^{-4} n/cm^2 sec											
2.32	4×10^{19}	971,145	4,412	2.62	0.213	0	0	0	0	0	0
4.63	8×10^{19}	943,122	8,691	9.59	1.59	0.005	0.003	0	0	0	0
8.11	1.4×10^{20}	902,596	14,869	25.9	7.69	0.038	0.024	0.001	0	0	0
11.6	2×10^{20}	863,812	20,769	46.8	20.4	0.125	0.109	0.003	0	0	0
23.2	4×10^{20}	746,171	38,578	130	122	0.987	1.84	0.075	0.003	0	0
46.3	8×10^{20}	556,772	66,873	290	599	5.68	22.7	1.82	0.115	0.012	0
85.0	1.47×10^{21}	340,943	98,097	497	1,951	20.6	148	20.5	2.37	0.425	0.043
122	2.1×10^{21}	214,932	115,204	606	3,548	38.4	391	71.4	11.2	2.79	0.434
243	4.2×10^{21}	46,217	132,484	721	8,728	97.0	1,776	492	141	53.8	20.4
386	8.4×10^{21}	2,136	121,946	669	14,119	158	4,343	1,523	637	323	328
650	1.47×10^{22}	21.2	99,957	548	14,691	165	5,299	2,008	968	540	1,238
1215	2.1×10^{22}	0.21	81,710	448	12,771	144	4,785	1,844	918	522	1,888

TABLE VIII (Cont)

Composition of Irradiated U-235 in Atoms per Million Initial U-235 Atoms
Resonance Flux per ln E Interval Equal to 1/12 Thermal Flux

Time days	nvt_2 n/cm^2	U-235	U-236	U-237	Np-237	Np-238	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
For Thermal Flux = 1×10^{15} n/cm^2 sec											
2.32	2×10^{20}	863,812	20,769	62.2	5.06	0.005	0.007	0	0	0	0
4.63	4×10^{20}	746,171	38,578	216	36.0	0.567	0.183	0.005	0	0	0
8.11	7×10^{20}	599,056	60,607	541	163	3.61	2.10	0.130	0.004	0	0
11.6	1×10^{21}	480,946	78,010	908	403	10.6	8.97	0.781	0.053	0.005	0
23.2	2×10^{21}	231,309	113,071	1,986	1,919	63.4	110	17.4	2.42	0.517	0.069
46.3	4×10^{21}	53,504	132,216	2,916	6,196	230	768	195	49.7	17.5	5.56
81.1	7×10^{21}	5,952	126,938	2,968	10,921	424	2,131	695	258	120	84.3
116	1×10^{22}	662	116,078	2,733	12,911	508	3,042	1,089	473	247	297
232	2×10^{22}	0.4	86,367	1,988	11,788	468	3,254	1,251	619	352	1,154
463	4×10^{22}	0	44,486	1,048	6,426	255	1,811	702	354	203	1,244
811	7×10^{22}	0	17,033	401	2,462	97.8	694	269	136	78.0	564

TABLE IX

Composition of Irradiated U-236 in Atoms per Million Initial U-236 Atoms
Resonance Flux per ln E Interval Equal to 1/12 Thermal Flux
For Thermal Flux of 2×10^{14} n/cm^2 sec

time days	nvt_2 n/cm^2	U-236	U-237	Np-237	Np-238	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
11.6	2×10^{20}	993,620	3,668	2,580	18.6	21.4	0.516	0.008	0.	0
23.2	4×10^{20}	987,282	4,717	7,475	67.3	171	8.56	0.314	0.018	0
46.3	8×10^{20}	974,725	5,060	18,132	182	1,000	99.7	7.61	0.910	0.056
81.1	1.4×10^{21}	956,189	5,000	32,655	340	3,303	534	71.3	14.2	1.61
116	2×10^{21}	938,005	4,906	45,048	475	6,352	1,327	246	65.0	11.4
232	4×10^{21}	879,853	4,602	74,057	792	17,449	5,214	1,639	669	288
463	8×10^{21}	774,142	4,049	97,020	1,043	31,461	11,293	4,910	2,550	2,884
811	1.4×10^{22}	638,905	3,342	95,492	1,029	34,928	13,291	6,459	3,617	8,786
1,160	2×10^{22}	527,292	2,758	82,784	892	31,207	12,038	6,001	3,416	12,640
2,320	4×10^{22}	278,037	1,454	44,583	481	17,016	6,599	3,323	1,904	11,968

TABLE X

Composition of Irradiated U-238 in Atoms per Million Initial U-238 Atoms
Resonance Flux per ln E Interval Equal to 1/30 Thermal Flux

Time	nvt_2 n/cm^2	U-238	U-237	Np-237	Np-238	Np-239	Pu-238	Pu-239	Pu-240	Pu-241
0	0	1,000,000	0	0	0	0	0	0	0	0
For Thermal Flux = 5×10^{13} n/cm^2 sec										
11.6 days	5×10^{19}	999,395	0.102	0.072	0.0001	170	0.0001	423	3.63	0.031
23.2 d	1×10^{20}	998,790	0.132	0.214	0.0004	176	0.0011	978	17.2	0.303
46.3 d	2×10^{20}	997,582	0.144	0.541	0.0012	176	0.0072	1,999	72.8	2.59
81.1 d	3.5×10^{20}	995,773	0.145	1.04	0.0024	176	0.0270	3,309	215	13.2
116 d	5×10^{20}	993,967	0.145	1.52	0.0036	175	0.0586	4,394	409	35.0
232 d	1×10^{21}	987,969	0.144	3.02	0.0073	174	0.233	6,827	1,263	194
463 d	2×10^{21}	976,084	0.142	5.59	0.0137	172	1.76	8,772	3,033	751
811 d	3.5×10^{21}	958,522	0.140	8.57	0.0210	169	11.1	9,309	4,734	1,517
3.17 years	5×10^{21}	941,277	0.137	10.7	0.0264	166	30.8	9,250	5,478	1,929
6.34 y	1×10^{22}	886,003	0.129	14.2	0.0351	156	94.8	8,727	5,720	2,140
For Thermal Flux = 2×10^{14} n/cm^2 sec										
463 days	8×10^{21}	907,712	0.518	12.9	0.120	639	14.1	8,921	5,819	2,195
811 d	1.4×10^{22}	844,130	0.481	14.6	0.135	594	26.5	8,297	5,514	2,103
3.17 years	2×10^{22}	785,001	0.448	14.4	0.133	552	29.1	7,716	5,131	1,956
6.34 y	4×10^{22}	616,227	0.351	11.6	0.108	434	24.0	6,057	4,028	1,537
For Thermal Flux = 1×10^{15} n/cm^2 sec										
11.6 days	1×10^{21}	987,969	1.88	1.28	0.033	3,331	0.037	5,416	927	129
23.2 d	2×10^{21}	976,084	2.32	3.39	0.106	3,393	0.239	8,233	2,743	645
46.3 d	4×10^{21}	952,739	2.41	7.05	0.240	3,332	1.03	9,154	5,086	1,697
81.1 d	7×10^{21}	918,765	2.33	10.4	0.363	3,197	2.47	8,927	5,965	2,233
116 d	1×10^{22}	886,003	2.25	12.0	0.424	3,083	3.73	8,612	5,942	2,267
232 d	2×10^{22}	785,001	1.99	12.8	0.453	2,731	5.84	7,630	5,301	2,031
463 d	4×10^{22}	616,227	1.57	10.4	0.367	2,144	6.02	5,990	4,161	1,595
811 d	7×10^{22}	428,594	1.09	7.20	0.256	1,491	4.58	4,166	2,894	1,109
3.17 years	1×10^{23}	298,093	0.757	5.01	0.178	1,037	3.24	2,898	2,013	771
6.34 y	2×10^{23}	88,859	0.226	1.49	0.053	309	0.971	864	600	230

TABLE X (Cont)

Composition of Irradiated U-238 in Atoms per Million Initial U-238 Atoms
Resonance Flux per ln E Interval Equal to 1/30 Thermal Flux

Time	nvt_2 n/cm^2	Pu-242	Am-241	Am-243	Cm-242	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248
0	0	0	0	0	0	0	0	0	0	
For Thermal Flux = 5×10^{13} n/cm^2 sec										
11.6 days	5×10^{19}	0	0	0	0	0	0	0	0	0
23.2 d	1×10^{20}	0.003	0	0	0	0	0	0	0	0
46.3 d	2×10^{20}	0.049	0.004	0	0	0	0	0	0	0
81.1 d	3.5×10^{20}	0.469	0.038	0.001	0.002	0	0	0	0	0
116 d	5×10^{20}	1.84	0.145	0.011	0.009	0	0	0	0	0
232 d	1×10^{21}	22.4	1.62	0.273	0.186	0.006	0	0	0	0
463 d	2×10^{21}	201	12.3	5.27	2.71	0.347	0.0015	0	0	0
811 d	3.5×10^{21}	855	40.2	42.4	14.2	5.29	0.0314	0.0126	0	0
3.17 years	5×10^{21}	1,793	64.9	133	29.4	25.6	0.181	0.0764	0.0004	0
6.34 y	1×10^{22}	5,021	90.0	764	52.8	348	3.09	2.81	0.0218	0.0049
For Thermal Flux = 2×10^{14} n/cm^2 sec										
463 days	8×10^{21}	3,861	22.2	472	31.7	166	1.38	0.930	0.0062	0.0005
811 d	1.4×10^{22}	7,136	22.5	1,432	47.9	1,042	9.84	13.6	0.120	0.0455
3.17 years	2×10^{22}	9,187	21.0	2,321	49.1	2,692	26.6	58.6	0.564	0.316
6.34 y	4×10^{22}	11,052	16.5	3,617	39.6	10,130	105	575	6.12	8.33
For Thermal Flux = 1×10^{15} n/cm^2 sec										
11.6 days	1×10^{21}	13.3	0.049	0.149	0.006	0.012	0	0	0	0
23.2 d	2×10^{21}	155	0.488	3.78	0.137	0.239	0.0010	0	0	0
46.3 d	4×10^{21}	1,067	2.38	58.3	1.71	8.22	0.0509	0.034	0.0001	0
81.1 d	7×10^{21}	3,160	4.31	327	7.20	94.5	0.751	0.396	0.0024	0
116 d	1×10^{22}	5,164	4.75	769	13.5	356	3.14	2.80	0.0214	0.0058
232 d	2×10^{22}	9,456	4.36	2,376	27.3	2,784	27.4	59.5	0.571	0.309
463 d	4×10^{22}	11,444	3.42	3,741	33.0	10,983	113	610	6.49	8.71
811 d	7×10^{22}	9,382	2.38	3,315	26.4	19,436	204	2,294	25.3	71.7
3.17 years	1×10^{23}	6,783	1.66	2,436	18.8	20,951	221	3,986	44.6	207
6.34 y	2×10^{23}	2,049	0.493	739	5.66	10,752	114	5,113	58.0	767

TABLE XI

Composition of Irradiated Pu-239 in Atoms per Million Initial Pu-239 Atoms
Resonance Flux per ln E Interval Equal to 1/30 Thermal Flux

Time	nvt_2 n/cm^2	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Pu-244	Am-241	Am-242
0	0	0	1,000,000	0	0	0	0	0	0
For Thermal Flux = 5×10^{13} n/cm^2 sec									
11.6 days	5×10^{19}	0	939,883	18,151	265	1.79	--	0.142	0
23.2 d	1×10^{20}	0	883,380	34,683	1,001	13.7	--	1.15	0.0012
46.3 d	2×10^{20}	0.014	780,360	63,335	3,574	101	--	8.19	0.0158
81.1 d	3.5×10^{20}	0.154	647,912	96,762	9,245	475	--	37.5	0.108
116 d	5×10^{20}	0.730	537,944	120,774	15,950	1,221	--	93.6	0.333
232 d	1×10^{21}	14.5	289,385	154,890	36,710	6,499	--	447	2.16
463 d	2×10^{21}	184	83,771	130,686	50,262	24,251	--	1,303	7.57
811 d	3.5×10^{21}	773	13,234	66,569	33,514	47,024	--	1,616	10.2
3.17 years	5×10^{21}	1,226	2,443	29,600	16,401	57,046	--	1,153	7.49
6.34 y	1×10^{22}	603	324	2,140	1,137	51,969	--	118	0.791
12.7 y	2×10^{22}	10.0	6.76	994	359	29,790	--	--	--
22.2 y	3.5×10^{22}	0	0	1,259	469	12,670	--	--	--
For Thermal Flux = 2×10^{14} n/cm^2 sec									
11.6 days	2×10^{20}	0.0027	780,360	63,335	3,580	101	0	2.05	--
23.2 d	4×10^{20}	0.0198	608,962	105,710	11,453	--	0	13.3	--
46.3 d	8×10^{20}	0.401	370,834	147,860	29,451	--	0.047	70.6	--
81.1 d	1.4×10^{21}	4.05	176,224	152,920	47,080	--	0.31	206	--
116 d	2×10^{21}	15.1	83,743	130,682	50,936	24,480	0.93	329	1.91
232 d	4×10^{21}	107	7,041	51,100	27,459	52,365	5.1	381	2.43
463 d	8×10^{21}	219	138	5,268	3,151	57,673	17	83.8	0.557
3.17 years	2×10^{22}	24.1	12.6	294	109	30,293	43	0.10	0.0007
6.34 y	4×10^{22}	0.136	0.073	408	154	9,689	61	--	--
12.7 y	8×10^{22}	0	0	222	85.2	991	64	--	--
31.7 y	2×10^{23}	0	0	11.2	4.32	--	51	--	--

TABLE XI (Cont)

Composition of Irradiated Pu-239 in Atoms per Million Initial Pu-239 Atoms
Resonance Flux per ln E Interval Equal to 1/30 Thermal Flux

Time		nvt_2 n/cm^2	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Pu-244	Am-241	Am-242
For Thermal Flux = 1 x 10^{15} n/cm^2 sec										
11.5	days	1 x 10^{21}	0.043	289,385	154,890	37,063	6,500	--	22.5	--
23.2	d	2 x 10^{21}	0.654	83,743	130,682	51,119	25,300	--	66.0	--
46.3	d	4 x 10^{21}	5.20	7,014	51,101	27,612	52,561	--	76.6	0.49
81.1	d	7 x 10^{21}	13.3	174	9,306	5,597	--	--	26.9	--
115	d	1 x 10^{22}	15.9	10.6	1,618	984	53,077	--	6.09	0.041
232	d	2 x 10^{22}	10.0	4.42	64.9	24.7	30,430	--	0.03	0.00014
463	d	4 x 10^{22}	2.77	1.23	89.9	34.1	9,733	--	--	--
3.17	years	1 x 10^{23}	0.058	0.026	38.3	14.7	319	--	--	--
6.34	y	2 x 10^{23}	0	0	4.81	1.85	--	--	--	--
12.7	y	4 x 10^{23}	0	0	0.070	0.027	--	--	--	--

TABLE XI (Cont)

Composition of Irradiated Pu-239 in Atoms per Million Initial Pu-239 Atoms
Resonance Flux per ln E Interval Equal to 1/30 Thermal Flux

Time	Am-243	Cm-242	Cm-243	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248	Bk-249	Cf-249	Cf-250	Cf-251	Cf-252
0	0	0	0	0	0	0	0	0	0	0	0	0	0
For Thermal Flux = 5×10^{13} n/cm^2 sec													
11.6 days	0	0	0	0	0	0	0	0	0	0	0	0	0
23.2 d	0.019	0	0	0	0	0	0	0	0	0	0	0	0
46.3 d	0.291	0.227	0	0	0	0	0	0	0	0	0	0	0
81.1 d	2.48	1.98	0.0021	0.027	0	0	0	0	0	0	0	0	0
116 d	9.32	7.04	0.0107	0.217	0	0	0	0	0	0	0	0	0
232 d	106	67.4	0.205	4.02	0.0115	0.002	0	0	0	0	0	0	0
463 d	890	391	2.38	74.8	0.356	0.049	0	0	0	0	0	0	0
811 d	3,495	830	8.83	588	3.90	1.18	0.005	0	0	0	0	0	0
3.17 years	6,721	813	12.2	1,815	14.2	6.78	0.040	0.005	0.00001	0	0	0	0
6.34 y	13,664	135	3.65	9,827	93.1	115	0.969	0.274	0.00081	0.00028	0.00046	0.00014	0.00021
12.7 y	12,652	--	--	25,565	263	892	9.07	6.65	0.0239	0.0112	0.0176	0.0060	0.0181
22.2 y	6,206	--	--	29,867	316	2,620	28.5	46.7	0.181	0.096	0.148	0.053	0.233
For Thermal Flux = 2×10^{14} n/cm^2 sec													
11.6 days	0.291	0.064	0	0	0	0	0	0	0	0	0	0	0
23.2 d	--	0.850	--	--	0	0	0	0	0	0	0	0	0
46.3 d	--	9.63	--	--	--	--	0	0	0	0	0	0	0
81.1 d	--	54.2	--	--	--	--	0	0	0	0	0	0	0
116 d	897	137	0.789	75.8	0.359	0.053	0.0001	0	0	0	0	0	0
232 d	4,616	453	4.85	938	6.63	2.37	0.0120	0.0014	0	0	0	0	0
463 d	12,000	440	7.59	6,478	58.4	52.2	0.400	0.084	0.00027	0.00002	0.00013	0.00004	0.00004
3.17 years	12,863	26.7	0.577	29,364	301	982	9.95	7.16	0.0333	0.0039	0.0227	0.0077	0.0314
6.34 y	4,807	0.15	--	38,905	410	3,826	41.9	79.7	0.415	0.056	0.318	0.113	0.934
12.7 y	500	--	--	20,485	218	6,659	75.1	404	2.21	0.315	1.77	0.638	8.72
31.7 y	--	--	--	1,029	11.0	2,763	31.5	885	4.93	0.73	4.03	1.47	27.4

TABLE XI (Cont)

