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BURNUP DETERMINATION OF NUCLEAR FUELS
PROJECT REPORT FOR THE
QUARTER JULY 1 - SEPTEMBER 30, 1967

Edited by

Frederick L. Lisman, William J. Maeck, and James E. Rein

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
IDAHO FALLS, IDAHO

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PROJECT REPORT
FOR THE QUARTER JULY 1 - SEPTEMBER 30, 1967

Edited by
Frederick L. Lisman, William J. Maeck, and James E. Rein

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SUMMARY

In this continuing program to develop methods for the accurate determination of the burnup of nuclear fuels, the main emphasis this quarter was the measurement of the absolute thermal fission yields of barium, cerium, cesium, krypton, neodymium, rubidium, strontium, xenon, and zirconium for Pu-239 by isotope dilution mass spectrometry. Also measured was $\hat{\alpha}_9$, the effective capture-to-fission ratio for Pu-239. The average values, determined from three different Pu-239 capsules, are:

Fission Yields for Pu-239

Isotope	% Fission Yield	Isotope	% Fission Yield
Kr-83	0.292±0.004	Xe-131	3.51±0.05
Kr-84	0.473±0.007	Xe-132	4.98±0.08
Kr- + Rb-85	0.560±0.006	Cs-133	6.95±0.07
Kr-86	0.748±0.011	Xe-134	7.01±0.11
Rb-87	0.975±0.010	Cs-137	6.56±0.07
Sr-88	1.32±0.02	Ba-138	5.24±0.06
Sr-90	2.05±0.02	Ce-140	5.44±0.06
Zr-91	2.45±0.02	Ce-142	4.89±0.05
Zr-92	3.01±0.03	Nd-143	4.36±0.05
Zr-93	3.84±0.04	Nd- + Ce-144	3.65±0.04
Zr-94	4.39±0.04	Nd-145	2.96±0.03
Zr-96	4.99±0.05	Nd-146	2.43±0.02
		Nd-148	1.66±0.02
		Nd-150	0.940±0.009

$\hat{\alpha}_9 = 0.4370 \pm 0.0044$

These values are tentative and as additional capsules are analyzed, refined data will be reported.

Previous project reports are:

Annual - 1962	TID 17385
Annual - 1963	IDO-14649
Annual - 1964	IDO-14656
Quarter, January - March, 1965	IDO-14660
Quarter, April - June, 1965	IDO-14663
Quarter, July - September, 1965	IDO-14667
Quarter, October - December, 1965	IDO-14676
Quarter, January - March, 1966	IDO-14678
Quarter, April - June, 1966	IDO-14681
Quarter, July - September, 1966	IN - 1064
Quarter, October - December, 1966	IN - 1113
Quarter, January - March, 1967	IN - 1157 Rev
Quarter, April - June, 1967	IN - 1178

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BURNUP DETERMINATION OF NUCLEAR FUELS

INTRODUCTION

The objective of this project is the development of methods for the accurate determination of burnup of nuclear fuels. The emphasis is on power reactor fuels having long residence time and high burnup rather than on short term, experimental irradiations. The principal measurement technique under study is chemical analysis for fission products. The use of long-lived, radiochemically determined fission products is not applicable because the errors of the in-pile decay correction are magnified with long-term irradiations, the nuclear constants are not known with sufficient accuracy, and the few available nuclides, Cs-137 and Sr-90, are volatile. The use of stable fission product nuclides, determined by isotope dilution mass spectrometry, has therefore been selected. The degree of reliability of a burnup determination based on stable fission product analysis depends on three factors; the accuracy of the values used for the fission yields, the linearity between the number of fissions and the number of fission product atoms present at the end of the irradiation, and the reliability of the analysis methods.

Specific goals of this project are the accurate measurement of the fission yields of selected nuclides for various fissioning isotopes, the selection of fission product nuclides for use as fission monitors for various fuel types, and the development of separation and mass spectrometric analysis methods for these nuclides. Presently, experiments are in progress to measure the absolute thermal fission yields for U-233, U-235, Pu-239, and Pu-241 to a high degree of accuracy. Later in the program, fast fission yields will be measured.

Initial values for many U-233 and U-235 thermal fission yields have been reported in previous reports IDO-14660, IDO-14663, IDO-14667, IDO-14676, IDO-14678, IDO-14681, IN-1064, IN-1113, IN-1157 Rev, and IN-1178.

ABSOLUTE Pu-239 THERMAL FISSION YIELDS FOR THE STABLE AND
LONG-LIVED FISSION PRODUCTS OF BARIUM, CERIUM, CESIUM,
KRYPTON, NEODYMIUM, RUBIDIUM, STRONTIUM, XENON, AND ZIRCONIUM

The major goal of the burnup project is the accurate determination of the thermal fission yields of long-lived and stable fission products. A series of capsules containing weighed amounts of the oxides of highly enriched U-233, U-235, Pu-239, and Pu-241 was prepared and irradiated in the MTR (1). During this quarter, the measurement of the absolute thermal fission yields of long-lived and stable isotopes of barium, cerium, cesium, krypton, neodymium, rubidium, strontium, xenon, and zirconium was completed for three Pu-239 capsules.

Irradiation History of Capsules

The Pu-239 standard burnup capsules, which contained weighed amounts of highly enriched PuO₂ blended with high-purity aluminum powder and weld-sealed in aluminum tubes, were irradiated in the MTR at a position where the unperturbed flux was 6×10^{13} n/cm²/sec and the cadmium ratio for cobalt was 15. The irradiation histories of these capsules are summarized in Table I.

Table I

COMPOSITION AND IRRADIATION HISTORY OF STANDARD
Pu-239 BURNUP CAPSULES

<u>Capsule</u>	<u>Mg PuO₂ in Capsule</u>	<u>MTR Cycles</u>	<u>Estimated Mwd</u>	<u>% Burnup</u> ^[a]
6-2-5	48.9±0.3	8	4994	37.68±0.66
6-2-6	48.8±0.3	8	4994	36.45±0.58
6-5-5	95.0±0.2	2	1290	10.01±0.22

[a] Defined as atoms of major isotope fissioned per 100 initial atoms of major isotope.

Dissolution of Capsules (W. A. Emel, R. E. Foster, Jr., S. F. Marsh)

Dissolution of the Pu-239 capsules was carried out in a 4-in. lead-shielded facility. The dissolution apparatus has been previously described (1,2). The dissolver flask and reflux condenser were fabricated from high-purity quartz and an acid mixture of 8.4M HBr-0.3M HI, prepared from quartz-distilled reagents, was the dissolution reagent.

The dissolutions proceeded smoothly and were visually complete in approximately 10 hr. The fission gases, krypton and xenon, appeared approximately 30 min after the start of dissolution and continued for about 8 hr.

After standing overnight (under total reflux), a small amount of black residue was observed in all three dissolution solutions. The solutions were filtered (using plastic laboratory ware) through a Gelman Metrical VM-6 filter. The dissolution flask and filter were quantitatively washed with approximately 30 ml of quartz-distilled 6M HCl and the filtrate was quantitatively transferred to a calibrated 250-ml quartz flask with the aid of 8M HBr rinses. The solutions were diluted to volume with quartz-distilled 8M HBr.

The residues were individually fused with NaNO_3 -NaOH and the melts were dissolved in a mixture of 6M HCl - 0.01M HF. The residue solutions were quantitatively analyzed for plutonium and selected fission products. The plutonium content of the residue was found to be less than 0.1% of the plutonium in the dissolver solution. Of the fission products, only those of ruthenium and technetium were present in significant amounts.

Determination of Number of Fissions (R. M. Abernathey, L. W. Buttars, W. A. Emel, M. E. Kussy, F. L. Lisman, R. E. McAtee, R. A. Nielsen, F. W. Spraktes, G. D. Workman)

The accuracy of the absolute fission yield measurements directly depends on the accuracy of the determined number of fissions in the irradiated capsules. For this reason, a large effort was placed on the pre- and postirradiation analysis for the atoms of the various plutonium isotopes from which the numbers of fissions were calculated.

The PuO_2 material (obtained from ORNL) that was weighed into the capsules was analyzed for both weight percent plutonium content and isotopic distribution. For the former, samples of the PuO_2 were weighed on a micro balance and dissolved in a mixture of 9M HBr - 0.14M HF, weight aliquots were mixed with weight aliquots of standard Pu-242 spike solution, and the mixtures were analyzed by isotope dilution mass spectrometry. Isotopic distributions were determined on unspiked samples. The determined weight percent plutonium was 86.88 ± 0.04 , compared to a theoretical value for stoichiometric PuO_2 of 88.19 for plutonium of the same isotopic composition. The calculated atoms of each plutonium isotope for the three plutonium capsules are summarized in Tables II thru IV.

