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BNWL-2282
UC-78

High Burn-Up Plutonium Isotopic Compositions Recommended for Use in Shielding Analysis

by
M. G. Zimmerman

June 1977

Prepared for the Energy Research and Development Administration under Contract EY-76-C-06-1830

Battelle
Pacific Northwest Laboratories

BNWL-2282

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BATTELLE
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ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
Under Contract EY-76-C-06-1830

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $_____*; Microfiche $3.00

*Pages	NTIS Selling Price
001-025	$4.50
026-050	$5.00
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M. G. Zimmerman

June 1977

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ACKNOWLEDGMENT

This report summarizes a study conducted by Battelle, Pacific Northwest Laboratories for the U.S. Energy Research and Development Agency under Contract No. E(45-1)-1830. The author wishes to thank E. T. Merrill and C. M. Heeb for their technical contributions to the data used, C. L. Brown for his assistance in preparing the report, and John Church, E.I. duPont de Nemours & Co., SRL, for his review and helpful suggestions regarding applicability to the industry.

SUMMARY

Isotopic compositions for plutonium generated and recycled in LWR's have been estimated *for use in shielding calculations*. The values were obtained by averaging isotopic values from many sources in the literature. These isotopic values should provide the basis for a reasonable prediction of exposure rates from the range of LWR fuel expected in the future. The isotopic compositions given are meant to be used for shielding calculations, and the values are not necessarily applicable to other forms of analysis, such as inventory assessment or criticality safety. Final results are presented in Table 1.

TABLE 1. High Burn-Up Plutonium Isotopic Compositions Recommended For Use In Shielding Analysis

	Composition, wt%, At End of Cycle				
Isotope	Equilibrium Uranium Cycle	First Pu Recycle	Second Pu Recycle	Third Pu Recycle	Fourth Pu Recycle
Pu-236	0.08 ppm	0.06 ppm	0.06 ppm	0.06 ppm	0.06 ppm
Pu-238	2.	2.5	3.5	4.	5.
Pu-239	57.	43.	37.2	29.	26.
Pu-240	24.	28.5	30.2	29.	26.
Pu-241	12.	16.	16.1	19.	19.
Pu-242	5.	10.	13.	19.	24.

These compositions provide a resource for shielding and dose rate analysis of new facilities that will fabricate and process high-burnup plutonium fuel.

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INTRODUCTION

The purpose of this report is to provide a set of reference isotopic compositions for plutonium that can be used in shielding and dose rate calculations for new, near-future plutonium plants. It is anticipated that as more data become available, these compositions may be adjusted slightly.

DISCUSSION

In order to select a reasonable and conservative plutonium isotopic content for use in shielding calculations several factors must be considered. These are:

- Burnup and radiation history of the fuel
- Time after reprocessing
- Shield material and thicknesses expected to be used
- Chemical form of the plutonium

The burnup and irradiation history of the nuclear fuel determines the actual plutonium isotopic composition at discharge. The relative contribution to radiation exposure of each plutonium isotope will be determined by the shielding material and thickness. Radiation exposure and shielding needs will be increased by the buildin as a function of time of certain daughter isotopes of plutonium such as Am-241 and U-237. The chemical form of the plutonium will determine the number of (α,n) reactions and hence the total neutron source.

The important assumption is the ratio of Pu-236, Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242 isotopes in the plutonium. Each of these isotopes have different radiation characteristics. The percentage of each in any given plutonium is determined by the characteristics and duration in the reactor where the plutonium was produced. Once the spent fuel (and plutonium) is

discharged from the reactor, however, the plutonium isotopic ratios are essentially fixed. Knowing these approximate ratios is important in shielding and dose rate analysis. Details of these considerations are given below.