Composition of Irradiated Pu-239 in Atoms per Million Initial Pu-239 Atoms
Resonance Flux per ln E Interval Equal to 1/30 Thermal Flux

Time	Am-243	Cm-242	Cm-243	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248	Bk-249	Cf-249	Cf-250	Cf-251	Cf-252
For Thermal Flux = 1 x 10^{15} n/cm^2 sec													
11.6 days	106	4.14	0.014	4.0	0.012	0.002	0	0	0	0	0	0	0
23.2 d	89C	30.3	0.19	76	0.36	0.054	0.0001	0	0	0	0	0	0
46.3 d	4,63C	116	1.19	944	6.67	2.39	0.0121	0.0017	0	0	0	0	0
81.1 d	--	177	--	--	--	--	--	--	--	--	--	--	--
116 d	13,935	169	2.73	10,655	101	122	1.026	0.287	0.0011	0.00019	0.00058	0.00018	0.00035
232 d	12,920	92.7	1.57	30,501	312	1,009	10.21	7.31	0.0368	0.00085	0.0245	0.0084	0.0373
463 d	4,828	25.6	0.43	42,633	449	4,051	44.3	83.1	0.474	0.0127	0.354	0.126	1.28
3.17 years	161	0.54	0.01	17,704	188	7,611	86.2	634	3.84	0.111	3.03	1.10	26.3
6.34 y	--	--	--	2,216	23.6	3,697	42.2	1,064	6.54	0.192	5.21	1.90	67.6
12.7 y	--	--	--	32.0	0.34	352	4.03	556	3.44	0.101	2.75	1.00	42.4

TABLE XIb

Composition of Irradiated Pu-239 in Atoms per Million Initial Pu-239 Atoms
Resonance Flux per ln E Interval Equal to 1/12 Thermal Flux
For Thermal Flux of 2×10^{14} n/cm^2 sec

Time	Thermal nvt_2 n/cm^2	Pu-239	Pu-240	Pu-241	Pu-242	Am-243	Cm-244	Cm-245
0	0	1,000,000	0	0	0	0	0	0
11.6 days	2×10^{20}	748,264	75,987	7,292	247	.21	0.011	0
23.2 d	4×10^{20}	559,898	119,113	22,057	1,600	16.2	0.302	0.00065
46.3 d	8×10^{20}	313,486	146,719	50,614	8,483	183	7.27	0.0288
81.1 d	1.4×10^{21}	131,336	124,708	67,869	25,184	1,053	78.8	0.488
116 d	2×10^{21}	55,023	87,057	61,211	42,012	2,740	317	2.51
232 d	4×10^{21}	3,028	16,700	17,203	66,354	10,838	3,152	36.2
463 d	8×10^{21}	---	437	486	52,287	19,647	16,066	227
3.17 years	2×10^{22}	---	256	142	16,800	11,319	43,790	683
6.34 y	4×10^{22}	---	207	118	2,513	1,892	33,682	537
12.7 y	8×10^{22}	---	---	---	---	---	7,879	126

TABLE XII

Composition of Irradiated Pu-242 in Atoms per Million Initial Pu-242 Atoms
For Napkin Ring Pile Flux

Time		nvt n/cm^2	Pu-242	Pu-244	Am-243	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248	Bk-249	Cf-249	Cf-250	Cf-251	Cf-252	Pu-240	Pu-241
0		0	1,000,000	0	0		0	0	0	0	0	0	0	0	0	0	0
For Thermal Flux = 2×10^{14} n/cm^2 sec																	
11.6	days	2×10^{20}	988,665	10.3	10,851	181	0.22	0	0	0	0	0	0	0	0	0.07	0.002
23.2	d	4×10^{20}	977,458	20.8	21,500	730	1.63	0.059	0	0	0	0	0	0	0	0.54	0.026
46.3	d	8×10^{20}	955,424	41.3	41,371	2,861	11.0	1.06	0.002	0	0	0	0	0	0	4.09	0.392
116	d	2×10^{21}	892,258	100	90,774	16,310	109	28.4	0.114	0.008	0	0	0	0	0	51.0	9.48
232	d	4×10^{21}	796,124	190	145,791	55,571	476	272	1.72	0.218	0.0005	0	0.0002	0	0	284	74.9
463	d	8×10^{21}	633,814	339	190,048	162,369	1,592	2,040	17.3	4.88	0.0176	0.0015	0.0093	0.0028	0.0050	1,186	387
3.17	years	2×10^{22}	319,819	622	144,388	425,428	4,516	18,011	186	160	0.764	0.0918	0.537	0.184	0.857	4,017	1,479
6.34	y	4×10^{22}	102,284	799	51,000	485,022	5,265	55,305	609	1,284	6.73	0.912	5.19	1.85	16.3	4,945	1,879
12.7	y	8×10^{22}	~11,000	825	5,273	242,648	2,655	87,251	985	5,633	30.8	4.40	24.7	8.93	124	2,542	976
22.2	y	1.4×10^{23}		749		56,284	617	64,213	731	10,613	58.9	8.58	47.9	17.4	302	593	228
For Thermal Flux = 1×10^{15} n/cm^2 sec																	
11.6	days	1×10^{21}	944,594	251	49,504	4,226	18.8	2.25	0.006	0	0	0	0	0	0	1.45	0.164
23.2	d	2×10^{21}	892,253	494	89,702	15,995	106	27.4	0.110	0.007	0	0	0	0	0	9.92	1.84
46.3	d	4×10^{21}	796,124	940	144,822	55,253	472	268	1.69	0.210	0.0004	0	0.0002	0	0	56.2	14.8
116	d	1×10^{22}	565,525	1,998	192,230	220,443	2,218	3,683	33.4	12.2	0.0515	0.0010	0.0292	0.0092	0.0201	349	119
232	d	2×10^{22}	319,819	3,097	143,981	441,160	4,676	18,385	190	162	0.840	0.0201	0.577	0.198	1.02	830	306
463	d	4×10^{22}	102,284	3,979	50,869	532,362	5,769	58,337	641	1,331	7.65	0.207	5.75	2.05	22.4	1,080	411
3.17	years	1×10^{23}	~3,500	3,998	1,682	209,598	2,290	97,865	1,109	8,573	52.0	1.50	41.0	14.8	363	437	168
6.34	y	2×10^{23}		3,353		26,117	286	46,349	529	13,762	84.7	2.48	67.5	24.5	881	54.6	21
12.7	y	4×10^{23}		2,339		377	4.13	4,382	50.1	7,083	43.8	1.29	35.0	12.8	541	0.79	0.30

TABLE XIII

Composition of Irradiated Cm-244 in Atoms per Million Initial Cm-244 Atoms
For Napkin Ring Pile Flux

Time	nvt_2 n/cm^2	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248	Bk-249	Cf-249	Cf-250	Cf-251	Cf-252
0	0	1,000,000	0	0	0	0	0	0	0	0	0
For Thermal Flux = 2×10^{14} n/cm^2 sec											
11.6 days	2×10^{20}	994,823	3,334	159	0.134	0	0	0	0	0	0
23.2 d	4×10^{20}	989,674	5,620	568	0.930	0.014	0	0	0	0	0
34.7 d	6×10^{20}	984,551	7,182	1,146	2.73	0.067	0.00003	0	0	0	0
46.3 d	8×10^{20}	979,454	8,243	1,840	5.65	0.191	0.00014	0	0.00002	0	0
116 d	2×10^{21}	949,424	10,139	6,941	42.8	4.12	0.0082	0.00032	0.0022	0.00044	0.00027
232 d	4×10^{21}	901,406	9,877	15,711	136	30.4	0.0968	0.0067	0.0432	0.0119	0.0125
463 d	8×10^{21}	812,532	8,909	31,372	318	166	0.711	0.0729	0.442	0.144	0.354
3.17 years	2×10^{22}	595,115	6,525	64,962	713	1,084	5.53	0.723	4.15	1.46	9.51
6.34 y	4×10^{22}	354,162	3,883	89,982	1,011	3,489	18.8	2.63	14.8	5.32	58.8
12.7 y	8×10^{22}	125,431	1,375	85,930	976	8,256	45.5	6.57	36.8	13.3	206
For Thermal Flux = 1×10^{15} n/cm^2 sec											
11.6 days	1×10^{21}	979,033	8,987	2,615	9.69	0.412	0.0004	0	0	0	0
23.2 d	2×10^{21}	958,505	10,212	6,968	43.0	4.12	0.0084	0.00006	0.0022	0.0004	0
34.7 d	3×10^{21}	938,408	10,220	11,460	88.1	13.8	0.0384	0.00042	0.0139	0.0034	0.0015
46.3 d	4×10^{21}	918,733	10,041	15,843	137	30.5	0.102	0.00140	0.0449	0.0124	0.0123
116 d	1×10^{22}	809,046	8,848	39,139	407	274	1.35	0.0301	0.879	0.295	1.00
232 d	2×10^{22}	654,555	7,158	67,935	744	1,115	6.22	0.162	4.55	1.60	12.0
463 d	4×10^{22}	428,442	4,686	98,201	1,101	3,689	21.8	0.610	16.8	6.03	87.6
3.17 years	1×10^{23}	120,152	1,314	90,833	1,033	11,207	68.4	1.99	54.1	19.6	547
6.34 y	2×10^{23}	14,436	158	35,756	408	13,666	84.2	2.47	67.2	24.4	915
12.7 y	4×10^{23}	208	2.28	3,175	36.3	6,304	39.0	1.15	31.2	11.4	486

TABLE XIV

Comparison of Measured and Calculated Yields for Napkin Rings
Yields in atoms per 10^6 atoms Pu-239 Initially Present

Napkin Ring	KAPL 52		ANL 55		KAPL 54		ANL 64		ANL 17		KAPL 63	
Reported nvt	3.7×10^{21}		4×10^{21}		7.5×10^{21}		1.1×10^{22}		1.4×10^{22}		1.46×10^{22}	
Thermal nvt assumed for comparison	2.6×10^{21}		3.9×10^{21}		5.3×10^{21}		9.6×10^{21}		1.25×10^{22}		1.65×10^{22}	
Calculated values for Flux of	5×10^{13}		5×10^{13}		2×10^{14}		2×10^{14}		2×10^{14}		2×10^{14}	
	Measured	Calculated	Measured	Calculated	Measured	Calculated	Measured	Calculated	Measured	Calculated	Measured	Calculated
Pu-238	170	410	---	1,200	287	170	123	200	55	140	34	60
Pu-239	38,500	40,000	7,560	7,800	1,640	1,500	50	84	24	48	25	25
Pu-240	111,000	103,000	55,700	56,000	25,700	25,000	1,150	2,100	223	700	302	330
Pu-241	46,600	45,000	30,800	29,000	16,400	14,500	750	1,200	109	360	98	135
Pu-242	35,700	34,500	48,700	50,000	54,800	58,000	54,800	54,000	34,900	46,000	44,500	37,000
Pu-244	---	0.5	1.6	1.2	9	9.0	21	22	18.2	29	34	38
Am-241	1,460	1,500	1,970	1,520	172	255	10	38	---	12	12	6
Am-243	1,800	3,300	12,300	4,400	7,760	7,500	20,000	13,500	17,600	14,800	11,500	14,000
Cm-242	549	610	240	860	180	500	158	330	92	170	40	67
Cm-243	3.8	4.9	---	10	16	7.0	---	6.2	---	3.5	14	1.4
Cm-244	204	205	1,180	880	2,780	2,250	8,160	9,700	13,000	15,700	25,000	24,000
Cm-245	2.0	1.2	13.3	6.0	34.8	17	108	90	154	157	256	250
Cm-246	0.36	0.23	3.4	2.0	14.4	8.5	117	110	252	260	767	600
Cm-247	---	---	---	0.01	0.037	0.05	1.37	0.86	3.76	2.4	8.3	6.0
Cm-248	---	---	---	---	---	0.008	---	0.23	---	0.84	3.0	2.7
Bk-249	---	---	---	---	7.8×10^{-6}	$\sim 1.8 \times 10^{-5}$	---	8×10^{-4}	6.3×10^{-4}	3.5×10^{-3}	9.2×10^{-3}	1.5×10^{-2}
Cf-249	---	---	---	---	---	---	4.5×10^{-5}	6.0×10^{-5}	---	3.5×10^{-4}	8.9×10^{-4}	1.5×10^{-3}
Cf-250	---	---	---	---	3.7×10^{-6}	$\sim 4.5 \times 10^{-6}$	5.2×10^{-4}	3.6×10^{-4}	1.4×10^{-3}	1.8×10^{-3}	8.1×10^{-3}	9.0×10^{-3}
Cf-251	---	---	---	---	---	$\sim 1 \times 10^{-6}$	1.2×10^{-4}	1.2×10^{-4}	---	7.8×10^{-4}	2.2×10^{-3}	3.2×10^{-3}
Cf-252	---	---	---	---	1.2×10^{-6}	$\sim 1 \times 10^{-6}$	3.8×10^{-4}	2.0×10^{-4}	9.8×10^{-4}	1.6×10^{-3}	1.32×10^{-2}	1.0×10^{-2}