The postirradiation atoms of each plutonium isotope were determined in a similar manner with unspiked and spiked aliquots of the dissolver solutions. The aliquots of the dissolver solutions were delivered with calibrated 1000- μl pipets. The calculated atoms for each plutonium isotope for the three Pu-239 capsules are presented in Tables II thru IV.

Table II

PLUTONIUM ANALYSIS FOR IRRADIATED Pu-239 CAPSULE CPP 6-2-5
AND CALCULATED NUMBERS OF ATOMS

Preirradiation Data

Initial Loading 48.9 ± 0.3 mg PuO₂
mg Pu/mg Oxide 0.8688 ± 0.0004
mg Pu in Capsule 42.48 ± 0.26 mg (0.61%)

	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Isotopic distribution, wt%	99.181	0.801	0.010	0.007
Std dev _{avg} (n=3)	0.001	0.001	0.0008	0.001
Number of atoms	1.0615×10^{20}	8.5403×10^{17}	1.2175×10^{16} [a]	7.4915×10^{15}
Std dev	0.0065×10^{20}	0.0529×10^{17}	0.0974×10^{16}	1.0713×10^{15}
Total atoms of plutonium	$(1.0702 \pm 0.0065) \times 10^{20}$ (0.61%)			

Postirradiation Data

Total atoms of Pu-239 in dissolver solution = $(4.8851 \pm 0.0068) \times 10^{19}$

	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Isotopic (atom %)	73.975	22.137	3.467	0.421
Std dev _{avg} (n=4)	0.013	0.010	0.003	0.001
Number of atoms	4.8851×10^{19}	1.4518×10^{19}	2.2816×10^{18}	2.7736×10^{17}
Std dev	0.0068×10^{19}	0.0045×10^{19}	0.0105×10^{18}	0.0064×10^{17}
Number of atoms in fused residue	6.3831×10^{16}	1.9281×10^{16}	2.9259×10^{15}	3.5010×10^{14}
Total atoms	4.8915×10^{19}	1.4537×10^{19}	2.5951×10^{18} [b]	2.7771×10^{17}
Atoms of plutonium lost other than Pu-239 fission	$(0.0693 \pm 0.0069) \times 10^{19}$ [c]			

Table II (Cont)

	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Total atoms of plutonium cor- rected for loss by processes other than Pu-239 fission		(6.7018±0.0107)×10 ¹⁹ (0.16%)		

[a] Corrected to reactor insertion date.

[b] Corrected to time of discharge from reactor.

[c] Assumes a 10% uncertainty in the calculated loss of plutonium.

Table III

PLUTONIUM ANALYSIS FOR IRRADIATED Pu-239 CAPSULE CPP 6-2-6
AND CALCULATED NUMBERS OF ATOMS

Preirradiation Data

	Initial Loading	56.2±0.3 mg PuO ₂		
	mg Pu/mg Oxide	0.8688±0.0004		
	mg Pu in Capsule	48.83±0.26 mg (0.53%)		
	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Isotopic distribution, wt%	99.181	0.801	0.010	0.007
Std dev _{avg} (n=3)	0.001	0.001	0.000 ₈	0.001
Number of atoms	1.2201x10 ²⁰	9.8163x10 ¹⁷	1.3994x10 ¹⁶ [a]	8.6108x10 ¹⁵
Std dev	0.0065x10 ²⁰	0.0530x10 ¹⁷	0.1119x10 ¹⁶	1.2313x10 ¹⁵
Total atoms of plutonium	(1.2301±0.0065)x10 ²⁰ (0.53%)			

Postirradiation Data

Total atoms of Pu-239 in dissolver solution = (5.7667±0.0127)x10¹⁹

	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Isotopic (atom %)	74.502	21.735	3.357	0.405
Std dev _{avg} (n=6)	0.017	0.015	0.005	0.003
Number of atoms	5.7667x10 ¹⁹	1.6824x10 ¹⁹	2.5984x10 ¹⁸	3.1348x10 ¹⁷
Std dev	0.0127x10 ¹⁹	0.0039x10 ¹⁹	0.0070x10 ¹⁸	0.0241x10 ¹⁷
Number of atoms in fused residue	6.1177x10 ¹⁶	1.7848x10 ¹⁶	2.7563x10 ¹⁵	3.3256x10 ¹⁴
Total atoms	5.7728x10 ¹⁹	1.6842x10 ¹⁹	2.9190x10 ¹⁸ [b]	3.1301x10 ¹⁷
Atoms of plutonium lost other than Pu-239 fission	(0.0734±0.0073)x10 ¹⁹ [c]			

Table III (Cont)

	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Total atoms of plutonium cor- rected for loss by processes other than Pu-239 fission				
				(7.8537±0.0152)×10 ¹⁹ (0.19%)

[a] Corrected to reactor insertion date.

[b] Corrected to time of discharge from reactor

[c] Assumes a 10% uncertainty in the calculated loss of plutonium.

Table IV

PLUTONIUM ANALYSIS FOR IRRADIATED Pu-239 CAPSULE CPP 6-5-5
AND CALCULATED NUMBERS OF ATOMS

Preirradiation Data

	Initial Loading	95.0±0.2 mg PuO ₂		
	mg Pu/mg. Oxide	0.8688±0.0004		
	mg Pu in Capsule	82.57±0.17 mg (0.21%)		
	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Isotopic distribution, wt%	99.181	0.801	0.010	0.007
Std dev _{avg} (n=3)	0.001	0.001	0.0008	0.001
Number of atoms	2.0632x10 ²⁰	1.6599x10 ¹⁸	2.3664x10 ¹⁶ [a]	1.4561x10 ¹⁶
Std dev	0.0043x10 ²⁰	0.0040x10 ¹⁸	0.1893x10 ¹⁶	0.2067x10 ¹⁶
Total atoms of plutonium	(2.0802±0.0044)x10 ²⁰ (0.21%)			

Postirradiation Data

Total atoms of Pu-239 in dissolver solution = (1.7589±0.0013)x10 ²⁰				
	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Isotopic (atom %)	94.375	5.373	0.235	0.010
Std dev _{avg} (n=4)	0.005	0.0025	0.0017	0.001
Number of atoms	1.7589x10 ²⁰	1.0014x10 ¹⁹	4.3799x10 ¹⁷	1.8636x10 ¹⁶
Std dev	0.0013x10 ²⁰	0.0008x10 ¹⁹	0.0320x10 ¹⁷	0.1864x10 ¹⁶
Number of atoms in fused residue	8.7672x10 ¹⁷	4.9914x10 ¹⁶	2.1831x10 ¹⁵	9.2897x10 ¹³
Total atoms	1.7677x10 ²⁰	1.0064x10 ¹⁹	4.9647x10 ¹⁷ [b]	1.8729x10 ¹⁶
Atoms of plutonium lost other than Pu-239 fission	(0.00024±0.00002)x10 ²⁰ [c]			

Table IV (Cont)

	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
Total atoms of plutonium cor- rected for loss by processes other than Pu-239 fission		(1.8737±0.0013)×10 ²⁰	(0.07%)	

[a] Corrected to reactor insertion date.

[b] Corrected to time of discharge from reactor.

[c] Assumes a 10% uncertainty in the calculated loss of plutonium.

The number of Pu-239 fissions was computed by the relationship:

$$F_9 = \text{Pu}^{\circ} - \text{Pu}$$

in which:

F_9 = number of Pu-239 fissions

Pu° = total preirradiated atoms of plutonium

Pu = total postirradiated atoms of plutonium.

To accurately use this relationship, one must account for those atoms of plutonium lost by fission and decay of Pu-241 and neutron capture of Pu-242. This was done with data obtained from a computer program which simulated the history of the irradiation. A schematic diagram of the program, along with the effective cross sections used, is given in Figure 1. It is of interest to note that the loss of plutonium by Pu-241 fission and decay and Pu-242 capture amounted to only 1.04% or less of that measured at the end of the irradiation. This provides a considerable margin in calculating the loss of plutonium inasmuch as a 10% error in the calculated atoms of plutonium lost would contribute approximately 0.1% error in the total atoms of postirradiation plutonium.

The number of fissions, the atom percent Pu-239 fissioned (defined as atoms of Pu-239 fissioned per 100 initial atoms of Pu-239), and the calculated ratio of Pu-241 fissions to Pu-239 fissions for the three Pu-239 capsules are given in Table V. The quoted errors are the propagated standard deviations.