PLUTONIUM ISOTOPE PRODUCTION

The plutonium isotopes are produced by neutron absorption and subsequent radionuclide decay of U-235 and U-238. The major process is that for U-238 shown below:

$$^{238}U + n \rightarrow {}^{239}U + \gamma$$
$$^{239}U \rightarrow {}^{239}Np + e^- \qquad t_{1/2} = 23.5 \text{ minutes}$$
$$^{239}Np \rightarrow {}^{239}Pu + e^- \qquad t_{1/2} = 2.35 \text{ days}$$

This process occurs when U-238 absorbs a neutron with less energy then necessary to fission the U-238 isotope. A similar process occurs less frequently with U-235

$$^{235}U + n \rightarrow {}^{236}U + \gamma$$
$$^{236}U + n \rightarrow {}^{237}U + \gamma$$
$$^{237}U \rightarrow {}^{237}Np + e^- \qquad t_{1/2} = 6.75 \text{ days}$$
$$^{237}Np + n \rightarrow {}^{238}Np + \gamma$$
$$^{238}Np \rightarrow {}^{238}Pu + e^- \qquad t_{1/2} = 2.12 \text{ days}$$

Other isotopes of plutonium usually present are Pu-240, Pu-241 and Pu-242. These are formed by neutron absorption in Pu-239. The possible neutron absorptions and decays causing buildup of plutonium isotopes and other transuranic isotopes is shown in Figure 1.[1] The isotope Pu-236 is also produced to a small extent.

The various amounts of plutonium isotopes produced by these reactions are dependent on the cross-sections involved; and the effective cross-sections are determined by the neutron spectra. Therefore, the plutonium isotopic composition at discharge is determined by the burnup history and fuel parameters and the beginning plutonium isotopic composition of the fuel.

As the plutonium is recycled the relative isotopic production trends will continue. The trend consists of a reduction in Pu-239 fraction, little change in the fraction of Pu-240 and Pu-241 and an increase in the fraction of the other isotopes.

Only Pu-239 and Pu-241 are fissile. The relative fraction of fissile plutonium (Pu-239 + Pu-241) decreases with each recycle. Because of this, a greater amount of plutonium must be added to a given core for each recycle to achieve the same burnup. This means that eventually it becomes uneconomical to recycle the plutonium. At this point the plutonium is either discarded or used in LMFBR's. *Our estimate is that plutonium will not be recycled beyond four times.*

RADIATION AND SHIELDING CHARACTERISTICS OF PLUTONIUM ISOTOPES

In order to estimate reasonably conservative isotopic compositions it is necessary to consider the shielding problems associated with the different isotopes. The neutron and gamma yields for the different plutonium isotopes are shown in Tables 2 and 3.[2]

TABLE 2. Neutron Yields From α,n Reactions and Spontaneous Fission

	n/sec gram of Pu					
Isotope	PuO_2	PuF_4	PuC	PuBe[a]	$Pu(NO_3)$[b]	Spontaneous Fission
Pu-236	-	-	-	-	-	3.7×10^4
Pu-238	1.34×10^4	2.04×10^6	1.49×10^4	4.38×10^7	2.7×10^4	2.62×10^3
Pu-239	38	6.35×10^3	37.3	1.36×10^5	76	.03
Pu-240	1.45×10^2	2.35×10^4	137.8	5.02×10^5	2.9×10^2	1.02×10^3
Pu-241	-	-	-	-	-	-
Pu-242	2.13	2.83×10^2	-	-	4.2	1.7×10^3
Am-241	2.68×10^3	4.08×10^5	2.98×10^3	8.76×10^6	5.46×10^3	-

(a) Estimated from Reference 13.

(b) A rough estimate indicates an approximate doubling of (α,n) yields compared to PuO_2.