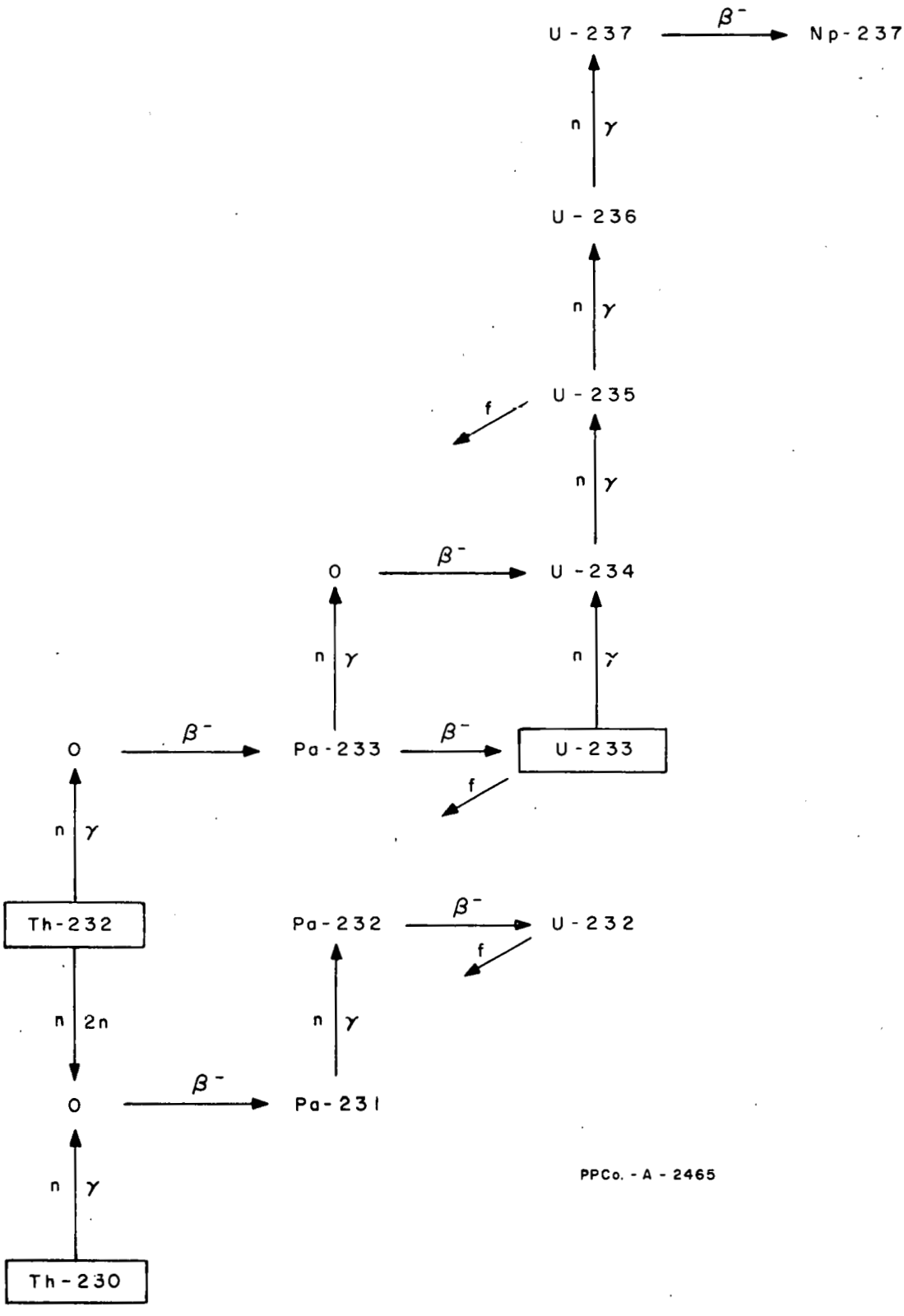


FIG. I
PRODUCTION OF HEAVY NUCLIDES FROM Th-230
Th-232 AND U-233

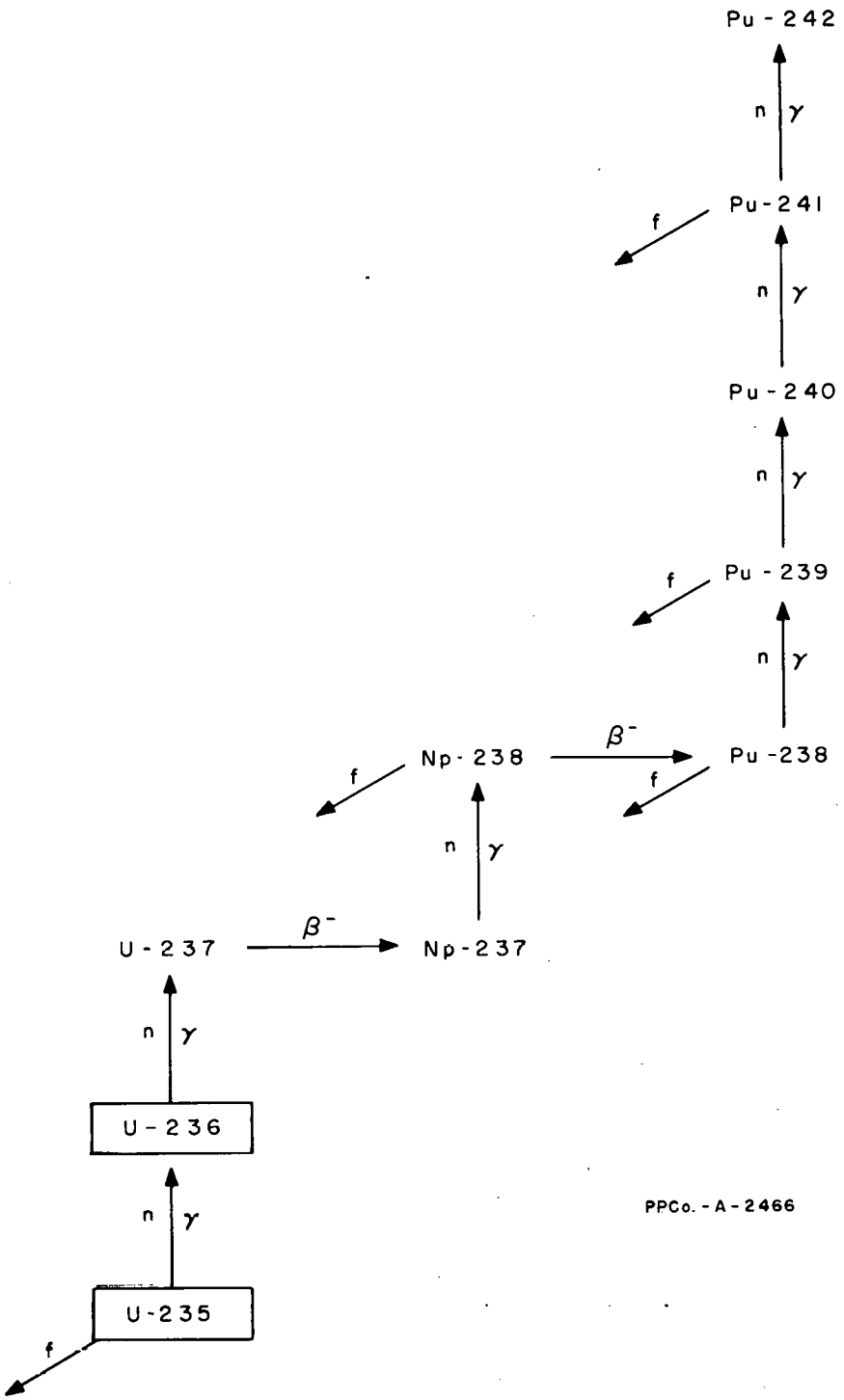


FIG. 2
PRODUCTION OF HEAVY NUCLIDES FROM U-235 AND U-236

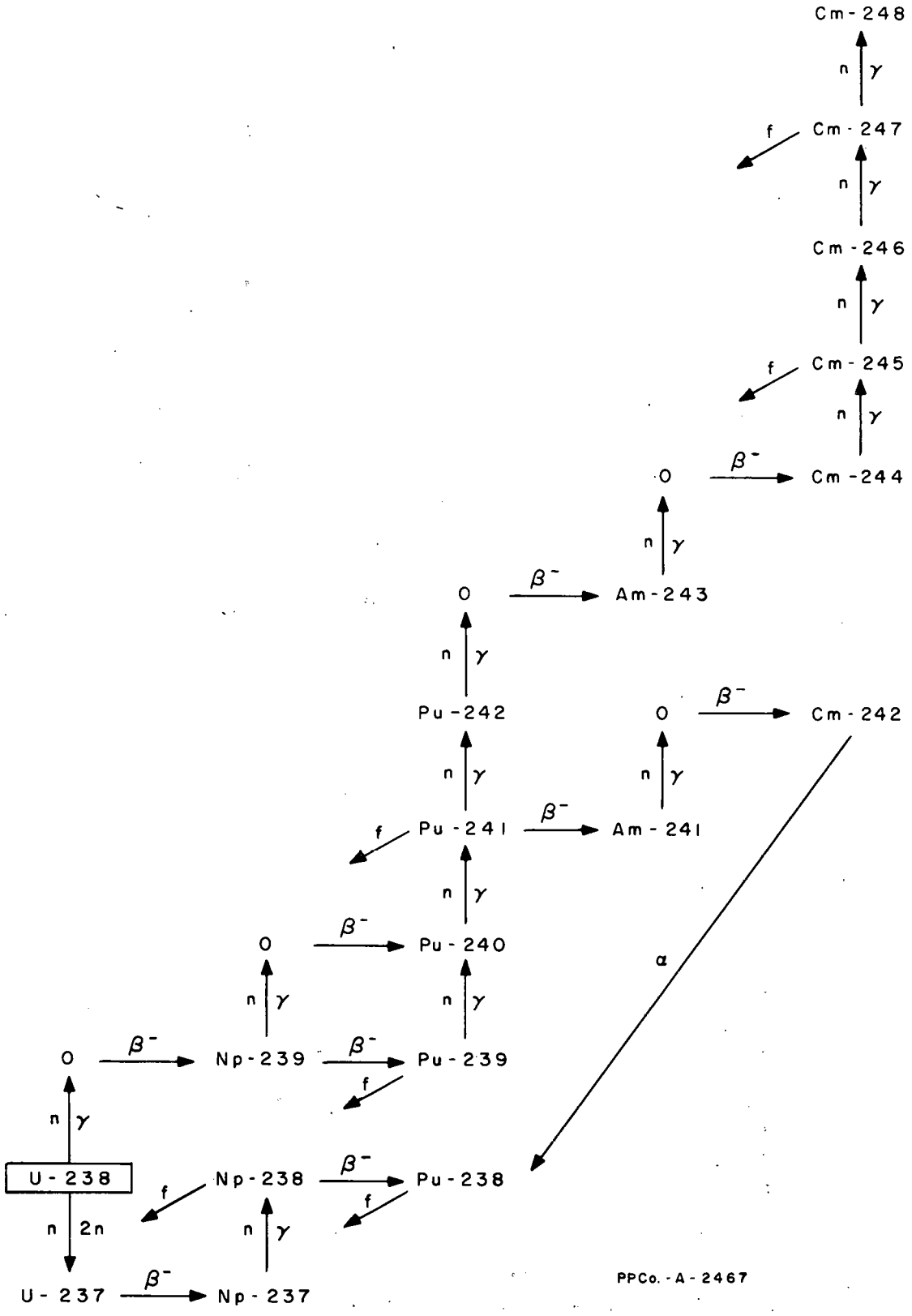


FIG. 3
PRODUCTION OF HEAVY NUCLIDES FROM U-238

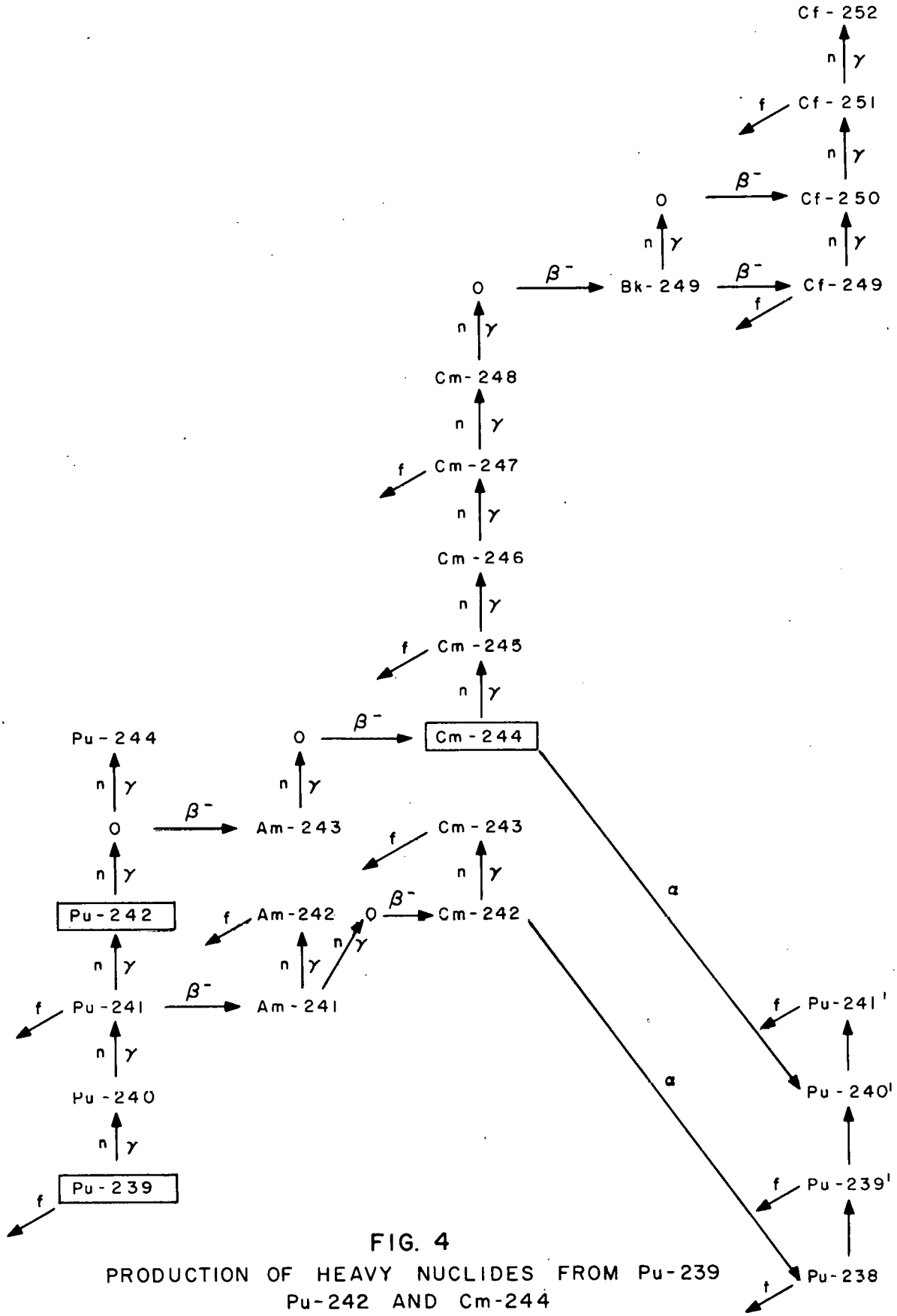


FIG. 4

PRODUCTION OF HEAVY NUCLIDES FROM Pu-239
Pu-242 AND Cm-244

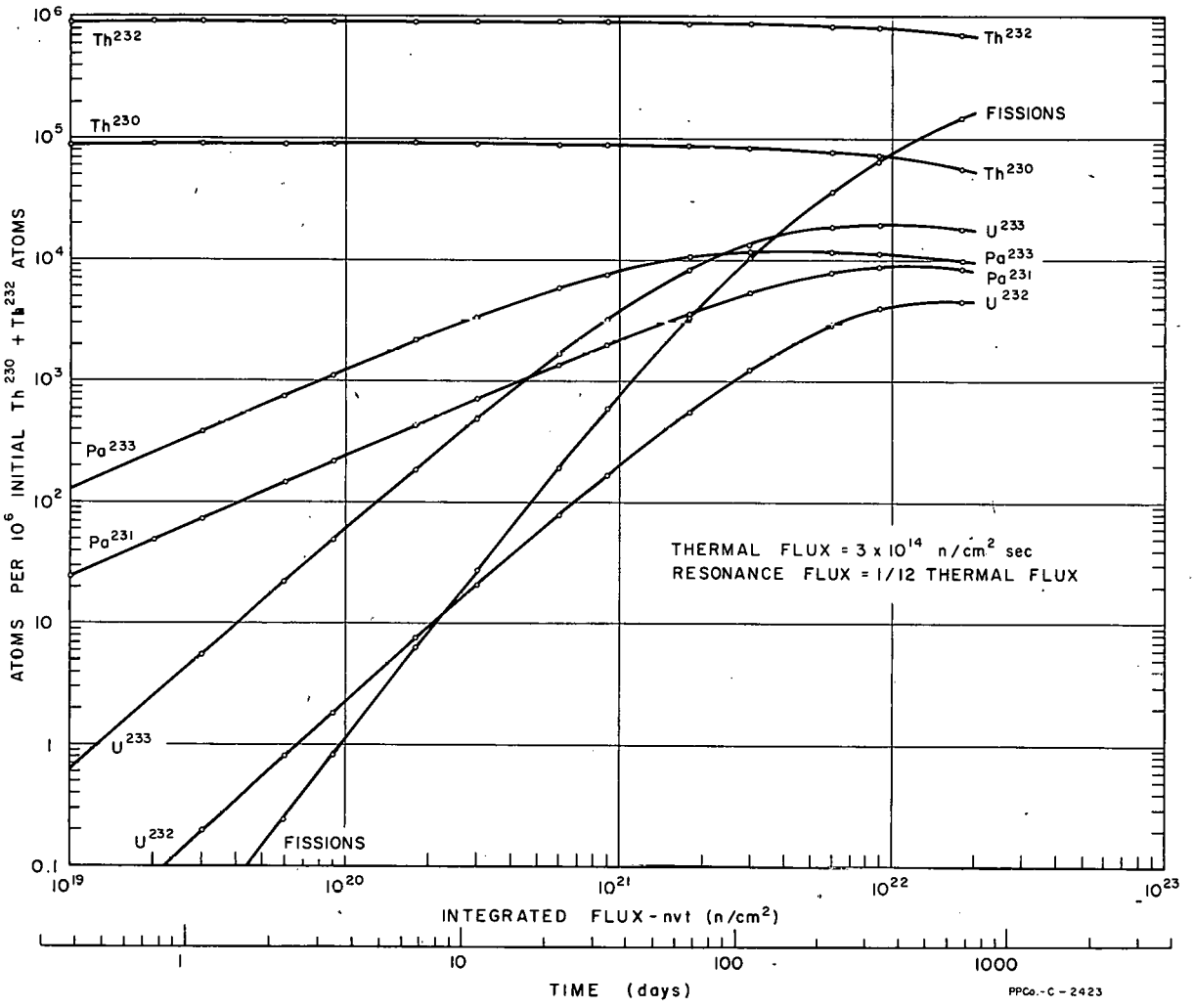


Fig. 5 Composition of pile irradiated ionium; sample initially contains 91% Th-232 plus 9% Th-230.

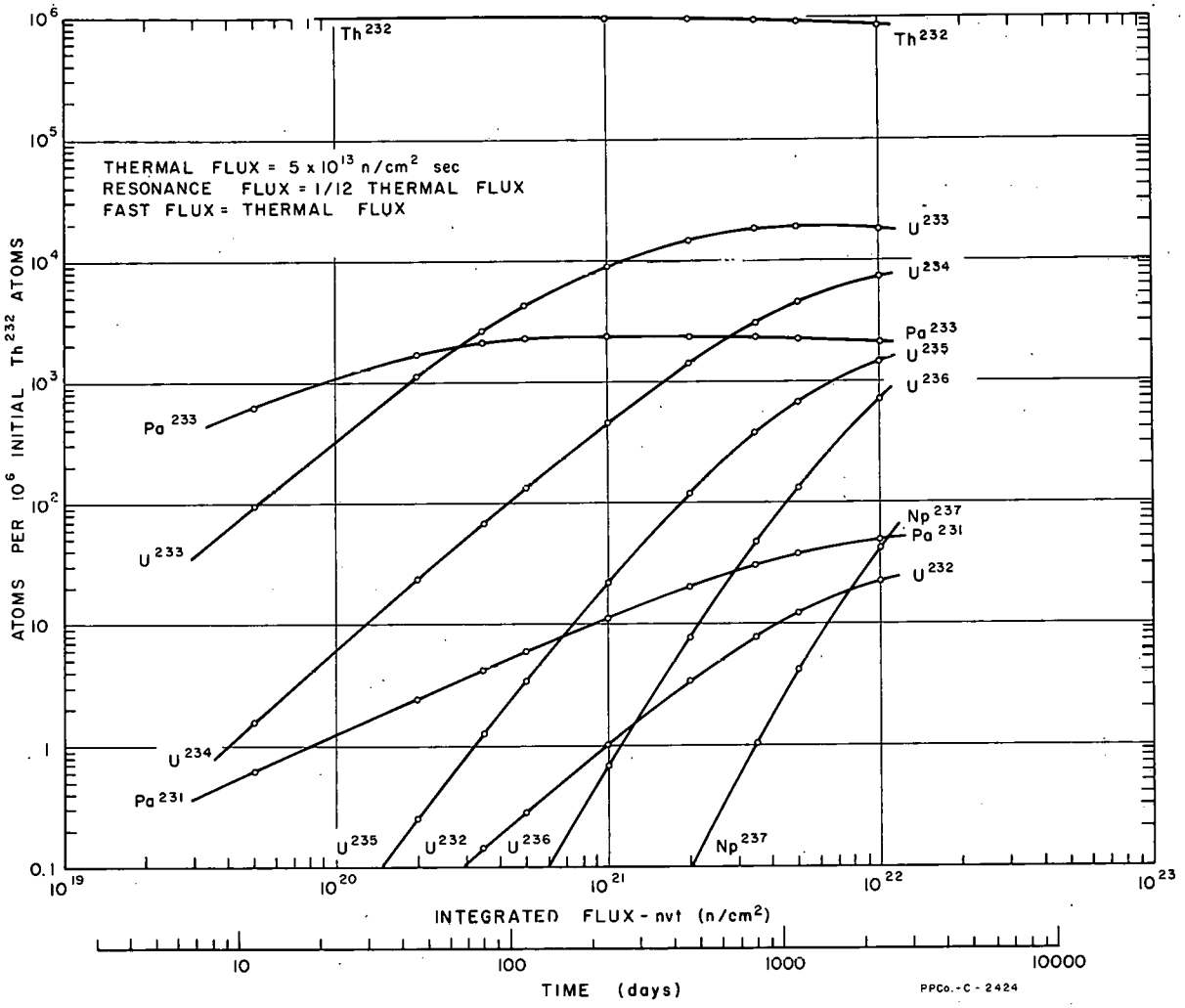


Fig. 6-1 Composition of pile irradiated thorium.