Capture-to-Fission Ratio (F. L. Lisman, F. W. Spraktes)

The effective capture-to-fission ratio, $\hat{\alpha}_9$, is readily calculated from the relationship:

$$\hat{\alpha}_9 = \frac{(N_0 + N_1 + N_2 + Y) - (N_0^{\circ} + N_1^{\circ} + N_2^{\circ})}{\text{Number of Pu-239 fissions}}$$

where:

N_0, N_0° = post and preirradiation atoms of Pu-240, respectively

N_1, N_1° = post and preirradiation atoms of Pu-241, respectively

N_2, N_2° = post and preirradiation atoms of Pu-242, respectively

Y = atoms of Pu-241 lost by fission and decay plus those atoms of Pu-242 lost by neutron capture.

Figure 1. COMPUTER PROGRAM. SCHEMATIC FOR BUILDUP AND BURNOUT OF PLUTONIUM ISOTOPES

Table V

NUMBER OF FISSIONS AND ATOM PERCENT FISSION FOR Pu-239 CAPSULES

	<u>Capsule 6-2-5</u>	<u>Capsule 6-2-6</u>	<u>Capsule 6-5-5</u>
1. Initial total plutonium (Pu ^o)	(1.0702±0.0065)x10 ²⁰	(1.2301±0.0065)x10 ²⁰	(2.0802±0.0044)x10 ²⁰
2. Post total plutonium corrected for losses other than Pu-239 fission (Pu)	(0.6702±0.0010 ₇)x10 ²⁰	(0.7854±0.0015)x10 ²⁰	(1.8737±0.0013)x10 ²⁰
3. Number of Pu-239 fissions (1-2)	(4.000±0.066)x10 ¹⁹	(4.447±0.066)x10 ¹⁹	(2.065±0.046)x10 ¹⁹
4. Initial Pu-239 atoms	(1.0615±0.0065)x10 ²⁰	(1.2201±0.0065)x10 ²⁰	(2.0632±0.0043)x10 ²⁰
5. Atom % Pu-239 fission (3÷4)x100	37.58±0.66	36.45±0.58	10.01±0.22
6. Calculated number of Pu-241 fissions	0.0665x10 ¹⁹	0.0703x10 ¹⁹	0.0023x10 ¹⁹
7. $\frac{\text{Pu-241 fissions}}{\text{Pu-239 fissions}}$ (6÷3)	0.0166	0.0158	0.0011

As stated in the previous section, the atoms of Pu-241 lost by fission and decay and the atoms of Pu-242 lost by neutron capture were obtained from a computer calculation. The values of $\hat{\alpha}_g$ calculated for the three Pu-239 capsules are summarized in Table VI. The quoted errors are the propagated standard deviation.

Fission Yield of Barium-138 (R. M. Abernathey, L. W. Buttars, R. E. Foster, Jr., M. E. Kussy, F. L. Lisman, R. A. Nielsen, F. W. Spraktes)

The absolute thermal fission yield of Ba-138 was determined on each of the three Pu-239 capsules. Four aliquots from each dissolver solution, each spiked with a known amount of Ba-138, were chemically separated and analyzed by mass spectrometry. The chemical separation and mass spectrometric procedures have been previously reported (3).

The analysis results and calculated absolute thermal fission yields are summarized in Table VII. The method of calculation and the error propagation for the determined number of Ba-138 atoms have been previously reported.

The accurate determination of absolute fission yields for samples which have undergone long-term irradiations requires correction for the buildup and burnout of the various fission products due to neutron capture. In the case of highly enriched Pu-239 samples that have undergone long-term irradiation, corrections also are necessary for fission product contributions from Pu-241 fission.

The method of correcting for buildup and burnout of the fission products is based on a series of computer programs which simulate the irradiation history of the sample. These programs yield the number of individual fission product atoms formed during the irradiation and their manner of formation. Details of these programs have been previously reported (3). The correction for fission product contributions from Pu-241 fission was obtained from the calculated ratio of Pu-241 to Pu-239 fissions (see Table V) and the reported heavy mass Pu-241 fission yields of Farrar et al (4) and the estimated light mass yields reported by Walker (5).

In the case of Ba-138, correction for buildup and burnout by neutron capture is small, being about 0.01%. Corrections for the Pu-241 fission contribution resulted in a 2.0% correction for capsules CPP 6-2-5 and CPP 6-2-6, and a 0.2% correction for capsule CPP 6-5-5.

Fission Yields of Cerium and Neodymium Isotopes (R. M. Abernathey, L. W. Buttars, W. A. Emel, F. L. Lisman, R. E. McAtee, R. A. Nielsen, F. W. Spraktes, G. D. Workman)

The absolute thermal fission yields of the long-lived and stable isotopes of cerium and neodymium were determined on each of the three dissolved Pu-239 capsules. Four aliquots from each dissolver solution, each spiked with known amounts of Ce-136 and Nd-150, and two aliquots without spike were chemically separated and analyzed by mass spectrometry. The preparation and standardization of the spike solutions have been

Table VI

CALCULATED EFFECTIVE CAPTURE-TO-FISSION RATIO FOR Pu-239

		Capsule 6-2-5	Capsule 6-2-6	Capsule 6-5-5
1.	N_0	$(1.4537 \pm 0.0045) \times 10^{19}$	$(1.6842 \pm 0.0039) \times 10^{19}$	$(1.0064 \pm 0.0008) \times 10^{19}$
2.	N_1	$(0.2595 \pm 0.0012) \times 10^{19}$	$(0.2919 \pm 0.0008) \times 10^{19}$	$(0.0496 \pm 0.0004) \times 10^{19}$
3.	N_2	$(0.0278 \pm 0.0001) \times 10^{19}$	$(0.0314 \pm 0.0002) \times 10^{19}$	$(0.0019 \pm 0.0002) \times 10^{19}$
4.	Y	$(0.0693 \pm 0.0069) \times 10^{19}$	$(0.0734 \pm 0.0073) \times 10^{19}$	$(0.0024 \pm 0.0002) \times 10^{19}$
5.	N_0^o	$(0.0854 \pm 0.0005) \times 10^{19}$	$(0.0982 \pm 0.0005) \times 10^{19}$	$(0.1660 \pm 0.0004) \times 10^{19}$
6.	N_1^o	$(0.0012 \pm 0.0001) \times 10^{19}$	$(0.0014 \pm 0.0001) \times 10^{19}$	$(0.0024 \pm 0.0002) \times 10^{19}$
7.	N_2^o	$(0.0007 \pm 0.0001) \times 10^{19}$	$(0.0008 \pm 0.0001) \times 10^{19}$	$(0.0015 \pm 0.0002) \times 10^{19}$
8.	Number of Pu-239 Fissions	$(4.000 \pm 0.066) \times 10^{19}$	$(4.447 \pm 0.066) \times 10^{19}$	$(2.065 \pm 0.046) \times 10^{19}$
	$\hat{\alpha}_9 = \frac{\Sigma(1 \text{ thru } 4) - \Sigma(5 \text{ thru } 7)}{(8)}$	(0.4307 ± 0.0074)	(0.4453 ± 0.0068)	(0.4312 ± 0.0096)
		$\hat{\alpha}_{9 \text{ avg}} = (0.4370 \pm 0.0044)^{[a]}$		

[a] $\hat{\alpha}_{9 \text{ avg}}$ was obtained by weighting the individual values as the reciprocal of their variances.

Table VII

THERMAL FISSION YIELD OF Ba-138 FOR Pu-239

	<u>Capsule 6-2-5</u>	<u>Capsule 6-2-6</u>	<u>Capsule 6-5-5</u>
Determined number of atoms	2.1445×10^{18}	2.3807×10^{18}	1.0809×10^{18}
Std dev _{avg} (n=4)	0.0080×10^{18}	0.0107×10^{18}	0.0051×10^{18}
Atoms after correction for neutron capture and decay	2.1448×10^{18}	2.3810×10^{18}	1.0809×10^{18}
Std dev, %, includes estimated uncertainties in correction factors for neutron capture	0.37	0.45	0.47
Number of Pu-239 fissions	$(4.000 \pm 0.066) \times 10^{19}$	$(4.447 \pm 0.066) \times 10^{19}$	$(2.065 \pm 0.046) \times 10^{19}$
Measured fission yield (%)	5.36 ± 0.09	5.35 ± 0.08	5.23 ± 0.12
$\frac{\text{Pu-241 fissions}}{\text{Pu-239 fissions}}$ (calculated)	0.0166	0.0158	0.0011
Assumed Pu-241 fission yield	6.83	6.83	6.83
Pu-239 fission yield (%)	5.25	5.24	5.22
Std dev	0.09	0.08	0.12

described previously (1). A three-column sequential procedure, similar to that used for the uranium capsules (1), was used for the separation of the individual lanthanides. The first column separated the lanthanides from the actinides and most fission products. Cerium is separated from the other lanthanides on a second column. The remaining lanthanides are chromatographically separated on a third column with α -hydroxyisobutyric acid at controlled pH as the elutriant. Details of the procedure are given in Appendix I.