TABLE 3. Gamma Spectra and Yields

gammas/100 disintegrations

Energy Range (MeV)	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	U-237	Pu-236	U-232	Th-228	Ra-224	Pb-212[a]	Bi-212[b]	Fission Spectra[c]
0.01 - 0.07	15.25	6.65	13.24	3.6-4	13.27	93.14	80.71	3.1-3	30.2	25.0	0.0	0.0	1.5	1.4-3
0.07 - 0.12	8.2-3[d]	6.8-3	1.2-2	1.7-3	0.0	0.047	56.6	1.2-2	0.0	1.6	0.0	33.7	0.0	2.8-3
0.12 - 0.21	1.0-3	4.9-3	1.2-3	1.6-4	0.0	5.9-3	25.0	6.6-5	8.2-2	0.30	0.0	0.24	0.0	8.3-3
0.21 - 0.4	0.0	2.88-3	0.0	0.0	0.0	1.7-3	2.4	0.0	7.2-3	0.29	4.2	50.6	4.6	9.1-3
0.4 - 0.6	0.0	1.23-3	0.0	0.0	0.0	9.0-5	0.0	2.8-4	0.0	0.0	0.0	0.16	40.3	1.6-2
0.6 - 0.9	5.5-5	9.4-5	1.8-5	0.0	0.0	1.0-3	0.0	2.4-4	0.0	0.0	0.0	0.0	13.5	2.7-2
0.9 - 1.2	8.0-7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0	4.7-2
1.2 - 1.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.23	6.1-2
1.6 - 2.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.7	9.9-2
2.0 - 2.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.2-1
2.5 - 3.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	36.0	1.9-1
3.0 - 3.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.6-1
3.5 - 4.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.7-1
4.0 - 5.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	8.6-2
5.0 - 6.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	5.0-2
6.0 - 7.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	5.1-2
		0.0												

(a) Includes gammas from Po-216 and Rn-220

(b) Includes gammas from Po-212 and Tl-208

(c) Spectra normalized to 1.0, yields equal to 14.98 MeV/fission which includes prompt and fission product gammas.

(d) Read as 8.2×10^{-3}

The major contributors to the dose rate in a given plutonium isotopic composition will depend on the shielding, geometry, and age since reprocessing. The half-lives of the plutonium isotopes and the daughters which contribute to the shielding problem are shown in Table 4.

TABLE 4. Half-Lives of Plutonium and Daughters Which Contribute Significantly To Radiation Exposure(11)

Daughters	Parents	Radioactive Decay Half-Life	Spontaneous Fission Half-Life
Pu-236	-	2.85 yr	3.5×10^{9} yr
Pu-238	-	86.4 yr	4.9×10^{10} yr
Pu-239	-	24390 yr	5.5×10^{15} yr
Pu-240	-	6580 yr	1.34×10^{11} yr
Pu-241	-	13.2 yr	-
Pu-242	-	3.79×10^{5} yr	7.1×10^{10} yr
Am-241	Pu-241	458 yr	2.0×10^{14} yr
U-237	Pu-241	6.75 day	-
U-232	Pu-236	72 yr	8×10^{13} yr
Th-228	U-232	1.91 yr	-
Ra-224	Th-228	3.64 day	-
Rn-220	Ra-224	55.3 sec	-
Po-216	Rn-220	0.145 sec	-
Pb-212	Po-216	10.64 hr	-
Bi-212	Pb-212	60.6 min	-
Po-212	Bi-212	0.304 μ sec	
Tl-208	Bi-212	3.10 min	-

Since each isotope has a different activity, the radiation source strength is determined by the isotopic composition. However, each isotope has a different gamma and x-ray spectra, and the isotope responsible for most of the dose rate can change depending on the shield thickness and geometry. In conditions of no shielding or very thin shields like hood gloves, the major contributors to the dose rate are those isotopes producing the most low energy gammas and x-rays. For this reason a geometry of plutonium with little self-shielding may produce a much greater dose rate then an equivalent amount of plutonium in a small dense source.

Several isotopes, notably Pu-236 and Pu-241 decay to daughters which contribute a significant dose rate. For this reason the time after reprocessing can change the relative contribution of the plutonium isotopes to the dose rate.

Most of the preceding discussion applies only to the gamma source. Because the neutron spectra from the different isotopes are essentially the same for shielding purposes, the geometry and shield thickness has little effect on the relative contribution to the dose rate by the different isotopes. Therefore, the dominate plutonium isotope from a neutron shielding standpoint is determined solely by the isotopes activity and density compared to the other plutonium isotopes.