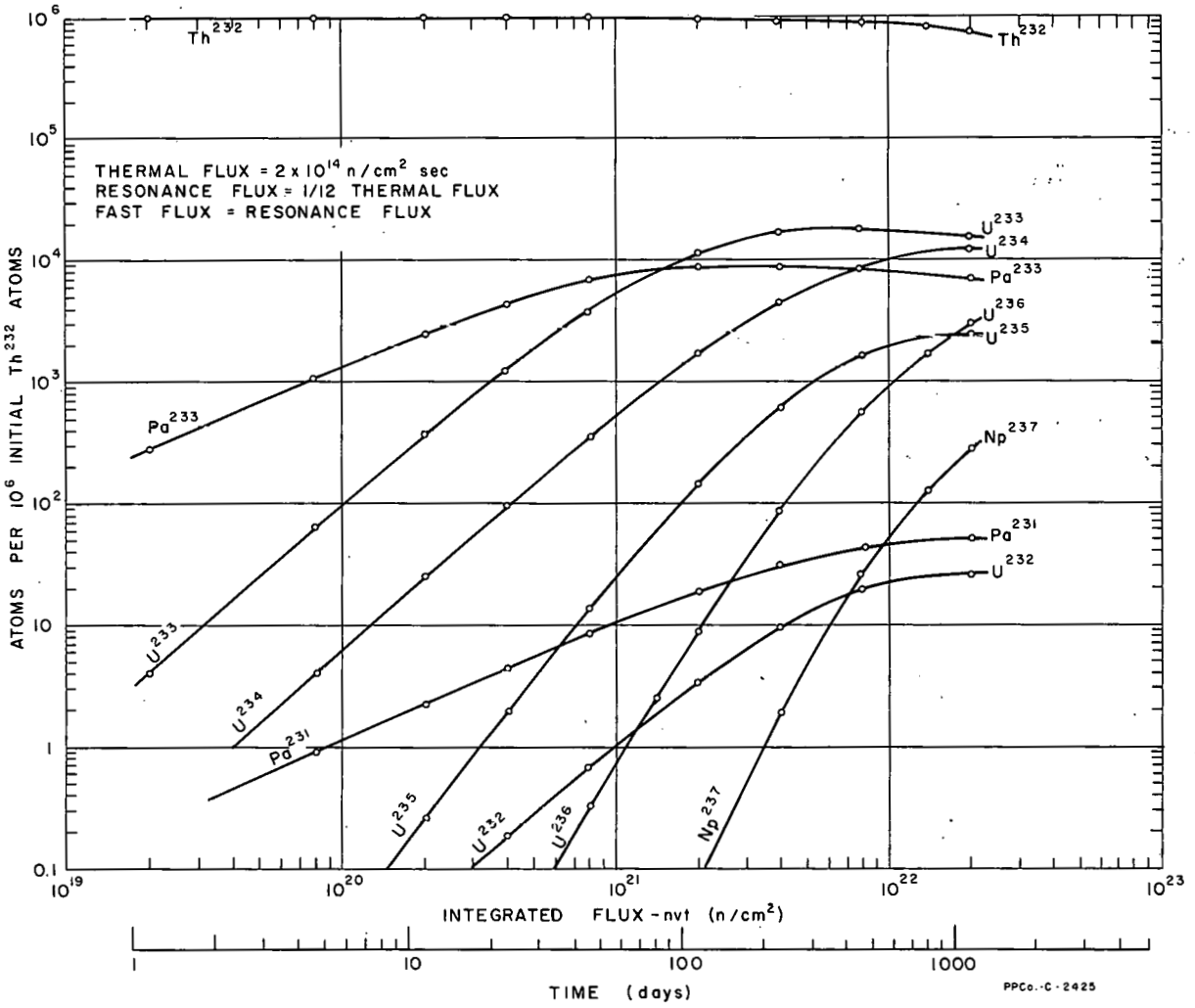


Fig. 6-2 Composition of pile irradiated thorium.

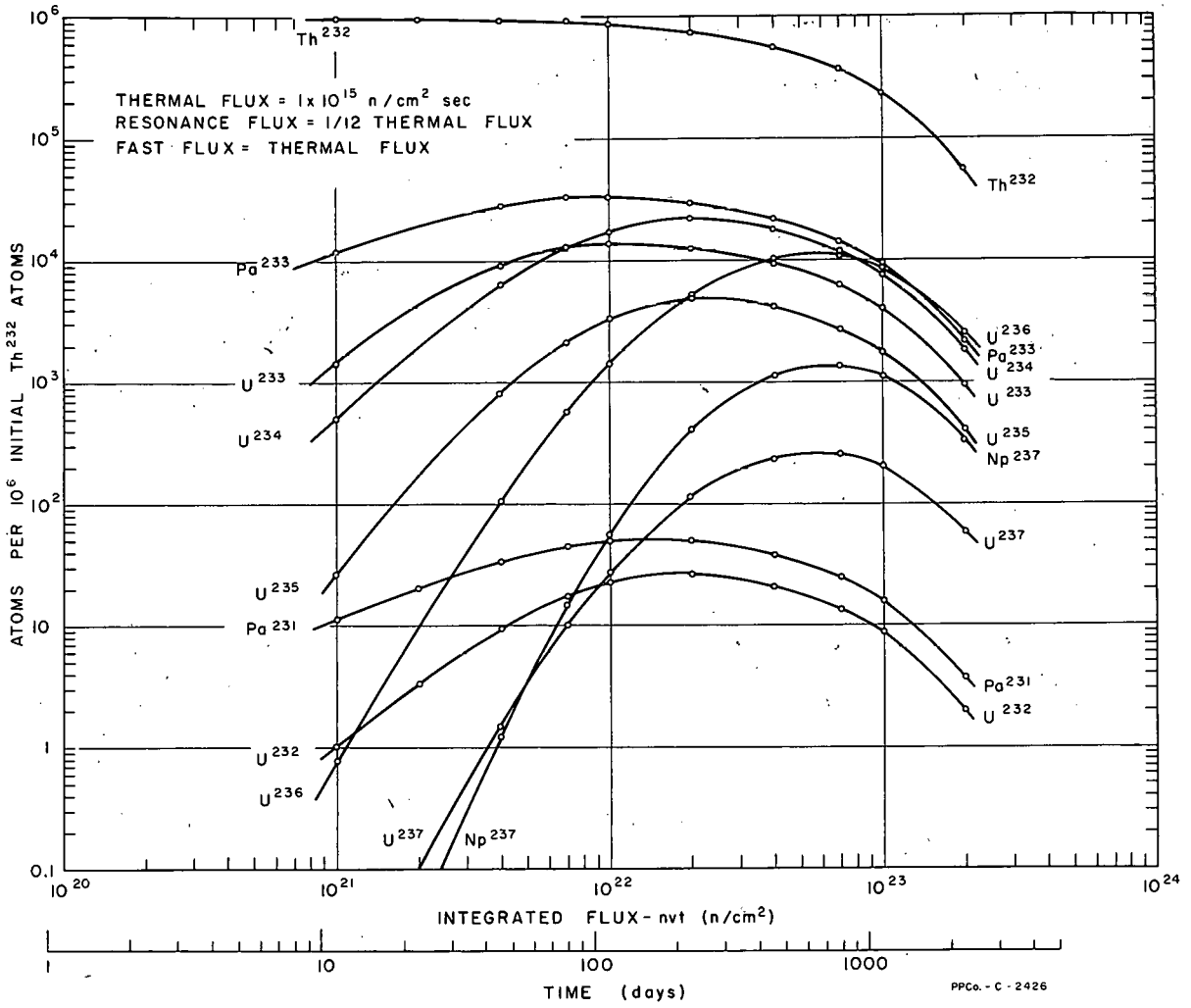


Fig. 6-3 Composition of pile irradiated thorium.

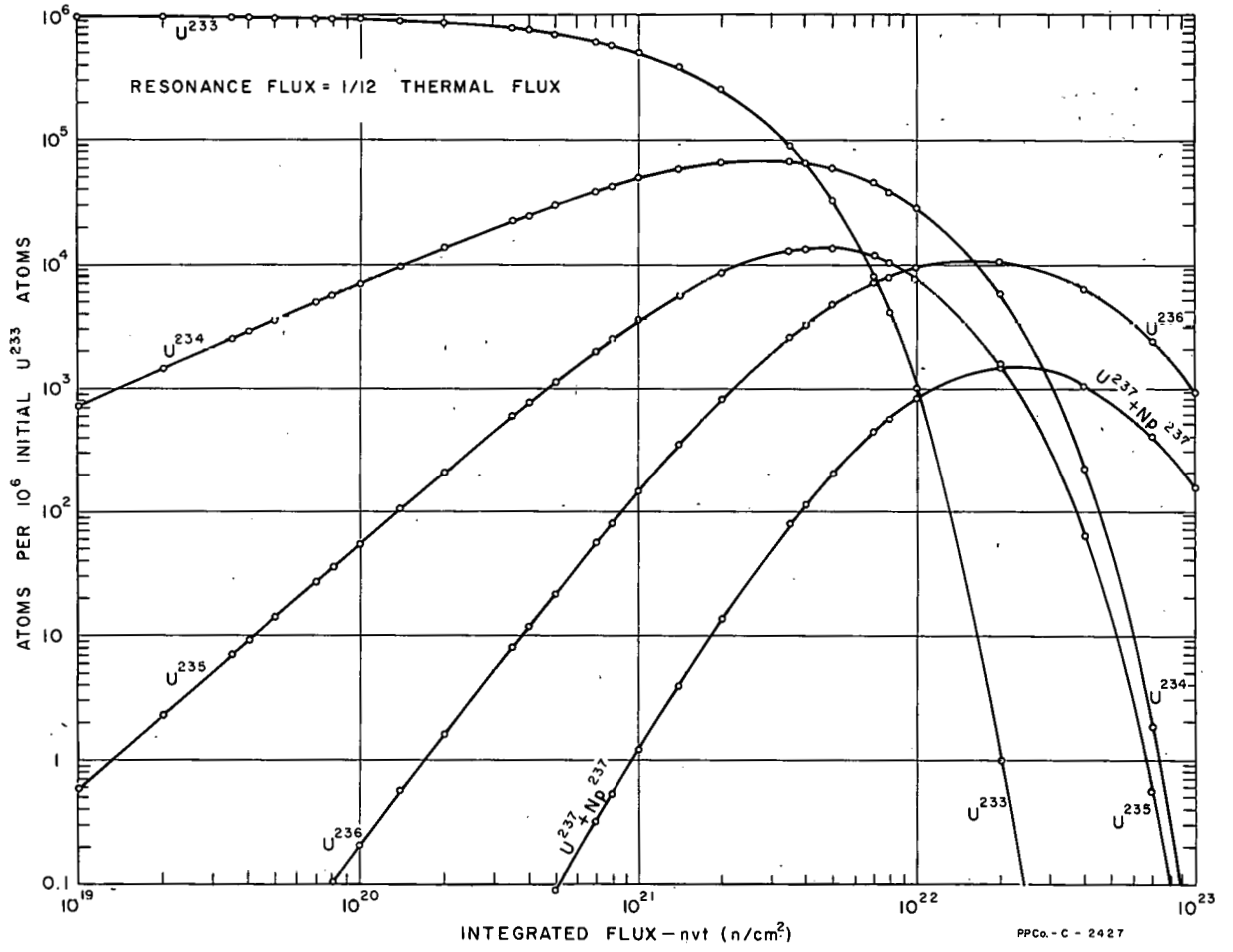


Fig. 7 Composition of pile irradiated U-233. (The compositions shown are dependent only upon "nvt", and will be representative of any reasonable reactor flux as long as $\phi_{res}/\phi_{thermal}$ equals 1/12.)

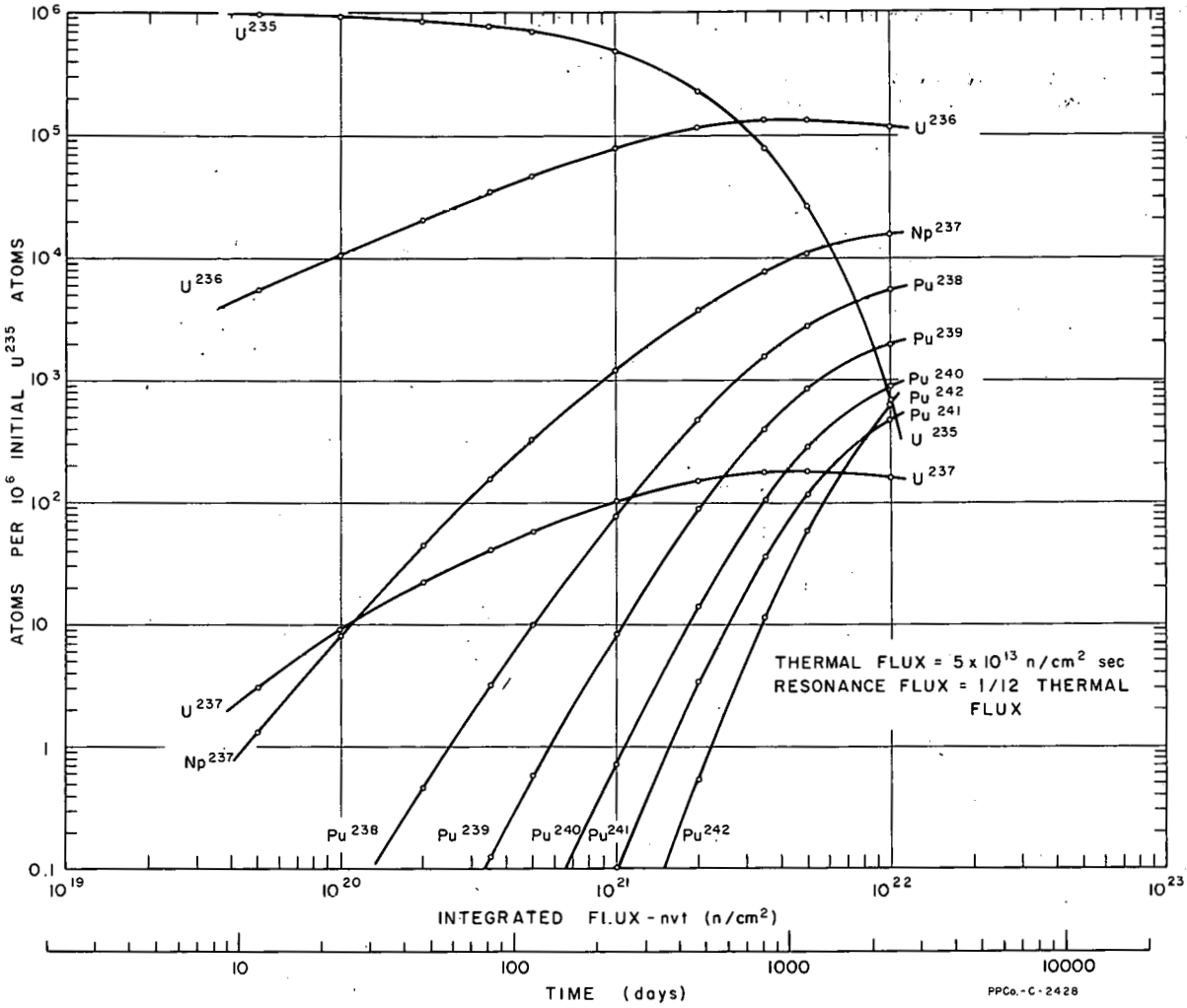


Fig. 8-1 Composition of pile irradiated U-235.