The analysis results and calculated Pu-239 fission yields for cerium are summarized in Tables VIII thru X and for neodymium in Tables XI thru XIII.

As in the case of the thermal fission yield of Ba-138, corrections for the buildup and burnout of the cerium and neodymium isotopes by neutron capture and radioactive decay and the Pu-241 fission contribution were based on computer programs which simulate the irradiation histories of the capsules. Details of these programs have been previously reported (6).

The yield of the mass 144 chain was calculated three different ways; from the neodymium analysis correcting for the holdup at Ce-144, from the cerium analysis correcting for the decay of Ce-144, and from the sum of the measured Ce-144 and Nd-144 atoms. These values are summarized in Table XIV.

Fission Yield of Cs-133 and Cs-137 for Pu-239 (R. M. Abernathey, L. W. Buttars, R. E. Foster, Jr., M. E. Kussy, F. L. Lisman, R. A. Nielsen, R. E. McAtee, F. W. Spraktes, G. D. Workman)

The absolute thermal fission yields of Cs-133 and Cs-137 were determined on each of the three irradiated Pu-239 capsules. Eight aliquots from each dissolver solution, four spiked with known amounts of Cs-133 and four without spike, were chemically separated and analyzed by mass spectrometry. The chemical separation and mass spectrometric procedures have been previously reported (7).

The analysis results and calculated fission yields are summarized in Table XV for Cs-133 and in Table XVI for Cs-137.

As in the case of the thermal fission yield of Ba-138, corrections for the buildup and burnout of fission product cesium by neutron capture and radioactive decay and the Pu-241 fission contribution were based on computer programmed calculations. Details of these programs have been previously reported (7).

Fission Yields of Krypton and Xenon For Pu-239 (R. M. Abernathey, L. L. Dickerson, R. E. Foster, Jr., F. L. Lisman, F. W. Spraktes)

These two gases were trapped from the dissolver off-gas on Molecular Sieve 5A at -190°C. The total concentration of each gas was determined by temperature programmed gas chromatography. The separated gas fractions from the chromatographic column were collected on charcoal

Table VIII

THERMAL FISSION YIELDS OF THE LONG-LIVED AND STABLE ISOTOPES
OF CERIUM FOR Pu-239 FROM CAPSULE 6-2-5

	Ce-140	Ce-142	Ce-144
Determined number of atoms	2.1790×10^{18}	1.9619×10^{18}	1.2223×10^{17}
Std dev _{avg} (n=4)	0.0144×10^{18}	0.0074×10^{18}	0.0155×10^{17}
Atoms after correction for neutron capture and radio- active decay	2.1724×10^{18}	1.9547×10^{18}	1.5363×10^{18} [a]
Std dev, % (includes esti- mated uncertainties in cor- rection factors for neutron capture and radioactive decay)	0.72	0.48	2.4
Number of Pu-239 fissions		$(4.000 \pm 0.066) \times 10^{19}$	
Measured fission yield (%)	5.43 ± 0.10	4.88 ± 0.08	3.84 ± 0.11
$\frac{\text{Pu-241 fissions}}{\text{Pu-239 fissions}}$ (calculated)		0.0166	
Assumed Pu-241 fission yield (%)	5.78	4.70	4.07
Pu-239 fission yield (%)	5.33	4.81	3.77
Std dev	0.10	0.08	0.11
[a] Includes in-pile and out-pile decay corrections.			

Table IX

THERMAL FISSION YIELDS OF THE LONG-LIVED AND STABLE ISOTOPES
OF CERIUM FOR Pu-239 FROM CAPSULE 6-2-6

	Ce-140	Ce-142	Ce-144
Determined number of atoms	2.4964×10^{18}	2.2437×10^{18}	1.2308×10^{17}
Std dev _{avg} (n=3) ^[b]	0.0062×10^{18}	0.0047×10^{18}	0.0053×10^{17}
Atoms after correction for neutron capture and radio- active decay	2.4892×10^{18}	2.2358×10^{18}	1.6188×10^{18} ^[a]
Std dev, % (includes esti- mated uncertainties in cor- rection factors for neutron capture and radioactive decay)	0.39	0.37	2.0
Number of Pu-239 fissions	$(4.447 \pm 0.066) \times 10^{19}$		
Measured fission yield (%)	5.59 ± 0.08	5.03 ± 0.08	3.64 ± 0.09
$\frac{\text{Pu-241 fissions}}{\text{Pu-239 fissions}}$ (calculated)	0.0158		
Assumed Pu-241 fission yield (%)	5.78	4.70	4.07
Pu-239 fission yield (%)	5.50	4.95	3.58
Std dev	0.08	0.08	0.09

[a] Includes in-pile and cut-pile decay corrections.

[b] One sample was inadvertently lost in the chemical separation procedure.

Table X

THERMAL FISSION YIELDS OF THE LONG-LIVED AND STABLE ISOTOPES
OF CERIUM FOR Pu-239 FROM CAPSULE 6-5-5

	Ce-140	Ce-142	Ce-144
Determined number of atoms	1.1313×10^{18}	1.0169×10^{18}	8.1477×10^{16}
Std dev _{avg} (n=5)	0.0033×10^{18}	0.0021×10^{18}	0.1189
Atoms after correction for neutron capture and radio- active decay	1.1306×10^{18}	1.0155×10^{18}	7.4750×10^{17} [a]
Std dev, % (includes esti- mated uncertainties in cor- rection factors for neutron capture and radioactive decay)	0.29	0.21	2.5
Number of Pu-239 fissions		$(2.065 \pm 0.046) \times 10^{19}$	
Measured fission yield (%)	5.47 ± 0.12	4.92 ± 0.11	3.62 ± 0.12
$\frac{\text{Pu-241 fissions}}{\text{Pu-239 fissions}}$ (calculated)		0.0011	
Assumed Pu-241 fission yield (%)	5.78	4.70	4.07
Pu-239 fission yield (%)	5.47	4.91	3.62
Std dev	0.12	0.11	0.12
[a] Includes in-pile and out-pile decay corrections.			

Table XI

THERMAL FISSION YIELDS OF STABLE NEODYMIUM ISOTOPIES
FOR Pu-239 FROM CAPSULE 6-2-5

	Nd-143	Nd-144	Nd-145	Nd-146	Nd-148	Nd-150
Determined number of atoms	1.5973×10^{18}	1.5246×10^{18}	1.1620×10^{18}	9.9713×10^{17}	6.6843×10^{17}	3.7607×10^{17}
Std dev _{avg} (n=4)	0.0062×10^{18}	0.0061×10^{18}	0.0048×10^{18}	0.0369×10^{17}	0.0280×10^{17}	0.0147×10^{17}
Atoms after correction for neutron capture and radioactive decay	1.7533×10^{18}	1.4578×10^{18}	1.1849×10^{18}	9.7464×10^{17}	6.6665×10^{17}	3.7626×10^{17}
Std dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	1.1	0.50	0.51	0.48	0.42	0.39
Number of Pu-239 fissions	$(4.000 \pm 0.066) \times 10^{19}$					
Measured fission yield (%)	4.38 ± 0.09	3.64 ± 0.06	2.96 ± 0.05	2.44 ± 0.04	1.67 ± 0.03	0.940 ± 0.016
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)	0.0166					
Assumed Pu-241 fission yield (%)	4.44	4.07	3.16	2.71	1.91	1.24
Pu-239 fission yield (%)	4.31	3.57	2.91	2.39	1.63	0.920
Std dev	0.08	0.06	0.05	0.04	0.03	0.016