The neutron and gamma radiation characteristics for each isotope are discussed below:

Pu-236

Typically recycle plutonium is expected to contain less then 0.1 ppm Pu-236. This isotope does not contribute significantly to neutron dose rates and the gamma dose rate is small compared to that from other isotopes. However because of decay daughters, the gamma dose rate will buildup with time. The decay scheme is shown in Figure 2. Equilibrium conditions will be reached in about 18 years. The major shielding problem is a 2.6 MeV gamma from Tl-208. Because of the high energy of this gamma ray, this isotope can contribute significantly to the dose rate under conditions of thick shields and several years time since reprocessing.

Pu-238

The amount of Pu-238 varies from about 1% to about 5% of the plutonium. This isotope, because of its abundance and relatively short half-life, is a major contributor to the dose rate. The gammas are mostly low energy and can be shielded by thin shields. However, there are enough high energy gammas to dominate the gamma dose rate if the shields are of the proper thickness.

The Pu-238 can also be the major contributor to the neutron dose rate. This is especially true in compounds with a potential for (α,n) reactions. The spontaneous fission half-life is also relatively short.

Pu-239

Pu-239 generally contributes less to the dose rate on a per atom basis then any of the others. The half-life for Pu-239 is much longer then for the other isotopes except α-decay of Pu-242. Only when Pu-239 makes up essentially all of the plutonium is it a major contributor to the dose rate. The Pu-239 can make up a significant portion of the gamma dose rates through thin lead shields. The Pu-239 can be more important if the dose rate due to induced fissions caused by neutron multiplication is significant. Since the Pu-239 and the Pu-241 are the only fissile isotopes, the neutron multiplication will depend on the amount of these isotopes.

Pu-240

Pu-240 may be about 25% of the plutonium. The gamma dose rate is mostly from low energy gammas and even thin shields reduce the contribution to insignificance.

This isotope can contribute about half of the neutron dose rate. This is due to both spontaneous fission and (α,n) neutrons.

Pu-241

Pu-241 may be about 17% of the plutonium. This isotope contributes primarily to the gamma dose rate through its daughters U-237 and Am-241. The decay scheme is shown in Figure 2. These isotopes are major contributors to the gamma dose rate for most shielding situations. The U-237 builds in to equilibrium within a month and the Am-241 builds in to significant levels in about a year after reprocessing.

Pu-241 contributes very little to the neutron dose rate. The Am-241 can contribute a small amount to the neutron dose rate through (α,n) reactions. Pu-241 is also fissile and may contribute to the induced fission neutron dose rate.

Pu-242

Pu-242 may be about 15% of the plutonium. This isotope contributes insignificantly to the gamma dose rate.

Pu-242 does contribute significantly to the neutron dose rate. This is mostly from spontaneous fission although some (α,n) neutrons can also be present.

From the foregoing discussion of the individual plutonium isotopes we can conclude that all the isotopes can contribute significantly to dose rates for differing conditions. However, in most instances in fabrication and reprocessing steps for plutonium recycle, the major contributors are Pu-238 and Pu-241 for the gamma dose rate and Pu-238, Pu-240 and Pu-242 for the neutron dose rate. Under conditions of thick shielding and long storage time, Pu-236 daughters may contribute a significant amount to the gamma dose rate. The proportions of the dose rate from these isotopes will vary depending on what shielding is employed and the specific isotopic composition.

Therefore in order to choose conservative isotopic compositions for recycle plutonium we would generally choose the composition with the least Pu-239 and the most Pu-238 and Pu-241. Generally the compositions with the most Pu-238 and Pu-241 would also have the most Pu-240 and Pu-242. However, in those cases where induced fissions are important we may want to choose a composition with the most Pu-239 and Pu-241 in order to assure conservatism.

REPORTED PLUTONIUM ISOTOPIC COMPOSITIONS FOR UO_2 AND PU-UO_2 FUELS

We have investigated several references for the isotopic composition of recycle plutonium. The mean isotopic composition and the range of values found for each isotope is shown in Table 5.