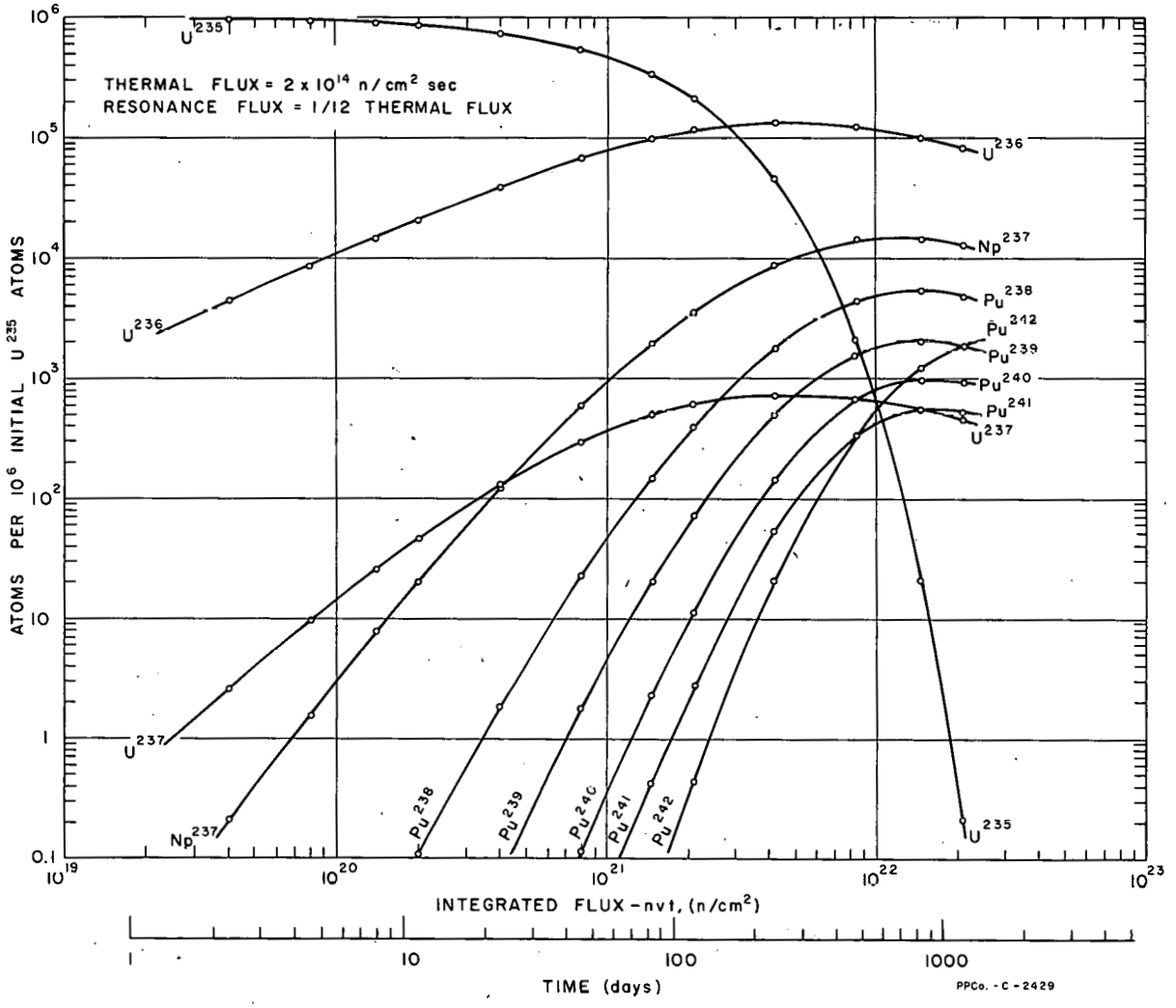


Fig. 8-2 Composition of pile irradiated U-235:

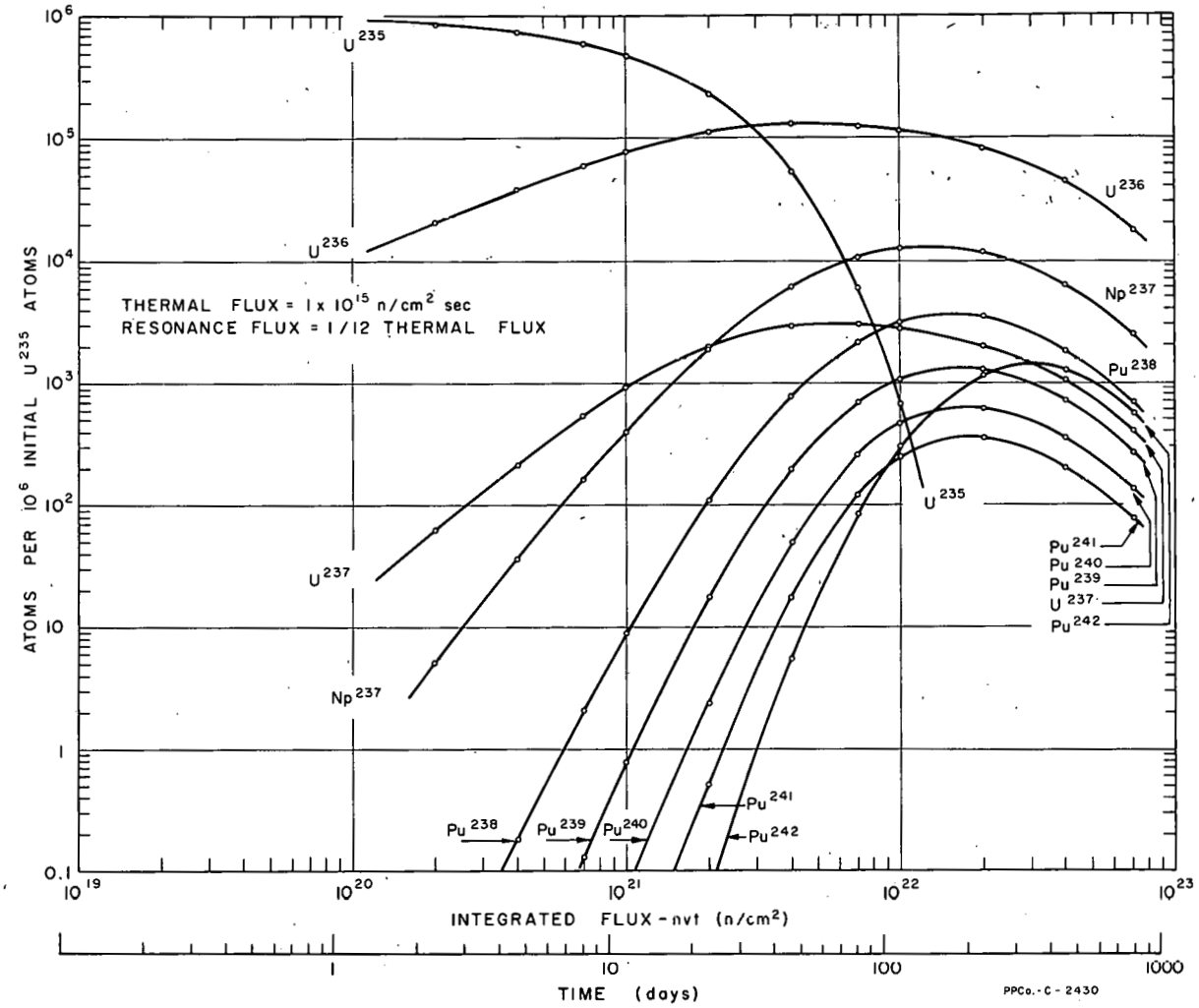


Fig. 8-3 Composition of pile irradiated U-235.

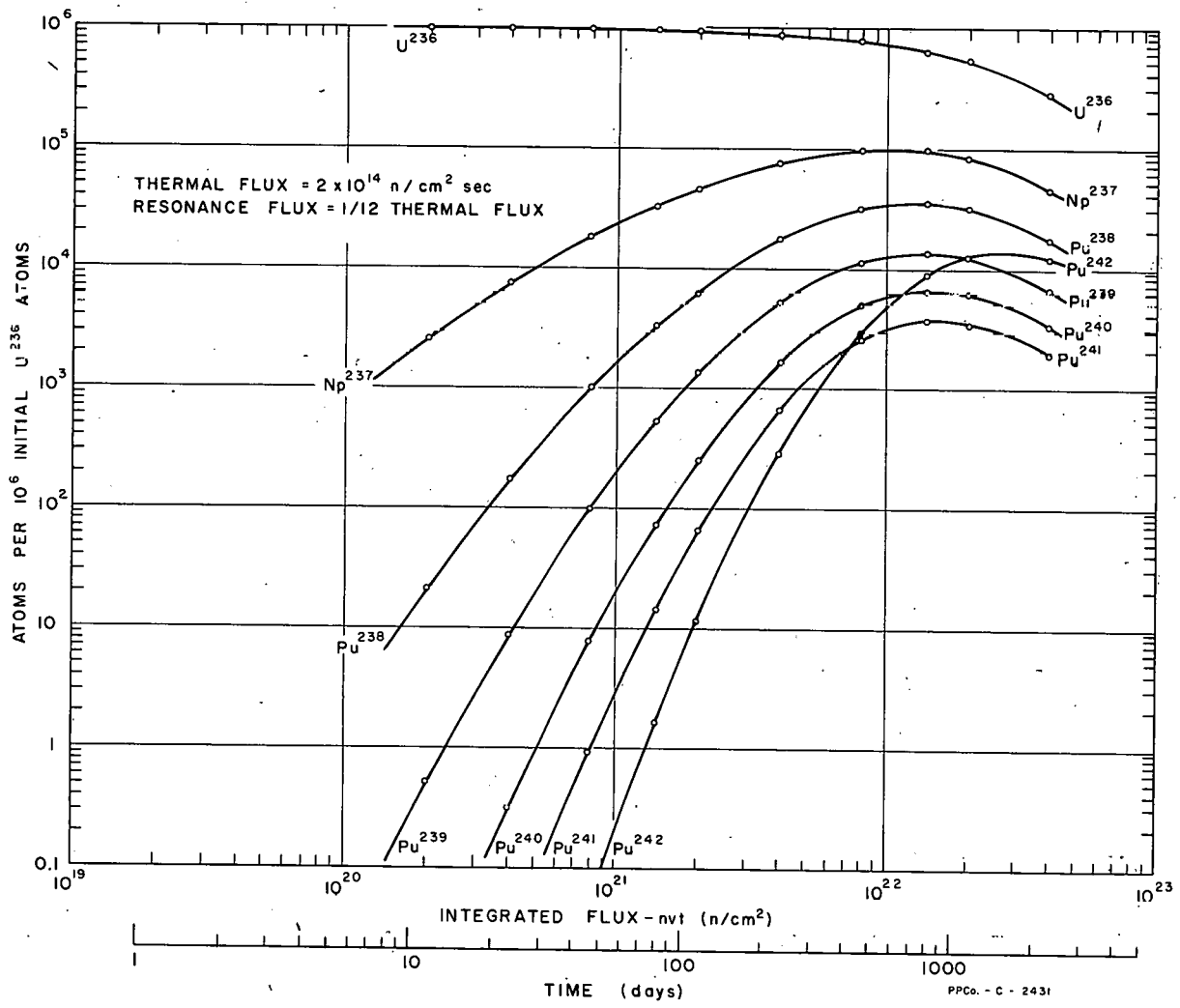


Fig. 9 Composition of pile irradiated U-236.

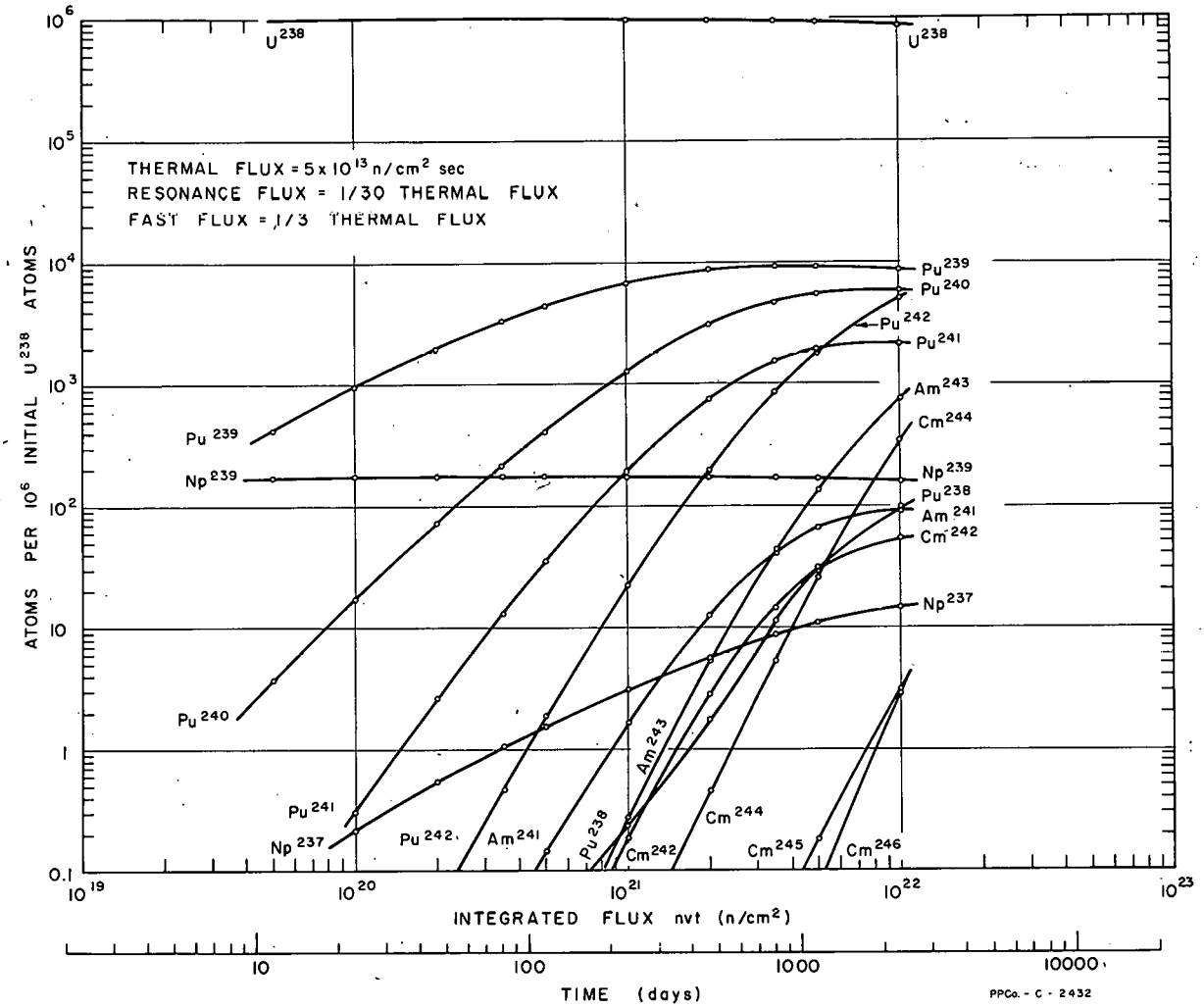


Fig. 10 1 Composition of pile irradiated U-238.