Table XII

THERMAL FISSION YIELDS OF STABLE NEODYMIUM ISOTOPES
FOR Pu-239 FROM CAPSULE 6-2-6

	Nd-143	Nd-144	Nd-145	Nd-146	Nd-148	Nd-150
Determined number of atoms	1.8360×10^{18}	1.7556×10^{18}	1.3382×10^{18}	1.1450×10^{18}	7.6713×10^{17}	4.3512×10^{17}
Std dev _{avg} (n=3) ^[a]	0.0112×10^{18}	0.0107×10^{18}	0.0076×10^{18}	0.0069×10^{18}	0.0476×10^{17}	0.0261×10^{17}
Atoms after correction for neutron capture and radioactive decay	2.0082×10^{18}	1.6774×10^{18}	1.3636×10^{18}	1.1201×10^{18}	7.6516×10^{17}	4.3533×10^{17}
Std dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	1.17	0.68	0.64	0.67	0.62	0.60
Number of Pu-239 fissions	$(4.447 \pm 0.066) \times 10^{19}$					
Measured fission yield (%)	4.52 ± 0.08	3.77 ± 0.06	3.07 ± 0.05	2.52 ± 0.04	1.72 ± 0.03	0.979 ± 0.016
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)	0.0158					
Assumed Pu-241 fission yield (%)	4.44	4.07	3.16	2.71	1.91	1.24
Pu-239 fission yield (%)	4.44	3.71	3.02	2.47	1.69	0.959
Std dev	0.08	0.06	0.05	0.04	0.03	0.015

[a] One sample was inadvertently lost in the chemical separation procedure.

Table XIII

THERMAL FISSION YIELDS OF STABLE NEODYMIUM ISOTOPES FOR Pu-239 FROM CAPSULE 6-5-5

	Nd-143	Nd-144	Nd-145	Nd-146	Nd-148	Nd-150
Determined number of atoms	8.8260×10^{17}	6.8439×10^{17}	6.0358×10^{17}	4.9979×10^{17}	3.3776×10^{17}	1.9418×10^{17}
Std dev _{avg} (n=4)	0.0326×10^{17}	0.0267×10^{17}	0.0223×10^{17}	0.0185×10^{17}	0.0125×10^{17}	0.0072×10^{17}
Atoms after correction for neutron capture and radioactive decay	8.9204×10^{17}	7.5775×10^{17}	6.0575×10^{17}	4.9769×10^{17}	3.3712×10^{17}	1.9418×10^{17}
Std dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.48	1.07	0.48	0.48	0.48	0.37
Number of Pu-239 fissions	$(2.065 \pm 0.046) \times 10^{19}$					
Measured fission yield (%)	4.32 ± 0.10	3.67 ± 0.09	2.93 ± 0.06	2.41 ± 0.05	1.63 ± 0.04	0.940 ± 0.021
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)	0.0011					
Assumed Pu-241 fission yield (%)	4.44	4.07	3.16	2.71	1.91	1.24
Pu-239 fission yield (%)	4.31	3.66	2.93	2.41	1.63	0.940
Std dev	0.10	0.09	0.06	0.05	0.04	0.021

Table XIV

THERMAL FISSION YIELD OF MASS 144 FOR Pu-239

	<u>Capsule 6-2-5</u>	<u>Capsule 6-2-6</u>	<u>Capsule 6-5-5</u>
1. Determined atoms of Ce-144[a]	$(0.1222 \pm 0.0015) \times 10^{18}$	$(0.1261 \pm 0.0005) \times 10^{18}$	$(0.8348 \pm 0.0122) \times 10^{17}$
2. Determined atoms of Nd-144[a]	$(1.5246 \pm 0.0061) \times 10^{18}$	$(1.7556 \pm 0.0086) \times 10^{18}$	$(6.8439 \pm 0.0267) \times 10^{17}$
3. Total number of 144 atoms (1 + 2)	$(1.6468 \pm 0.0063) \times 10^{18}$	$(1.8817 \pm 0.0086) \times 10^{18}$	$(7.6787 \pm 0.0293) \times 10^{17}$
4. Total number of 144 atoms corrected for buildup and burnout by neutron capture	$(1.4651 \pm 0.0157) \times 10^{18}$	$(1.6807 \pm 0.0185) \times 10^{18}$	$(7.5661 \pm 0.0289) \times 10^{17}$
5. Number of Pu-239 fissions	$(4.000 \pm 0.066) \times 10^{19}$	$(4.447 \pm 0.066) \times 10^{19}$	$(2.065 \pm 0.046) \times 10^{19}$
6. Measured fission yield % [(4)x100/(5)]	3.66±0.07	3.78±0.07	3.66±0.08
7. <u>Pu-241 fissions</u> Pu-239 fissions (calculated)	0.0166	0.0158	0.0011
8. Assumed Pu-241 fission yield (%)	4.07	4.07	4.07
Pu-239 fission yield (%) [6-(7)(8)]	3.59	3.71	3.66
Std dev	0.07	0.07	0.08
[a] The determined atoms of Ce-144 and Nd-144 have been normalized to the same date.			

Table XV

THERMAL FISSION YIELD OF Cs-133 FOR Pu-239

	Capsule 6-2-5	Capsule 6-2-6	Capsule 6-5-5
Determined number of atoms	$(2.7170 \pm 0.0149) \times 10^{18}$	$(3.1548 \pm 0.0170) \times 10^{18}$	$(1.4377 \pm 0.0032) \times 10^{18}$
Atoms after correction for neutron capture	$(2.7551 \pm 0.0173) \times 10^{18}$	$(3.1975 \pm 0.0197) \times 10^{18}$	$(1.4443 \pm 0.0053) \times 10^{18}$
Number of Pu-239 fissions	$(4.000 \pm 0.066) \times 10^{19}$	$(4.447 \pm 0.066) \times 10^{19}$	$(2.065 \pm 0.046) \times 10^{19}$
Measured fission yield (%)	6.89 $\pm$ 0.12	7.19 $\pm$ 0.12	6.99 $\pm$ 0.15
$\frac{\text{Pu-241 fissions}}{\text{Pu-239 fissions}}$ (calculated)	0.0166	0.0158	0.0011
Assumed Pu-241 fission yield (%)	6.56	6.56	6.56
Pu-239 fission yield (%)	6.78	7.08	6.99
Std dev	0.12	0.11	0.15

Table XVI

THERMAL FISSION YIELD OF Cs-137 FOR Pu-239

	Capsule 6-2-5	Capsule 6-2-6	Capsule 6-5-5
Determined number of atoms	$(2.4530 \pm 0.0134) \times 10^{18}$	$(2.7983 \pm 0.0066) \times 10^{18}$	$(1.2598 \pm 0.0023) \times 10^{18}$
Atoms after correction for neutron capture and radioactive decay ^[a]	$(2.6323 \pm 0.0145) \times 10^{18}$	$(3.0020 \pm 0.0072) \times 10^{18}$	$(1.3468 \pm 0.0024) \times 10^{18}$
Number of Pu-239 fissions	$(4.000 \pm 0.066) \times 10^{19}$	$(4.447 \pm 0.066) \times 10^{19}$	$(2.065 \pm 0.046) \times 10^{19}$
Measured fission yield (%)	6.58 $\pm$ 0.11	6.75 $\pm$ 0.10	6.53 $\pm$ 0.14
$\frac{\text{Pu-241 fissions}}{\text{Pu-239 fissions}}$ (calculated)	0.0166	0.0158	0.0011
Assumed Pu-241 fission yield (%)	6.62	6.62	6.62
Pu-239 fission yield (%)	6.47	6.64	6.52
Std dev	0.11	0.10	0.14

^[a] Corrected for in-pile decay.

at -80°C and transferred to the mass spectroscopy laboratory for isotopic distribution analysis. Details of the apparatus and the procedure for collection and analysis have been reported (1).

The analysis results and calculated absolute thermal fission yields are summarized in Tables XVII, XVIII, and XIX for krypton and in Tables XX, XXI, and XXII for xenon. The error associated with each value is the standard deviation.

Corrections for buildup and burnout of the various isotopes by neutron capture and radioactive decay were made using the previously described computer programs (6).

Fission Yields of Rb-85 and Rb-87 for Pu-239 (R. M. Abernathey, L. W. Buttars, R. E. Foster, Jr., M. E. Kussy, F. L. Lisman, R. A. Nielsen, R. E. McAtee, F. W. Spraktes, G. D. Workman)

The absolute thermal fission yields of Rb-85 and Rb-87 were determined by isotope dilution mass spectrometry. Eight aliquots from each dissolver solution, four spiked with a known amount of Rb-85 and four without spike, were chemically separated and analyzed by mass spectrometry. The chemical separation and mass spectrometric procedures have been previously reported (3).