TABLE 5. Mean Isotopic Compositions From Plutonium Recycle

	Equilibrium Uranium Cycle	First Pu Recycle	Second Pu Recycle	Third Pu Recycle
Pu-236	0.08 ppm	0.06 ppm	0.06 ppm	-
Pu-238	1.9 ± .6	2.3 ± 1.0	3.4 ± 1.5	4.3 ± 0.7
Pu-239	57.8 ± 3.4	46.9 ± 9.2	38.2 ± 8.8	29.0 ± 2.0
Pu-240	23.7 ± 2.3	28.0 ± 4.0	30.3 ± 3.3	29.8 ± 2.8
Pu-241	12.2 ± 2.1	16.0 ± 4.0	16.2 ± 8.2	18.8 ± 2.8
Pu-242	4.2 ± 1.1	10.0 ± 5.0	11.7 ± 3.7	19.2 ± 0.8

From this table we averaged the values for our recommendation in Table 1. In order to get the fourth plutonium recycle results in Table 1 we extrapolated the results for the first three recycles. Also, we show in Table 6 the predicted average plutonium isotopic composition for 1975, 1980, 1985.[3] The various isotopic compositions from the individual references are shown in Tables 7, 8, 9, and 10. From these tables we can observe that there is a large variation in plutonium isotopic composition between the different sources.

TABLE 6. Average Composition of Plutonium Available for Recycle[3]

Year	^{236}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
1975	0.000006	1.0	64	22	10	3
1980	0.0000007	1.5	58	24	11	5
1985	0.0000007	1.7	54	25	12	7

This variation in results from the different references is explained mostly by the burnup assumed. During exposure of the fuel in a reactor core, the production of plutonium is not constant with time. Initially when most of the power is produced from U-235 fissions, only a small amount of plutonium is produced. As the U-235 is burned out and more power is produced from Pu-239 the effective production rate for the Pu-238, Pu-240, Pu-241 and Pu-242 isotopes increases relative to Pu-239. Therefore, the production rate of these isotopes is greatest just before the fuel is discharged. In the cases where fuel burnup and enrichment was given by the references used for this report, the burnup was about 27,000 MWD/MT for BWRs and 33,000 MWD/MTM for PWRs with 2.7% enrichment for BWRs to 3.3% enrichment for PWRs.

The isotopic compositions are generally based on average core burnups. Some portions of the fuel may achieve higher burnups and therefore higher proportions of Pu-238, Pu-241 and Pu-242. Likewise other portions of the core will achieve lower burnup. Therefore the plutonium isotopics given can only be considered as approximate averages of the actual plutonium isotopic composition.

TABLE 7. Plutonium Isotopic Compositions Discharged From The Uranium Cycle

	Plutonium Composition from First Uranium Cycle, wt%		Plutonium Composition from Equilibrium Uranium Cycle, wt%										
	(9)[a]	(3)	(3)	(5)	(6)	(6)	(7)	(8)	(10)PWR	(10)BWR	(1)	(15)PWR	(15)BWR
Pu-236	-	0.004 ppm	0.07 ppm	-	-	-	-	-	-	-	8.8-06	-	-
Pu-238	1	0.6	1.6	2.5	2	2.5	1.9	1.9	1.5	1.3	1.5	1.5	1.3
Pu-239	70	69	56	57	61	55.5	57.9	59.2	58.0	61.3	54.5	55.	55.1
Pu-240	17	20	26	23	24	23.6	24.7	24.0	22.8	21.4	21.6	23.9	26.0
Pu-241	10	8	11	11	10	13.5	11.0	11.1	13.6	12.7	12.5	14.3	12.7
Pu-242	2	2	5	5.2	3	4.9	4.4	3.8	4.3	3.4	4.9	5.3	4.8

a. These numbers refer to the references.