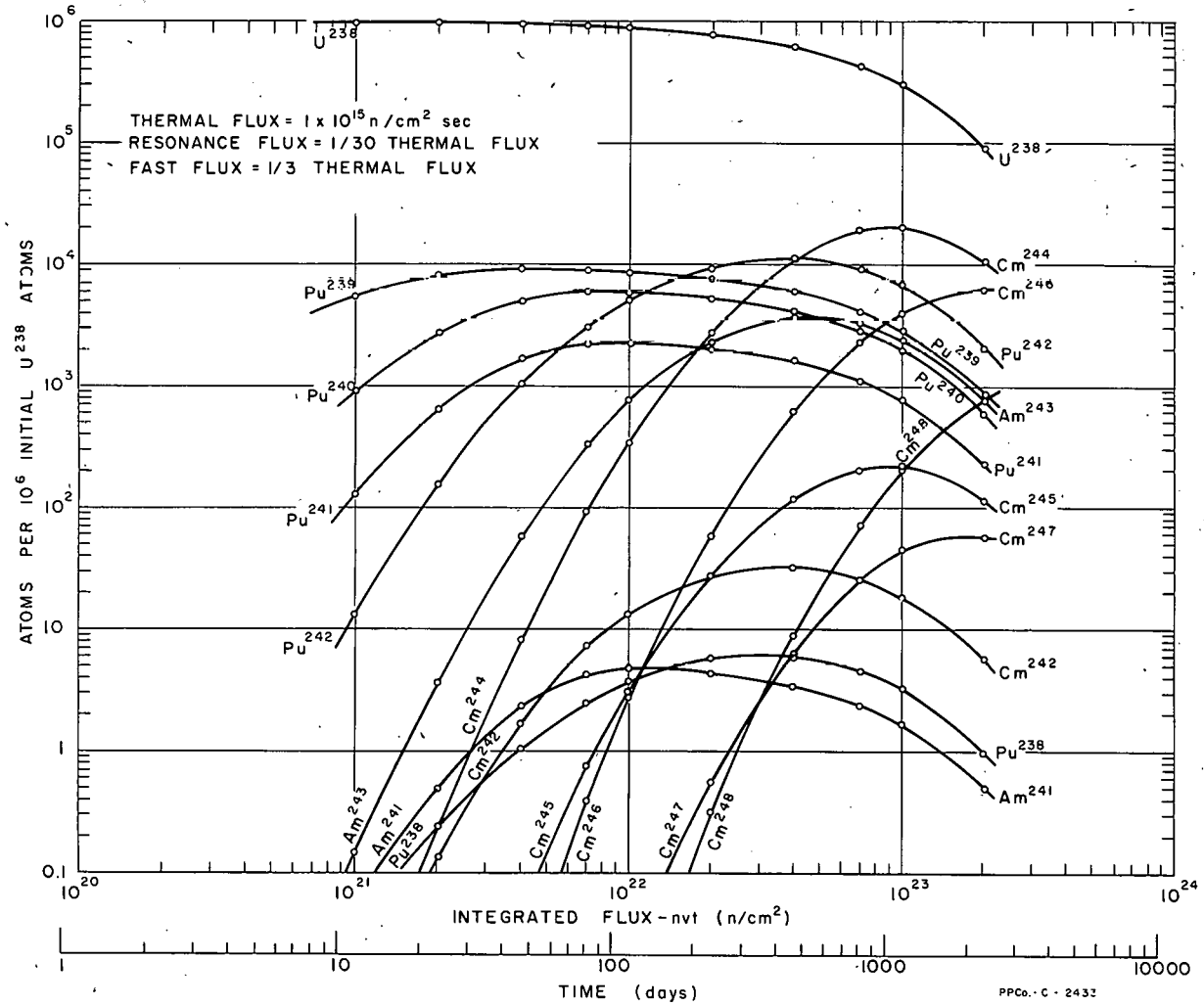


Fig. 10-2 Composition of pile irradiated U-238

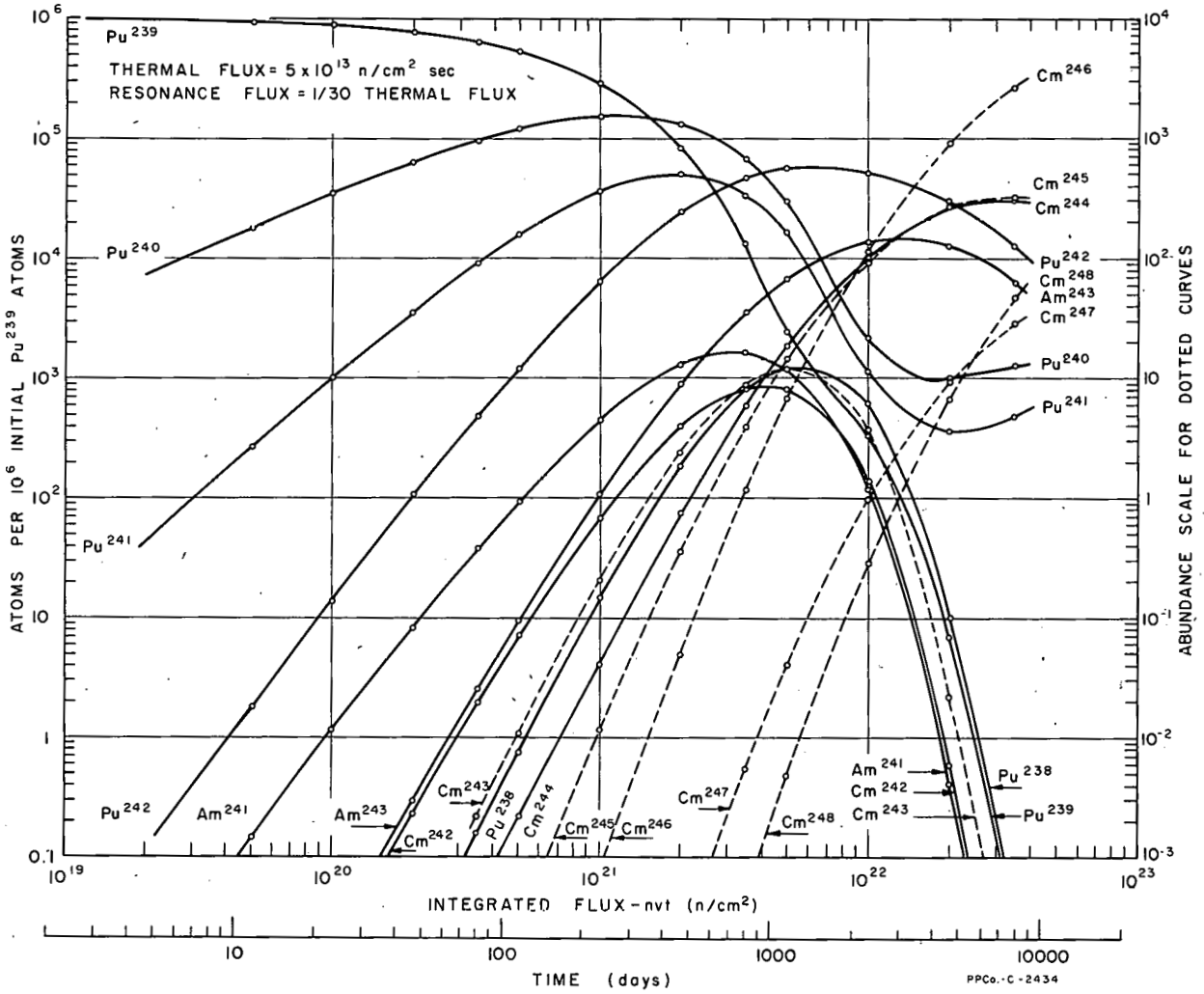


Fig. 11-1a Composition of pile irradiated Pu-239.

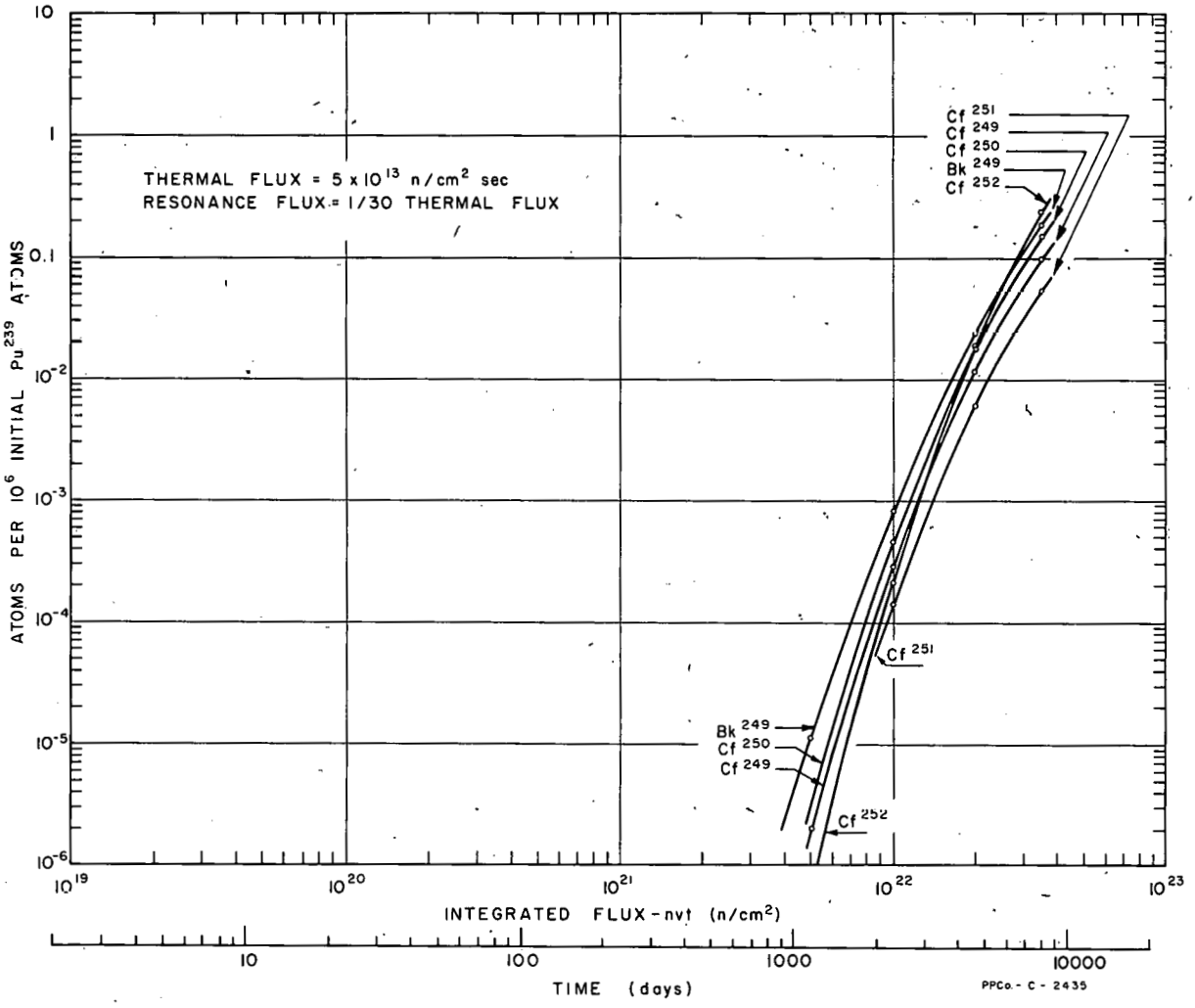


Fig. 11-1b Buildup of very heavy nuclides in pile irradiated Pu-239.

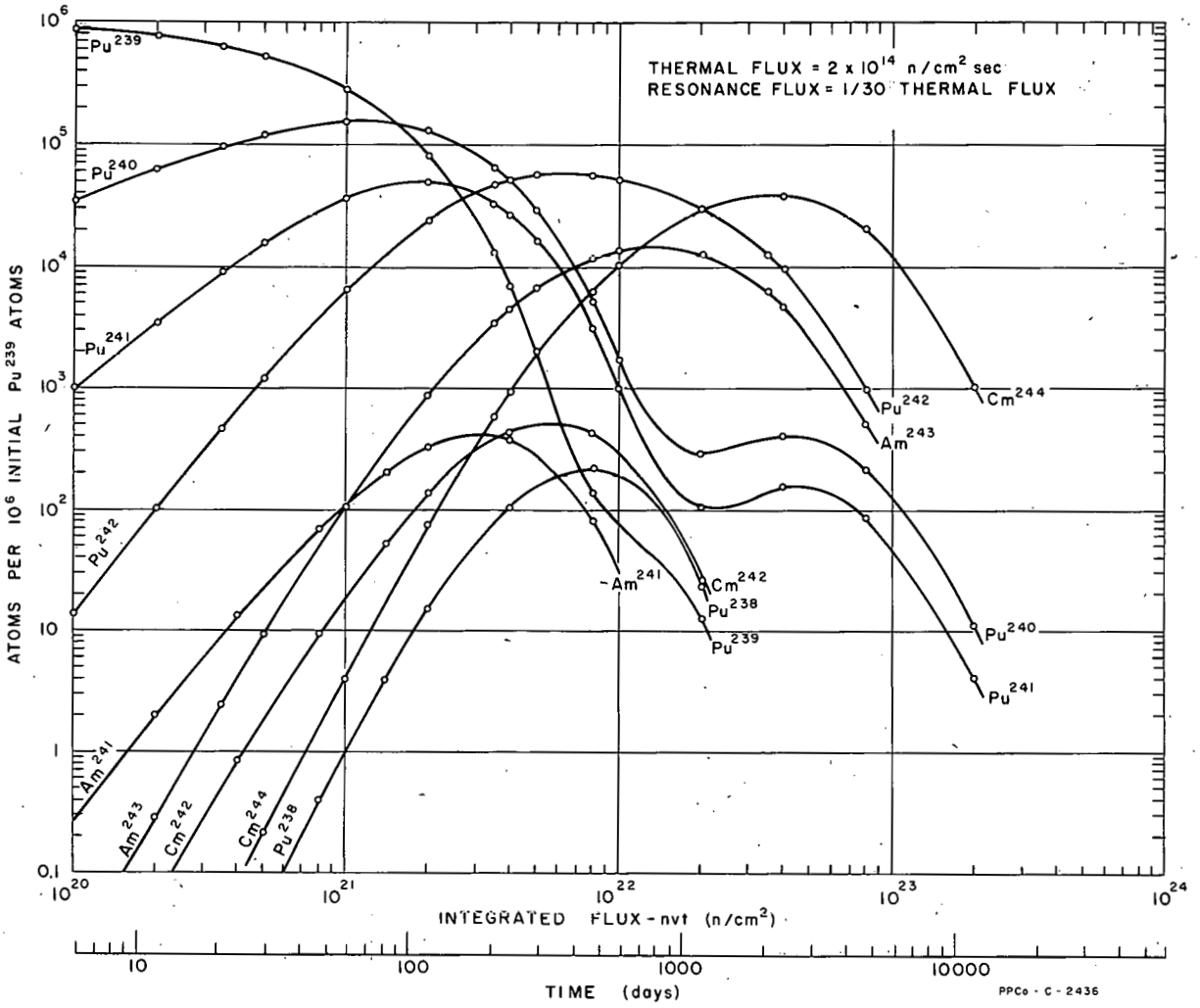


Fig. 11-2a Composition of pile irradiated Pu-239.

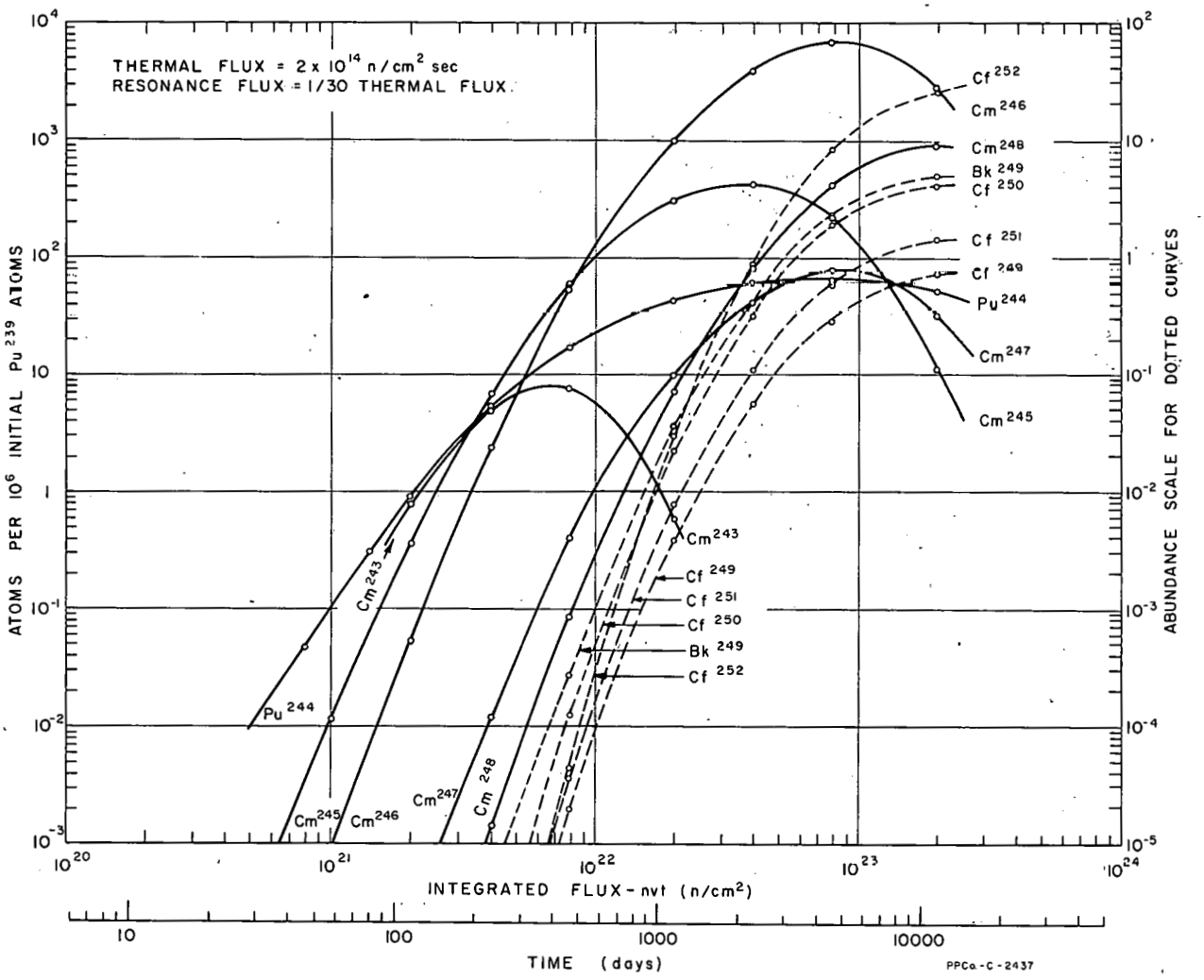


Fig. 11-2b Buildup of very heavy nuclides in pile irradiated Pu-239.

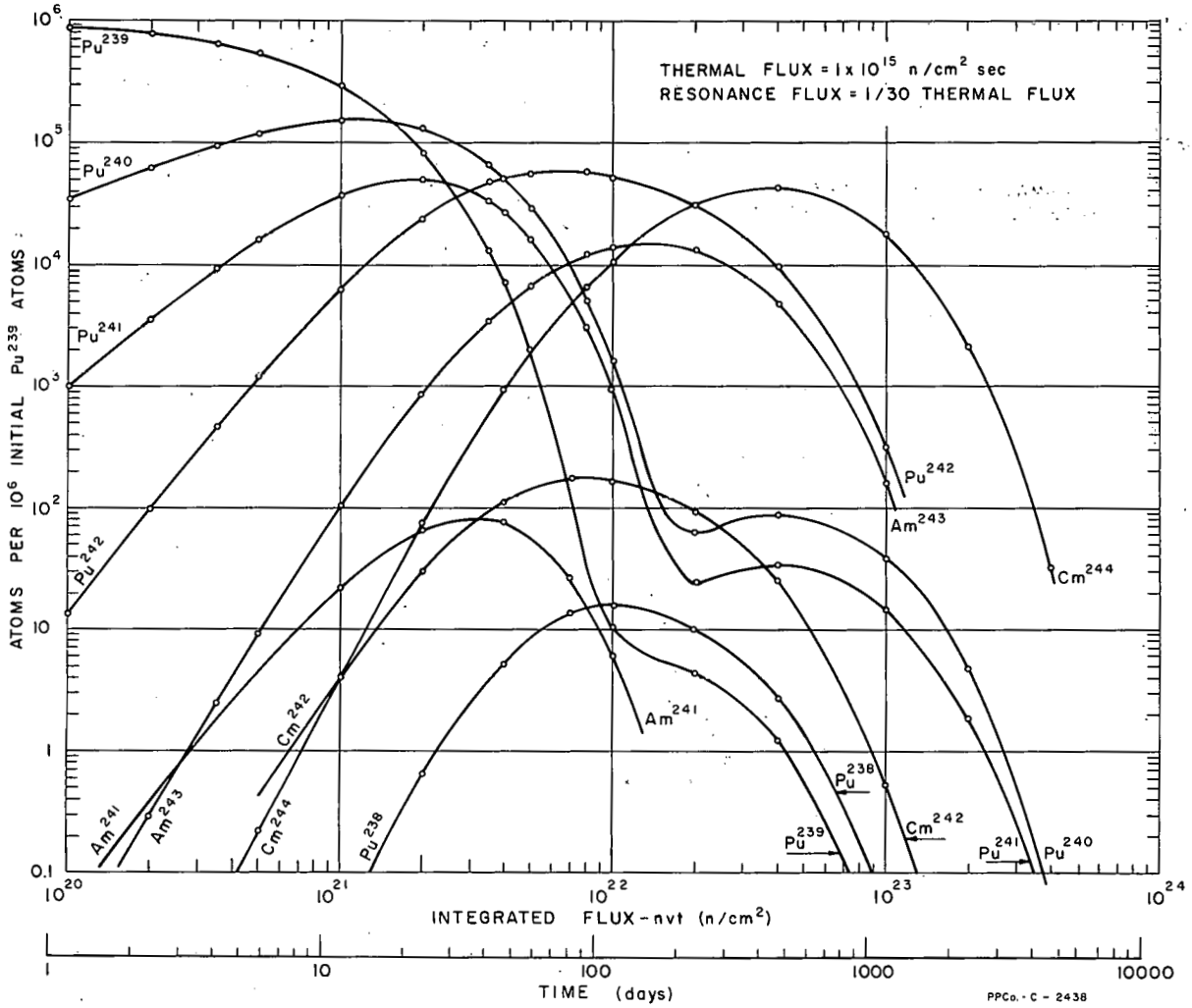


Fig. 11-3a Composition of pile irradiated Pu-239.