The analysis results and calculated fission yields are summarized in Table XXIII. The indicated errors are the propagated standard deviation of the mean value. The method of calculation and the error propagation for the determined number of Rb-85 and Rb-87 atoms have been reported (3).

To calculate the yield of the 85 mass chain, the number of determined Rb-85 atoms was added to the determined atoms of 10.76-yr Kr-85.

Corrections for the buildup and burnout by neutron capture, radioactive decay, and the Pu-241 fission contribution for fission product rubidium were made with the aid of computer programed calculations. Details of these programs have been previously reported (3).

Fission Yields of Sr-88 and Sr-90 for Pu-239 (R. M. Abernathey, L. W. Buttars, R. E. Foster, Jr., M. E. Kussy, F. L. Lisman, R. A. Nielsen, R. E. McAtee, F. W. Spraktes, G. D. Workman)

The absolute thermal fission yields of Sr-88 and Sr-90 were determined by isotope dilution mass spectrometry for standard burnup capsules CPP 6-2-5 and CPP 6-2-6. Eight aliquots from each dissolver solution, four spiked with a known amount of Sr-87 and four without spike, were chemically separated and then analyzed by mass spectrometry. The chemical separation and mass spectrometric procedure have been reported (3).

Table XVII

THERMAL FISSION YIELDS OF KRYPTON ISOTOPES FOR Pu-239 FROM CAPSULE 6-2-5

std cc Kr = 0.0233±0.0004₇

	<u>Kr-83</u>	<u>Kr-84</u>	<u>Kr-85 (10-yr)</u>	<u>Kr-86</u>
Isotopic distribution, atom %	17.07	30.15	6.70	46.08
Determined number of atoms	(1.0703±0.0214)×10 ¹⁷	(1.8897±0.0378)×10 ¹⁷	(4.2014±0.0840)×10 ¹⁶	(2.8891±0.0578)×10 ¹⁷
Atoms after correction for neutron capture and radio- active decay	(1.1382±0.0227)×10 ¹⁷	(1.8234±0.0365)×10 ¹⁷	(5.0439±0.1009)×10 ¹⁶ [a]	(2.8877±0.0577)×10 ¹⁷
Number of Pu-239 fissions		(4.000±0.066)×10 ¹⁹		
Measured fission yield (%)	0.284±0.007	0.456±0.012	0.126±0.003	0.722±0.019
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0166	
Assumed Pu-241 fission yield (%)	0.10	0.20	0.07	0.40
Pu-239 fission yield (%)	0.283	0.452	0.125	0.715
Std dev	0.007	0.012	0.003	0.019

[a] Corrected for in-pile and out-pile decay.

Table XVIII

THERMAL FISSION YIELDS OF KRYPTON ISOTOPES FOR Pu-239 FROM CAPSULE 6-2-6

std cc Kr = 0.0276 ± 0.0005

	Kr-83	Kr-84	Kr-85 (10-yr)	Kr-86
Isotopic distribution, atom %	16.83	33.17	6.56	46.45
Determined number of atoms	$(1.2489 \pm 0.0250) \times 10^{17}$	$(2.2388 \pm 0.0448) \times 10^{17}$	$(4.8678 \pm 0.0974) \times 10^{16}$	$(3.4468 \pm 0.0689) \times 10^{17}$
Atoms after correction for neutron capture and radioactive decay	$(1.3252 \pm 0.0265) \times 10^{17}$	$(2.1630 \pm 0.0432) \times 10^{17}$	$(5.7557 \pm 0.1151) \times 10^{16} [a]$	$(3.4452 \pm 0.0689) \times 10^{17}$
Number of Pu-239 fissions		$(4.447 \pm 0.066) \times 10^{19}$		
Measured fission yield (%)	0.298 ± 0.007	0.486 ± 0.012	0.129 ± 0.003	0.775 ± 0.019
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0158	
Assumed Pu-241 fission yield (%)	0.10	0.20	0.07	0.40
Pu-239 fission yield (%)	0.296	0.483	0.128	0.768
Std dev	0.007	0.012	0.003	0.019

[a] Corrected for in-pile and out-pile decay.

Table XIX

THERMAL FISSION YIELDS OF KRYPTON ISOTOPES FOR Pu-239 FROM CAPSULE 6-5-5

std cc Kr = $0.0128 \pm 0.0002_6$

	<u>Kr-83</u>	<u>Kr-84</u>	<u>Kr-85 (10 yr)</u>	<u>Kr-86</u>
Isotopic distribution, atom %	17.80	29.58	6.60	46.01
Determined number of atoms	$(6.1257 \pm 0.1225) \times 10^{16}$	$(1.0179 \pm 0.0204) \times 10^{17}$	$(2.2713 \pm 0.0454) \times 10^{16}$	$(1.5834 \pm 0.0317) \times 10^{17}$
Atoms after correction for neutron capture and radioactive decay	$(6.1950 \pm 0.1239) \times 10^{16}$	$(1.0110 \pm 0.0202) \times 10^{17}$	$(2.6556 \pm 0.0531) \times 10^{16} [a]$	$(1.5833 \pm 0.0317) \times 10^{17}$
20 Number of Pu-239 fissions			$(2.065 \pm 0.046) \times 10^{19}$	
Measured fission yield (%)	0.300 ± 0.009	0.489 ± 0.014	0.129 ± 0.004	0.767 ± 0.023
<u>Pu-241 fissions</u> <u>Pu-239 fissions</u> (calculated)			0.0011	
Assumed Pu-241 fission yield (%)	0.10	0.20	0.07	0.40
Pu-239 fission yield (%)	0.300	0.489	0.128	0.766
Std dev ,	0.009	0.014	0.004	0.023
[a] Corrected for in-pile and out-pile decay.				

Table XX

THERMAL FISSION YIELDS OF XENON ISOTOPES FOR Pu-239 FROM CAPSULE 6-2-5

std cc Xe = 0.4185 ± 0.0084

	Xe-131	Xe-132	Xe-134	Xe-136 ^[a]
Isotopic distribution atom %	12.10	18.22	25.23	44.45
Determined number of atoms	$(1.3610 \pm 0.0272) \times 10^{18}$	$(2.0501 \pm 0.0410) \times 10^{18}$	$(2.8389 \pm 0.0568) \times 10^{18}$	$(5.0015 \pm 0.1000) \times 10^{18}$
Atoms after correction for neutron capture and radioactive decay	$(1.3991 \pm 0.0280) \times 10^{18}$	$(2.0110 \pm 0.0402) \times 10^{18}$	$(2.8274 \pm 0.0565) \times 10^{18}$	
Number of Pu-239 fissions			$(4.000 \pm 0.066) \times 10^{19}$	
Measured fission yield (%)	3.50 ± 0.09	5.03 ± 0.13	7.07 ± 0.18	
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0166	
Assumed Pu-241 fission yield (%)	3.01	4.47	7.81	
Pu-239 fission yield (%)	3.45	4.95	6.94	
Std dev	0.09	0.13	0.18	

[a] No attempt was made to distinguish those atoms of Xe-136 resulting from Xe-135 (n,γ) Xe-136 reactions; hence, no Xe-136 fission yield can be reported.

Table XXI

THERMAL FISSION YIELDS OF XENON ISOTOPES FOR Pu-239 FROM CAPSULE 6-2-6

std cc Xe = 0.4848 ± 0.0097

	Xe-131	Xe-132	Xe-134	Xe-136 ^[a]
Isotopic distribution, atom %	12.11	18.25	25.28	44.36
Determined number of atoms	$(1.5784 \pm 0.0316) \times 10^{18}$	$(2.3787 \pm 0.0476) \times 10^{18}$	$(3.2950 \pm 0.0659) \times 10^{18}$	$(5.7819 \pm 0.1156) \times 10^{18}$
Atoms after correction for neutron capture and radioactive decay	$(1.6208 \pm 0.0324) \times 10^{18}$	$(2.3350 \pm 0.0467) \times 10^{18}$	$(3.2821 \pm 0.0656) \times 10^{18}$	
Number of Pu-239 fissions			$(4.447 \pm 0.066) \times 10^{19}$	
Measured fission yield (%)	3.64 ± 0.09	5.25 ± 0.13	7.38 ± 0.18	
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0158	
Assumed Pu-241 fission yield (%)	3.01	4.47	7.81	
Pu-239 fission yield (%)	3.59	5.18	7.26	
Std dev	0.09	0.13	0.18	

[a] No attempt was made to distinguish those atoms of Xe-136 resulting from Xe-135 (n,γ) Xe-136 reactions; hence, no Xe-136 fission yield can be reported.