TABLE 8. Plutonium Isotopic Composition Discharged From The First Plutonium Recycle

	Plutonium Composition, wt%									
	(9)	(3)	(5)	(7)PWR	(8)	(10)PWR	(10)BWR	(7)BWR	(15)PWR	(15)BWR
Pu-236	-	0.06 ppm	-	-	-	-	-	-	-	-
Pu-238	2	1.7	3.2	3.46	2.9	1.2	1.6	3.4	2.2	2.0
Pu-239	56	51	40	38.2	39.6	37.7	41.9	41.7	39.1	41.3
Pu-240	24	27	30	29.4	25.6	29.1	27.6	29.2	30.6	31.9
Pu-241	13	12	15	17.2	16.8	19.9	18.7	15.2	17.9	15.8
Pu-242	5	8	10	11.7	15.0	12.2	10.2	10.2	10.3	8.9

TABLE 9. Plutonium Isotopic Composition Discharged From The Second Plutonium Recycle

	Plutonium Composition, wt%					
	(9)	(3)	(5)	(7)	(15)PWR	(15)BWR
Pu-236	-	0.06 ppm	-	-	-	-
Pu-238	4	1.9	4.2	4.9	2.9	2.8
Pu-239	47	47	34	29.4	32.2	33.7
Pu-240	28	27	30	33.5	30.1	32.4
Pu-241	13	13	16	17.4	19.4	17.7
Pu-242	8	11	15	14.9	15.4	13.5

Other reasons for the differences observed in Tables 7-10 include differences in cross-sectional data and sophistication of techniques. The older results especially may be incorrect because of new cross-sectional data made available since they were calculated.

A comparison of our average isotopic composition in Table 1 and two other references is shown in Table 11. These two references did not present the ^{238}Pu wt%, therefore the comparison is made as a function of the ratio of the amount of a given plutonium isotope to that of ^{239}Pu. The results from references 15 and 16 differ from the Table 1 averages, however, the range of values in Tables 6 and 7 includes the values for the 1st and 2nd plutonium recycles from references 15 and 16 in Table 11. The third and fourth recycle values from Table 1 differ from those values given in references 15 and 16 because of the way in which the plutonium is recycled. The recycle plutonium given in Table 1 has been segregated. Segregation means that the plutonium from the first plutonium recycle is recycled separately from other plutonium into the second recycle and similarly into the third and fourth recycle. In references 15 and 16, the plutonium from each recycle step is mixed with plutonium from the rest of the UO_2 fueled core. This reduces the amounts of ^{240}Pu, ^{241}Pu, and ^{242}Pu relative to ^{239}Pu. The conservative approach for shielding analysis is to assume the plutonium is segregated.

TABLE 10. Plutonium Isotopic Compositions From The Third Plutonium Recycle

	Plutonium Composition, wt%		
	(5)	(15)PWR	(16)BWR
Pu-236	-	-	-
Pu-238	5	3.7	3.6
Pu-239	31	27.0	27.8
Pu-240	27	28.3	32.5
Pu-241	16	21.5	18.7
Pu-242	20	19.4	18.4

TABLE 11. Ratio of Individual Plutonium Isotopes to Pu-239 in Discharged LWR Fuel

	Ratios from Pu Recycle in PWR's[15]					Ratios from[16] Pu Recycle in BWR's		Ratios from Average Pu Recycle in LWR's(a)			
	1	2	3	4	5	1	4	1	2	3	4
Pu-239	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Pu-240	0.58	0.59	0.60	0.60	0.59	0.65	0.68	0.66	0.81	1.0	1.0
Pu-241	0.48	0.51	0.53	0.53	0.54	0.48	0.54	0.37	0.43	0.66	0.73
Pu-242	0.33	0.47	0.61	0.72	0.81	0.40	0.92	0.23	0.35	0.66	0.92

a. From Table 1

It would be difficult to include references 15 and 16 results in the averages because no ^{238}Pu values are given. The ^{238}Pu cannot be ignored in the shielding analysis.