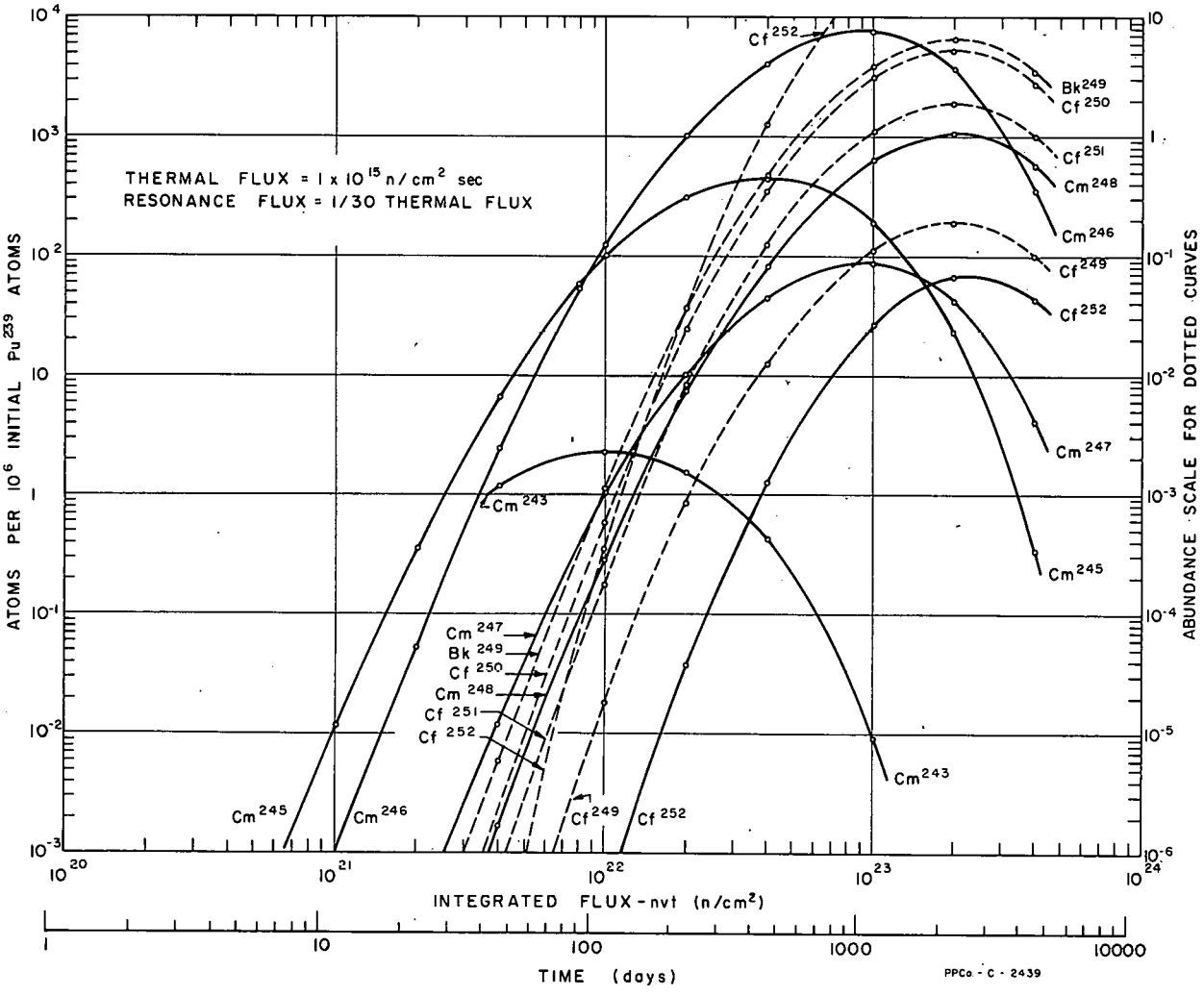


Fig. 11-3b Buildup of very heavy nuclides in pile irradiated Pu-239.

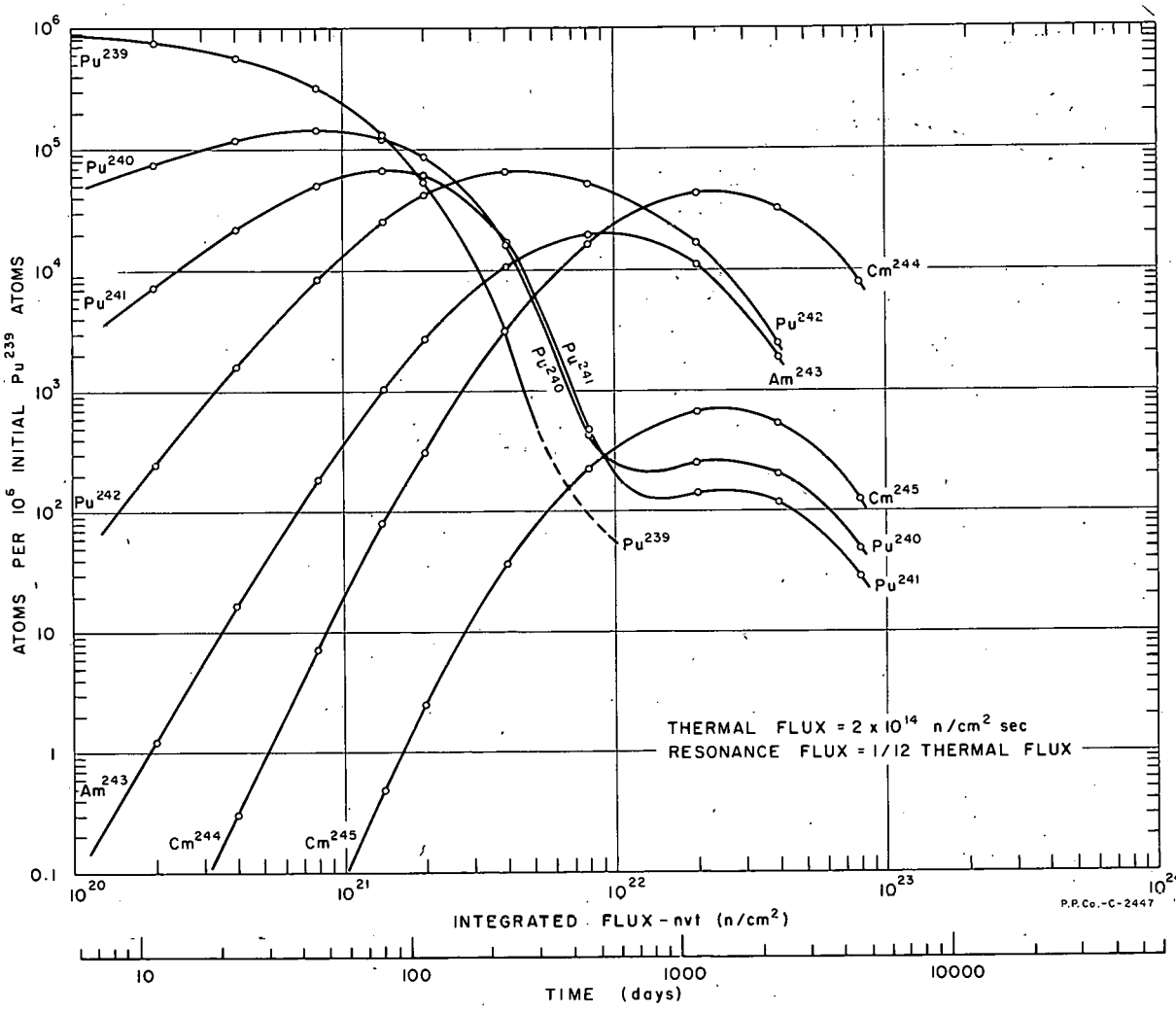


Fig. 11b Composition of pile irradiated Pu-239 for a pile flux with $\phi_{res}/\phi_{th} = 1/12$.

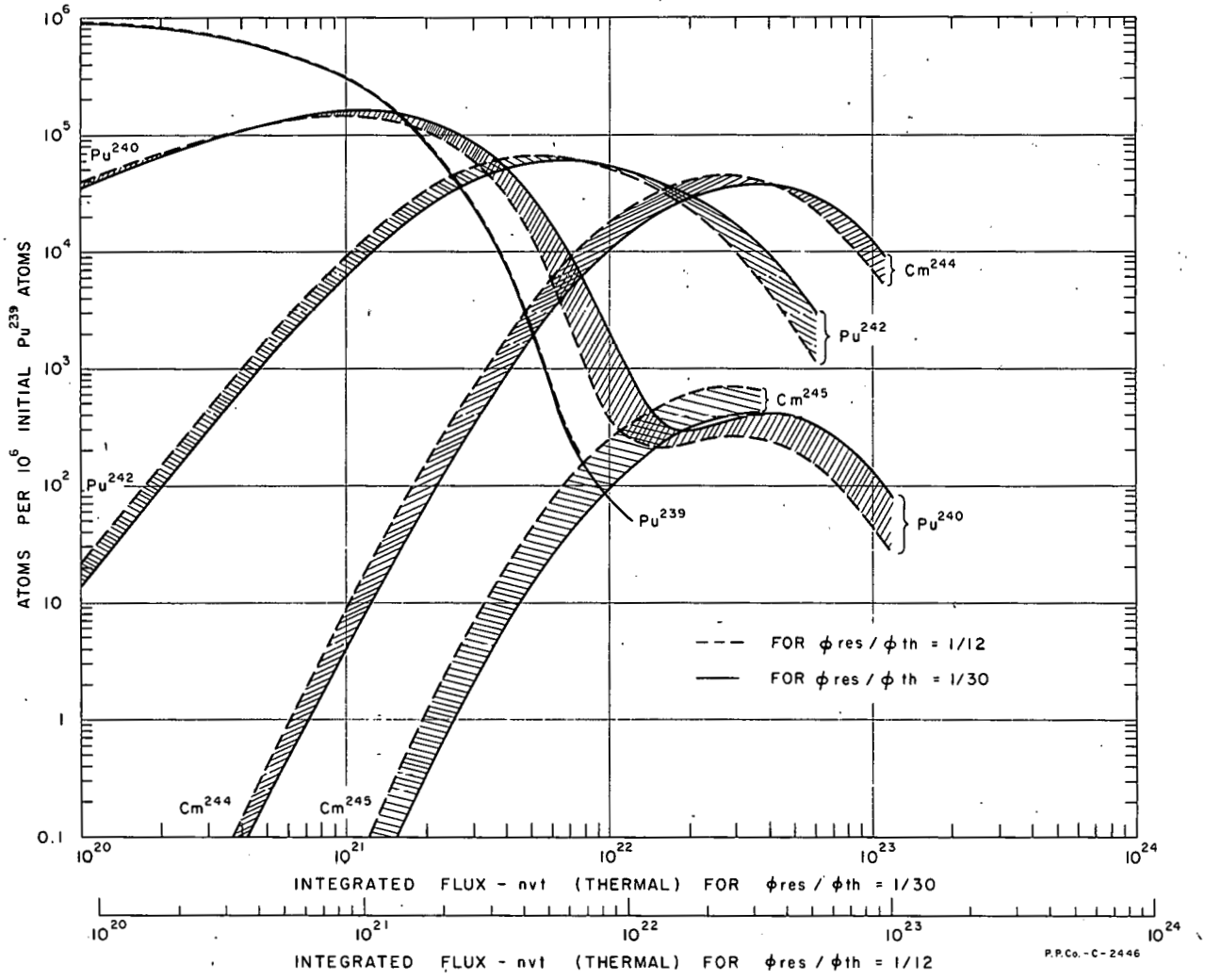


Fig. 11c Comparison of the buildup of heavy nuclides in Pu-239 for a pile flux with ϕ_{res}/ϕ_{th} = 1/12 and for a pile flux with ϕ_{res}/ϕ_{th} = 1/30. Curves plotted so that the Pu-239 burnout curves are superimposed.

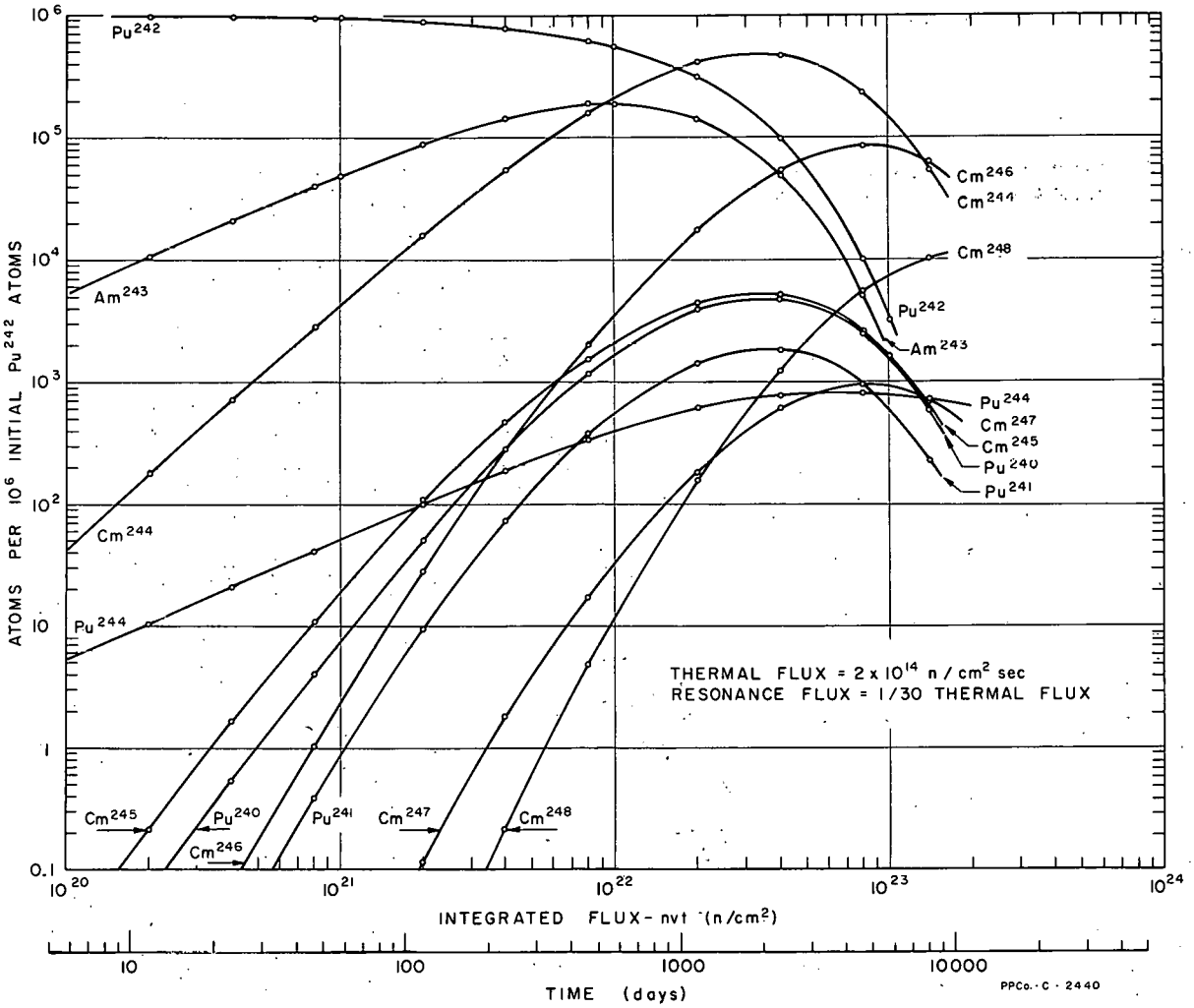


Fig. 12-1a Composition of pile irradiated Pu-242.