Table XXII

THERMAL FISSION YIELDS OF XENON ISOTOPES FOR Pu-239 FROM CAPSULE 6-5-5

std cc Xe = 0.2090±0.0042

	Xe-131	Xe-132	Xe-134	Xe-136 ^[a]
Isotopic distribution, atom %	12.72	17.81	25.24	44.23
Determined number of atoms	$(7.1476 \pm 0.1429) \times 10^{18}$	$(1.0008 \pm 0.0200) \times 10^{18}$	$(1.4183 \pm 0.0284) \times 10^{18}$	$(2.4854 \pm 0.0497) \times 10^{18}$
Atoms after correction for neutron capture and radioactive decay	$(7.1783 \pm 0.1436) \times 10^{17}$	$(0.9838 \pm 0.0197) \times 10^{18}$	$(1.4139 \pm 0.0283) \times 10^{18}$	
Number of Pu-239 fissions			$(2.065 \pm 0.046) \times 10^{19}$	
Measured fission yield (%)	3.48±0.10	4.76±0.14	6.85±0.20	
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0011	
Assumed Pu-241 fission yield (%)	3.01	4.47	7.81	
Pu-239 fission yield (%)	3.47	4.76	6.84	
Std dev	0.10	0.14	0.20	

[a] No attempt was made to distinguish those atoms of Xe-136 resulting from Xe-135 (n,γ) Xe-136 reactions; hence, no Xe-136 fission yield can be reported.

Table XXIII

THERMAL FISSION YIELDS OF Rb-85 AND Rb-87 FOR Pu-239

	Capsule 6-2-5		Capsule 6-2-6		Capsule 6-5-5	
	Rb-85	Rb-87	Rb-85	Rb-87	Rb-85	Rb-87
Determined number of atoms	$1.7952 \pm 0.0223 \times 10^{17}$	$3.8850 \pm 0.0317 \times 10^{17}$	$2.0531 \pm 0.0137 \times 10^{17}$	$4.4379 \pm 0.0284 \times 10^{17}$	$9.3132 \pm 0.1066 \times 10^{16}$	$1.9895 \pm 0.0178 \times 10^{17}$
Determined atoms of 10.76-yr Kr-85[a]	$(0.4201 \pm 0.0084) \times 10^{17}$		$(0.4868 \pm 0.0097) \times 10^{17}$		$(2.2713 \pm 0.0454) \times 10^{16}$	
Total mass-85 atoms	$(2.2153 \pm 0.0238) \times 10^{17}$		$(2.5399 \pm 0.0167) \times 10^{17}$		$(1.1584 \pm 0.0116) \times 10^{17}$	
Atoms after correction for neutron capture	$2.2173 \pm 0.0238 \times 10^{17}$	$3.8852 \pm 0.0317 \times 10^{17}$	$2.5422 \pm 0.0167 \times 10^{17}$	$4.4381 \pm 0.0284 \times 10^{17}$	$1.1585 \pm 0.0116 \times 10^{17}$	$1.9895 \pm 0.0178 \times 10^{17}$
Number of Pu-239 fissions	$(4.000 \pm 0.066) \times 10^{19}$		$(4.447 \pm 0.066) \times 10^{19}$		$(2.065 \pm 0.046) \times 10^{19}$	
Measured fission yield (%)	0.554 $\pm$ 0.011	0.971 $\pm$ 0.018	0.572 $\pm$ 0.009	0.998 $\pm$ 0.016	0.561 $\pm$ 0.013	0.963 $\pm$ 0.023
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)	0.0166		0.0158		0.0011	
Assumed Pu-241 fission yield (%)	0.30	0.50	0.30	0.50	0.30	0.50
Pu-239 fission yield (%)	0.549	0.963	0.567	0.990	0.561	0.963
Std dev	0.011	0.018	0.009	0.016	0.013	0.023

[a] Not corrected for either out-pile or in-pile decay. The sum of the determined Kr-85 atoms and the determined Rb-85 atoms is the total number of mass-85 atoms, because the krypton is quantitatively removed from the dissolver solution at time of dissolution.

The analysis results and calculated fission yields are summarized in Table XXIV. The indicated errors are the propagated standard deviation of the mean value. The method of calculation and the error propagation for the determined number of Sr-88 and Sr-90 atoms have been previously reported (3).

Corrections for the buildup and burnout by neutron capture, radioactive decay, and the Pu-241 fission contribution, were made with the aid of computer programmed calculations. Details of these programs have been previously reported (3).

Fission Yields of the Stable Zirconium Isotopes for Pu-239 (R. M. Abernathy, L. W. Buttars, W. A. Emel, F. L. Lisman, R. A. Nielsen, R. E. McAtee, F. W. Spraktes, G. D. Workman)

The absolute thermal fission yields of the stable zirconium isotopes were determined on each of the three dissolved Pu-239 capsules. Eight aliquots from each dissolver solution, four spiked with known amounts of Zr-90 and four without spike, were chemically separated and analyzed by mass spectrometry. The preparation and standardization of the spike solutions has been previously described (7).

The chemical separation procedure was the thorium iodate coprecipitation method employed for the uranium capsules (7) with the addition of a plutonium cleanup step. The plutonium was removed from the zirconium by means of an anion exchange column. The sample was placed on the column from 10.3M HCl and washed with 10.3M HCl to remove plutonium and potassium. It is necessary to remove any potassium present in the sample as K₂O₂ will interfere with the Zr-94 and Zr-96 determination. The zirconium then was eluted with 8M HNO₃. The procedure is detailed in Appendix II.

The analysis results and calculated Pu-239 fission yields are summarized in Tables XXV, XXVI, and XXVII.

As in the case of the other thermal fission yields, corrections were necessary for the buildup and burnout by neutron capture and the Pu-241 fission contribution. These corrections were made with the aid of computer programmed calculations. Details of these programs have been previously reported (7).

Average Fission Yield Values for Pu-239

The fission yield values determined on each of the three Pu-239 capsules have been averaged to obtain final values. These values are summarized in Table XXVIII. The average values and associated errors were obtained by weighting the individual values as the reciprocals of their variances.

For comparison, the Pu-239 fission yield values determined by mass spectrometry by Fickel and Tomlinson (8,9) have been included in Table XXVIII.

Table XXIV

THERMAL FISSION YIELDS OF Sr-88 AND Sr-90 FOR Pu-239

	Capsule 6-2-5		Capsule 6-2-6	
	Sr-88	Sr-90	Sr-88	Sr-90
Determined number of atoms	$(5.2633 \pm 0.0562) \times 10^{17}$	$(7.5579 \pm 0.0196) \times 10^{17}$	$(5.9751 \pm 0.0717) \times 10^{17}$	$(8.5639 \pm 0.0260) \times 10^{17}$
Atoms after correction for neutron capture and radio- active decay	$(5.2624 \pm 0.0563) \times 10^{17}$	$(8.1616 \pm 0.0530) \times 10^{17} [a]$	$(5.9741 \pm 0.0717) \times 10^{17}$	$(9.3500 \pm 0.0626) \times 10^{17} [a]$
Number of Pu-239 fissions	$(4.00 \pm 0.066) \times 10^{19}$		$(4.447 \pm 0.066) \times 10^{19}$	
Measured fission yield (%)	1.32 $\pm$ 0.03	2.04 $\pm$ 0.04	1.34 $\pm$ 0.03	2.10 $\pm$ 0.03
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)	0.0166		0.0158	
Assumed Pu-241 fission yield (%)	0.70	1.40	0.70	1.40
Pu-239 fission yield (%)	1.30	2.02	1.33	2.08
Std dev	0.03	0.04	0.02	0.03

[a] Corrected for both in-pile and out-pile decay.

Table XXV

THERMAL FISSION YIELDS OF STABLE ZIRCONIUM ISOTOPES FOR Pu-239 FROM CAPSULE 6-2-5

	Zr-91	Zr-92	Zr-93	Zr-94	Zr-96
Determined number of atoms	9.7657×10^{17}	1.1974×10^{18}	1.5344×10^{18}	1.7449×10^{18}	1.9959×10^{18}
Std dev _{avg} (n=3) ^[a]	0.0410×10^{17}	0.0050×10^{18}	0.0064×10^{18}	0.0073×10^{18}	0.0084×10^{18}
Atoms after correction for neutron capture	$(9.7641 \pm 0.0410) \times 10^{17}$	$(1.1973 \pm 0.0050) \times 10^{18}$	$(1.5354 \pm 0.0066) \times 10^{18}$	$(1.7439 \pm 0.0073) \times 10^{18}$	$(1.9950 \pm 0.0084) \times 10^{18}$
Number of Pu-239 fissions			$(4.000 \pm 0.066) \times 10^{19}$		
Measured fission yield (%)	2.44 ± 0.04	2.99 ± 0.05	3.84 ± 0.07	4.36 ± 0.07	4.99 ± 0.08
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0166		
Assumed Pu-241 fission yield (%)	1.7	2.1	2.5	3.2	5.0
Pu-239 fission yield (%)	2.41	2.96	3.80	4.31	4.90
Std dev	0.04	0.05	0.07	0.07	0.08

[a] One sample was inadvertently lost during chemical separation.