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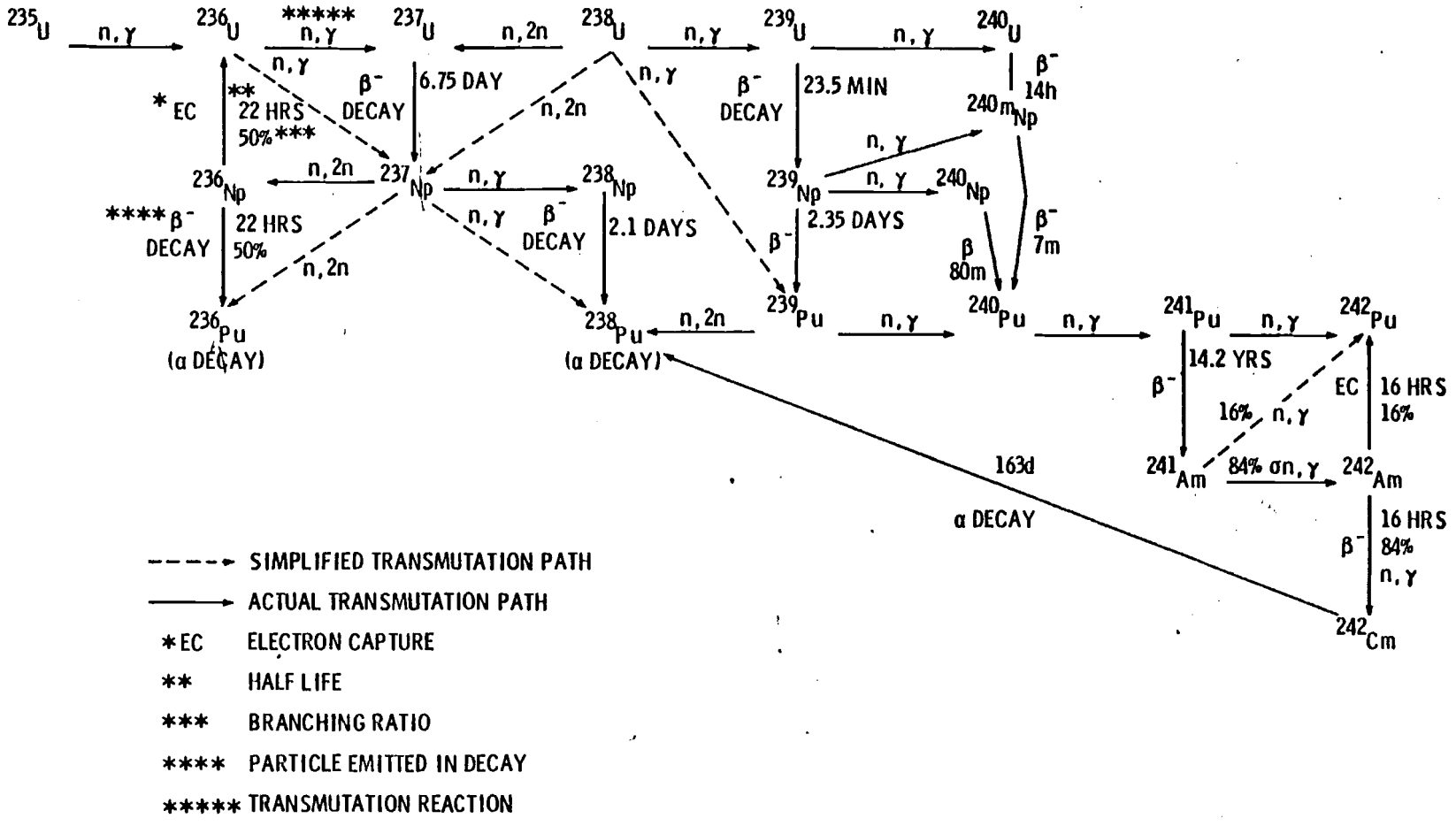
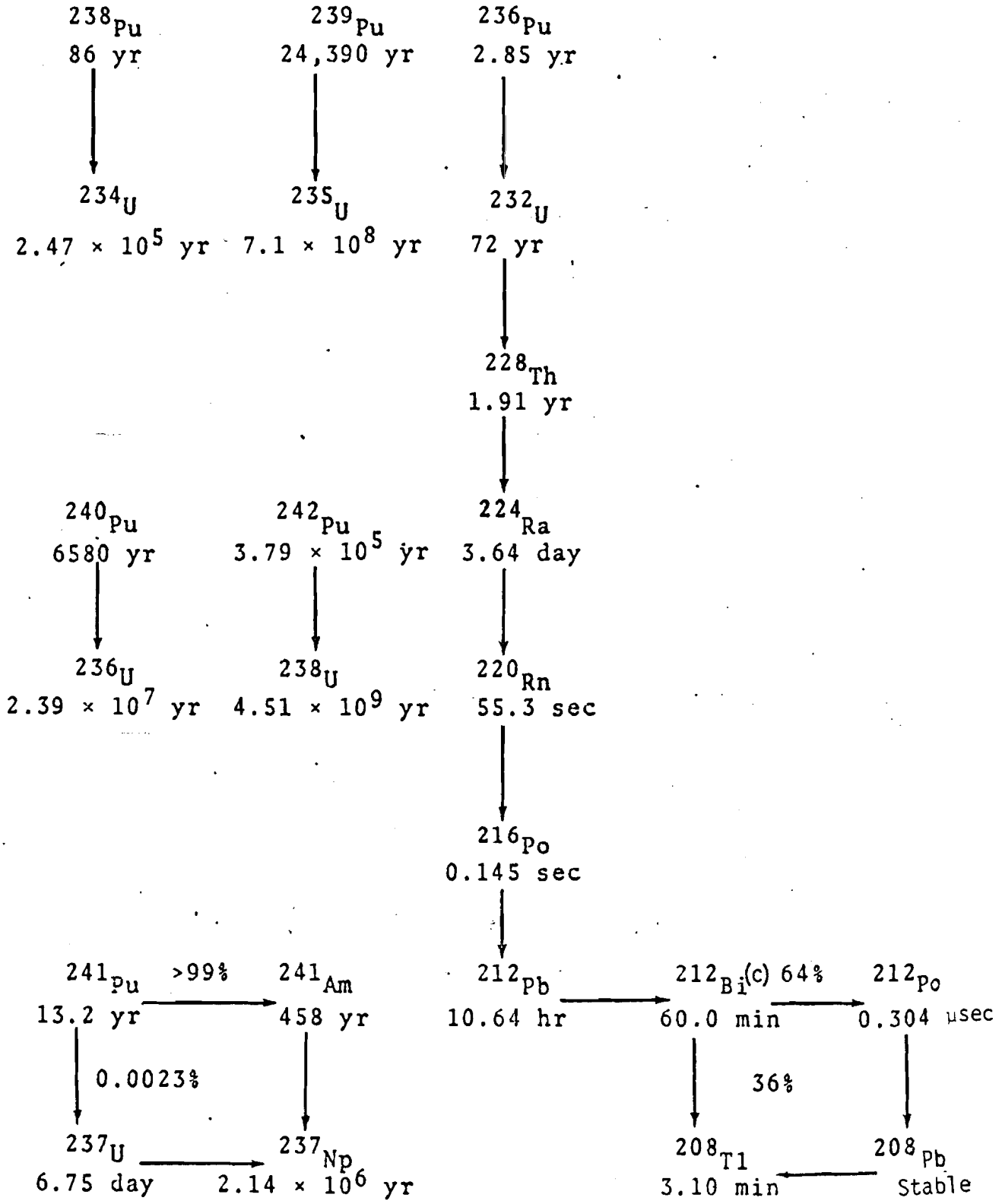


FIGURE 1. Actual and Simplified Transmutation Paths Used in the Zodiac Program [14]

Figure 2

Plutonium Isotopic Decay Daughters[a]



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