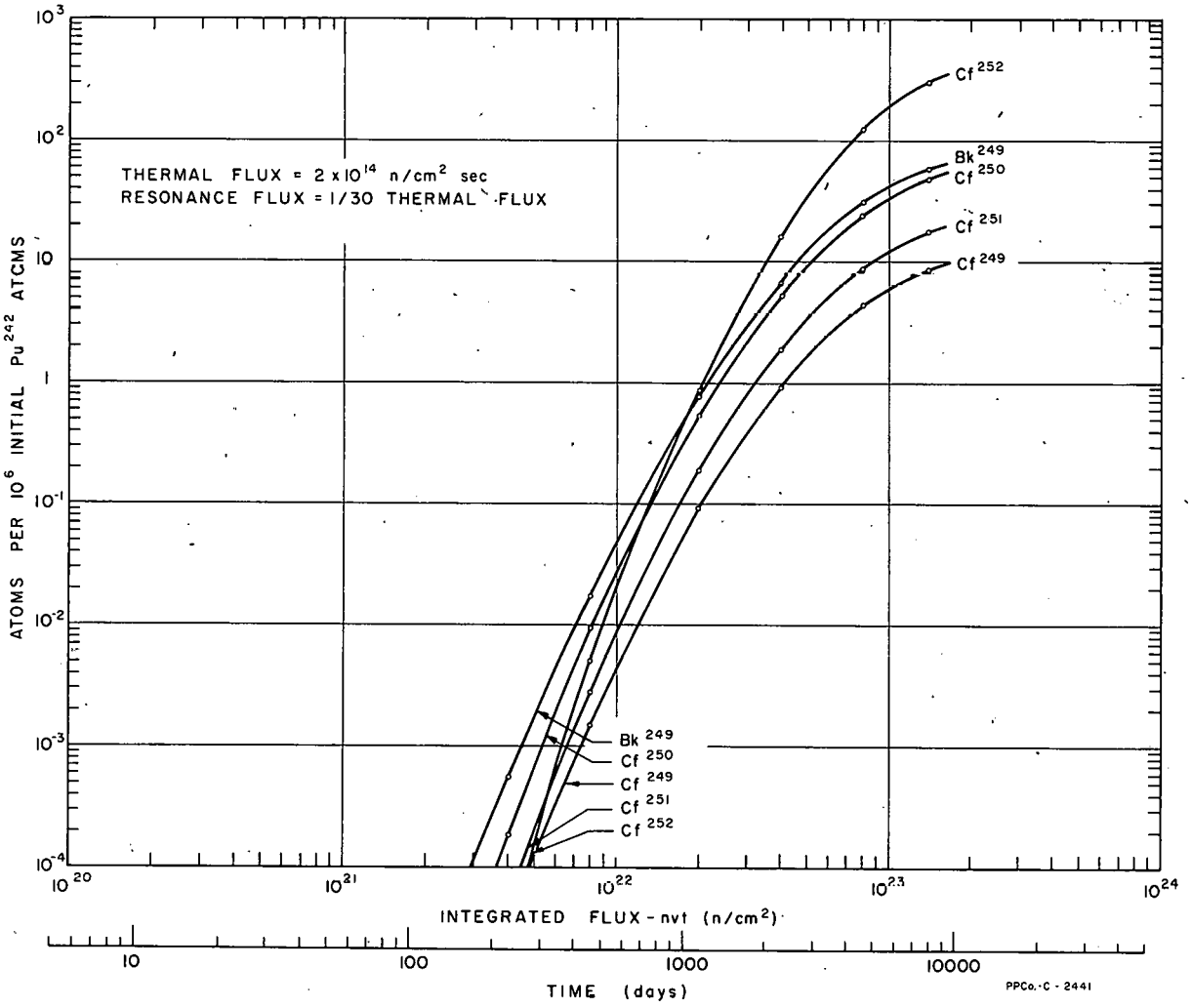


Fig. 12-1b Buildup of very heavy nuclides in pile irradiated Pu-242.

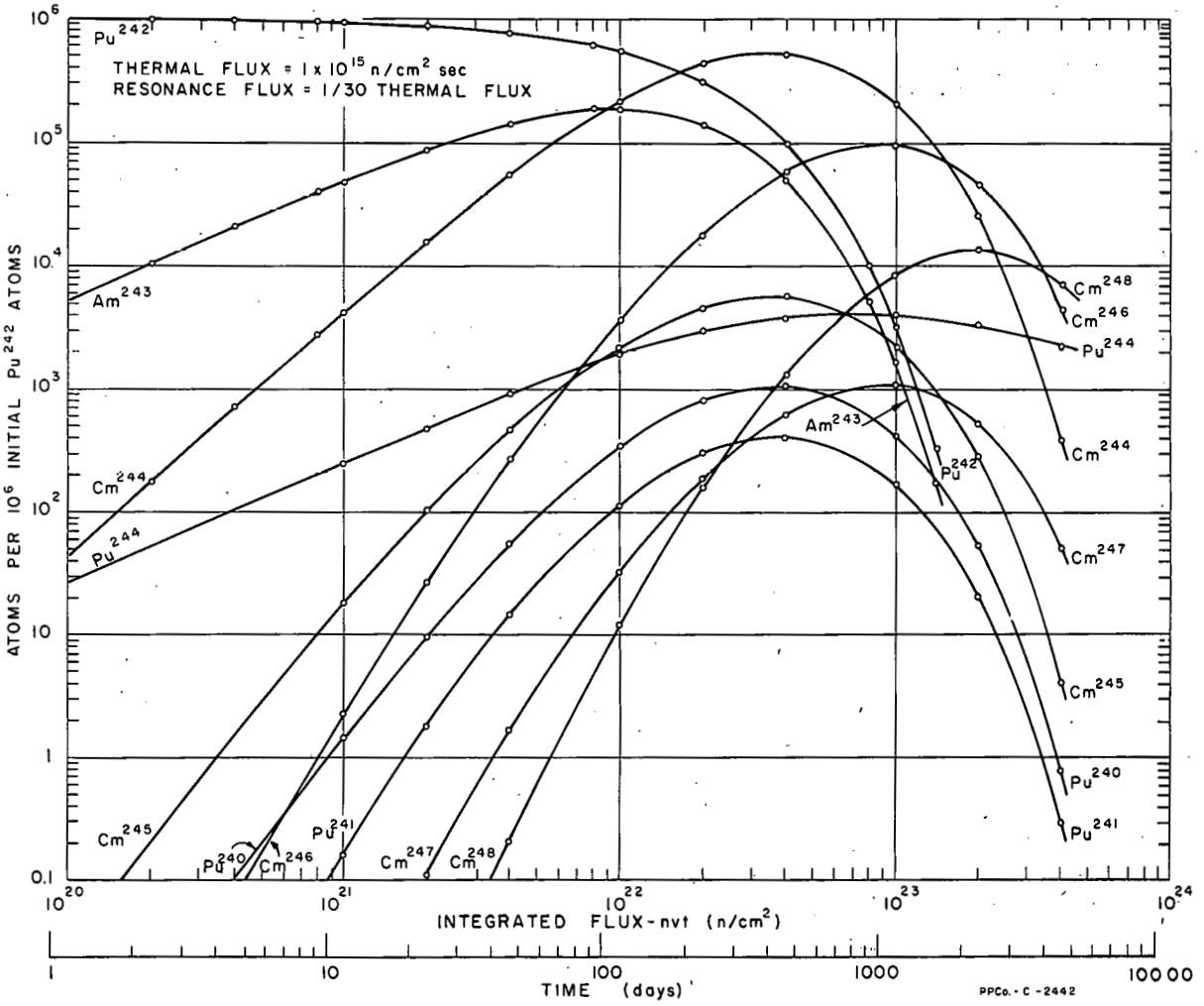


Fig. 12-2a Composition of pile irradiated Pu-242.

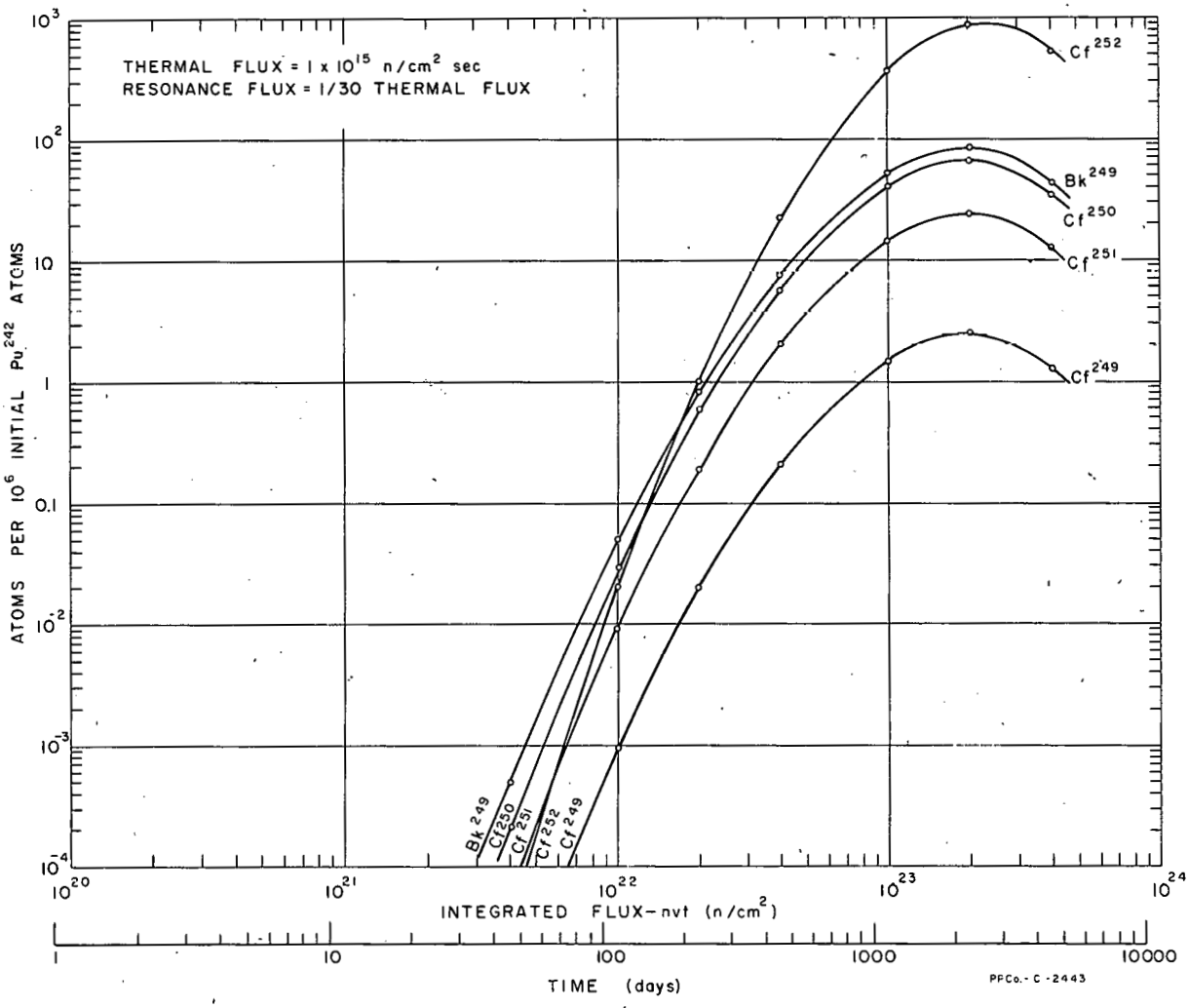


Fig. 12-2b Buildup of very heavy nuclides in pile irradiated Pu-242.

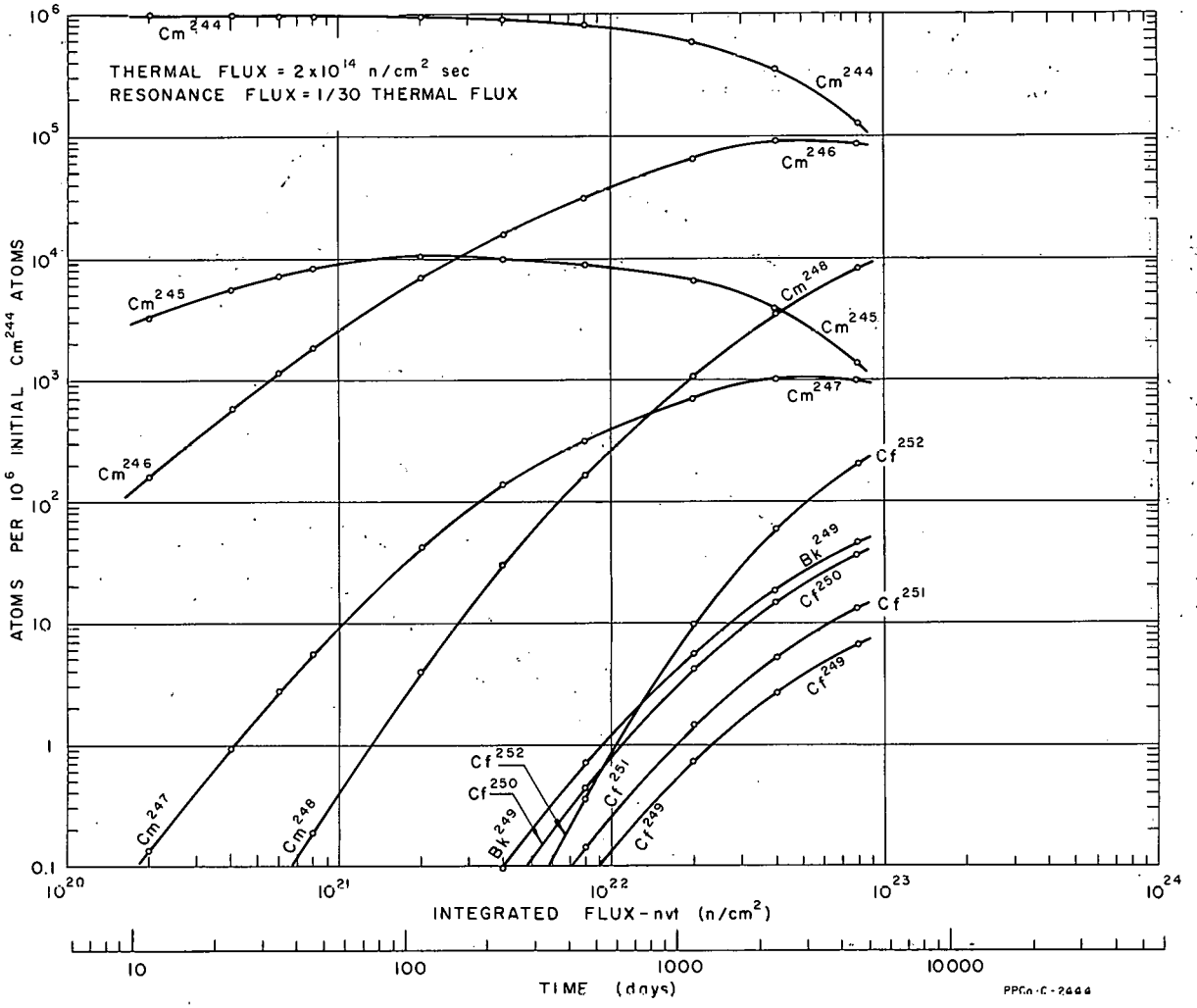


Fig. 13-1 Composition of pile irradiated Cm-244.

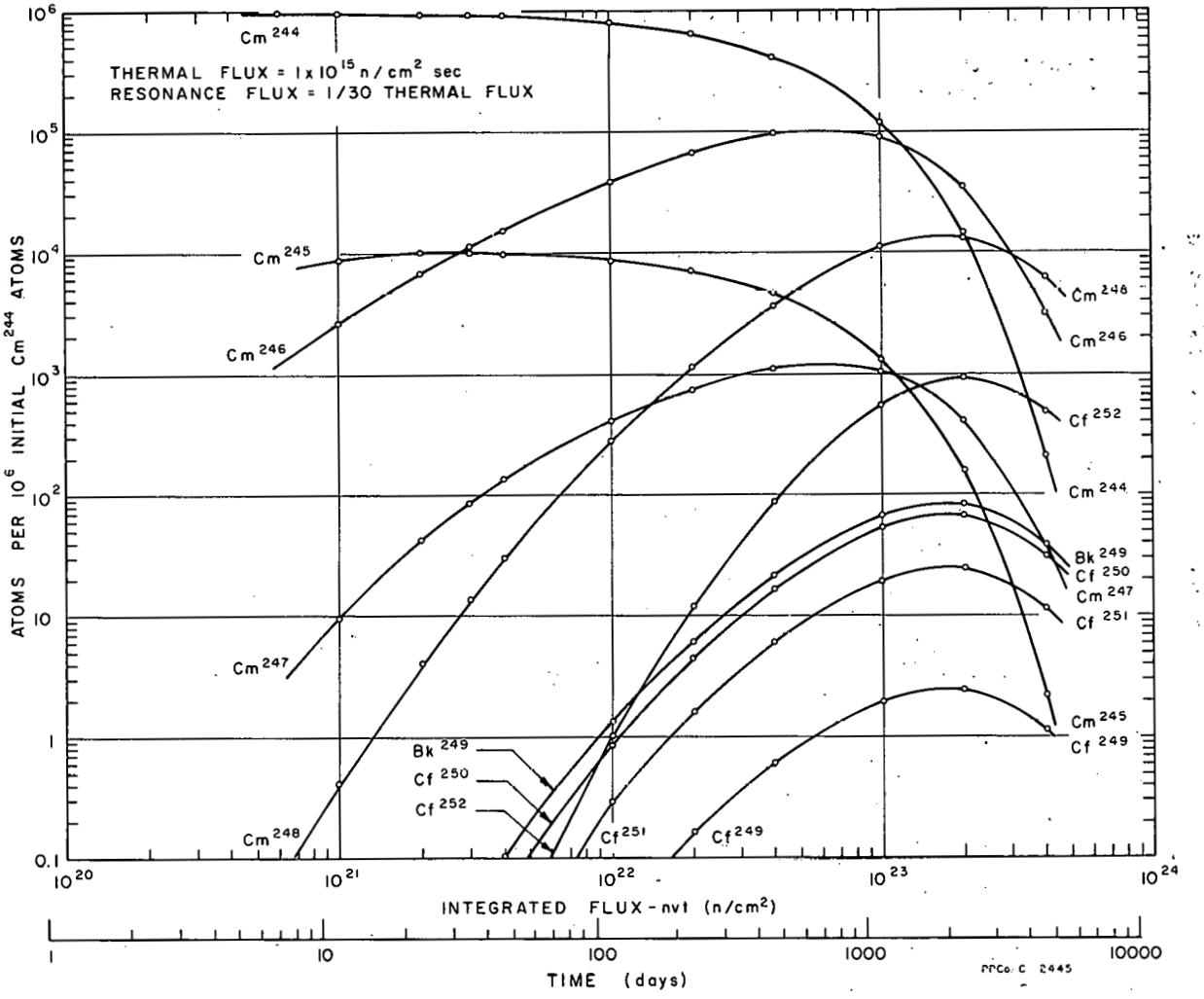
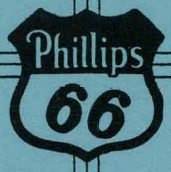


Fig. 13-2 Composition of pile irradiated Cm-244.


Phillips
66