Table XXVI

THERMAL FISSION YIELDS OF STABLE ZIRCONIUM ISOTOPES FOR Pu-239 FROM CAPSULE 6-2-6

	Zr-91	Zr-92	Zr-93	Zr-94	Zr-96
Determined number of atoms	1.1222×10^{18}	1.3757×10^{18}	1.7492×10^{18}	2.0076×10^{18}	2.2892×10^{18}
Std dev _{avg} (n=1)	0.0035×10^{18}	0.0047×10^{18}	0.0054×10^{18}	0.0062×10^{18}	0.0073×10^{18}
Atoms after correction for neutron capture	$(1.1220 \pm 0.0035) \times 10^{18}$	$(1.3756 \pm 0.0047) \times 10^{18}$	$(1.7503 \pm 0.0054) \times 10^{18}$	$(2.0065 \pm 0.0062) \times 10^{18}$	$(2.2882 \pm 0.0073) \times 10^{18}$
Number of Pu-239 fissions			$(4.447 \pm 0.066) \times 10^{19}$		
Measured fission yield (%)	2.52 ± 0.04	3.09 ± 0.05	3.93 ± 0.06	4.51 ± 0.07	5.14 ± 0.08
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0158		
Assumed Pu-241 fission yield (%)	1.7	2.1	2.5	3.2	5.0
Pu-239 fission yield (%)	2.50	3.06	3.89	4.46	5.07
Std dev	0.04	0.05	0.06	0.07	0.08

Table XXVII

THERMAL FISSION YIELDS OF STABLE ZIRCONIUM ISOTOPES FOR Pu-239 FROM CAPSULE 6-5-5

	Zr-91	Zr-92	Zr-93	Zr-94	Zr-96
Determined number of atoms	5.0315×10^{17}	6.1957×10^{17}	7.8839×10^{17}	9.0280×10^{17}	1.0264×10^{18}
Std dev _{avg} (n=4)	0.0206×10^{17}	0.0254×10^{17}	0.0323×10^{17}	0.0370×10^{17}	0.0042×10^{18}
Atoms after correction for neutron capture	$(5.0312 \pm 0.0206) \times 10^{17}$	$(6.1957 \pm 0.0254) \times 10^{17}$	$(7.8848 \pm 0.0323) \times 10^{17}$	$(9.0270 \pm 0.0370) \times 10^{17}$	$(1.0263 \pm 0.0042) \times 10^{18}$
Number of Pu-239 fissions			$(2.065 \pm 0.046) \times 10^{19}$		
Measured fission yield (%)	2.44 ± 0.05	3.00 ± 0.07	3.82 ± 0.08	4.37 ± 0.10	4.97 ± 0.11
<u>Pu-241 fissions</u> Pu-239 fissions (calculated)			0.0011		
Assumed Pu-241 fission yields (%)	1.7	2.1	2.5	3.2	5.0
Pu-239 fission yield (%)	2.43	3.00	3.81	4.37	4.96
Std dev	0.05	0.07	0.08	0.10	0.11

Table XXVIII

AVERAGE Pu-239 FISSION YIELDS

% Fission Yield			% Fission Yield		
Isotopic Mass	This Work	Fickel & Tomlinson ^[a]	Isotopic Mass	This Work	Fickel & Tomlinson ^[b]
Kr-83	0.292±0.004	0.29	Xe-131	3.51±0.05	3.77
Kr-84	0.473±0.007	0.47	Xe-132	4.98±0.08	5.26
Kr- + Rb-85	0.560±0.006	0.53	Cs-133	6.95±0.07	6.90
Kr-86	0.748±0.011	0.76	Xe-134	7.01±0.11	7.46
Rb-87	0.975±0.010	0.91	Cs-137	6.56±0.07	6.48
Sr-88	1.32±0.02	1.43	Ba-138	5.24±0.06	6.31
Sr-90	2.05±0.02	2.16	Ce-140	5.44±0.06	5.58
Zr-91	2.45±0.02	2.59	Ce-142	4.89±0.05	4.97
Zr-92	3.01±0.03	3.12	Nd-143	4.36±0.05	4.56
Zr-93	3.84±0.04	3.94	Nd- + Ce-144	3.65±0.04	3.84
Zr-94	4.39±0.04	4.45	Nd-145	2.96±0.03	3.12
Zr-96	4.99±0.05	5.13	Nd-146	2.43±0.02	2.57
			Nd-148	1.66±0.02	1.71
			Nd-150	0.940±0.009	1.02

[a] Uncertainty reported to be less than 3% (relative) on the individual fission yields.

[b] Uncertainty reported to be 2% (relative) on the individual fission yields.

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APPENDIX I

PROCEDURE FOR THE CHEMICAL SEPARATION OF FISSION PRODUCT RARE EARTH ELEMENTS FROM Pu-239 CAPSULES

W. A. Emel

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CHEMICAL SEPARATION

1. To a 50-ml polypropylene test tube, add 1 ml of sample, spikes of Ce-136, Nd-150, and Sm-144, 2 drops of nickel carrier (10 mg/ml), and 1 drop of 30% H_2O_2 .
2. Swirl to mix and heat under a heat lamp for 5 min.
3. Add 10M NaOH until $Al(OH)_3$ precipitates, then add a 2-ml excess.
4. Allow 5 min for complete precipitation, centrifuge, and discard the supernatant solution.
5. Wash the precipitate several times with quartz-distilled (QD) water. Discard the washings.
6. Immediately dissolve the precipitate in 2 ml of a mixture of 1M lactic acid - 0.05M penta-sodiumdiethylenetriaminepenta-acetic acid (Na_5DTPA) (pH = 2.90).
7. Transfer the dissolved precipitate to the first column.

If necessary, warm in a hot water bath to speed dissolution. The pH of the 1M lactic acid - 0.05M Na_5DTPA is adjusted to pH = 2.90 with QD NH_4OH .

Prepare the column material from 1 volume of 0.6M bis(2-ethylhexyl) phosphoric acid (D_2EHP) to two volumes of 100-200 mesh chromosorb "W". Add acetone as necessary to aid even coating of the D_2EHP on the chromosorb "W". Air dry overnight. Prepare a water slurry of this column material and transfer 1.5 ml of this slurry to an 8-mm ID polyethylene eye dropper. Pack the material to give a flow rate of 1 drop per 5 sec. Wash the column with 4 ml of QD 8M HNO_3 , 4 ml of QD H_2O , and 3 ml of 1M lactic acid - 0.05M Na_5DTPA .

8. Wash the column with three 1-ml portions of 1M lactic acid - 0.05M Na_5DTPA and two 1-ml portions of QD water. Discard the effluent.
9. Elute the lanthanides with four 1-ml portions of QD 8M HNO_3 into a 10-ml polypropylene tube containing 6 drops of 1M KBrO_3 .
10. Swirl to mix and heat gently under a heat lamp for 5 min.
11. Transfer the cooled solution to the second column and collect any effluent in a 50-ml polypropylene tube.

The second column is prepared similar to the first column using 1 volume of 3M D_2EHP and 5 volumes of chromosorb "W". The column is washed with four 1-ml portions of a freshly prepared solution of QD 8M HNO_3 - 0.1M KBrO_3 .
12. Rinse the column with two 1-ml portions of QD 8M HNO_3 -0.1M KBrO_3 solution. Collect all the effluent in the same 50-ml tube as used in step 11 for the subsequent separation of neodymium and samarium starting at step 15.

The effluent contains all of the lanthanides except cerium.
13. Elute the Ce with four 1-ml portions of QD 6M HCl - 1% H_2O_2 . Collect the effluent in a 10-ml polyethylene tube.
14. Evaporate to dryness and transfer the tube and residue to the mass spectrometry laboratory for Ce analysis.
15. To the solution from step 12, add 2 drops of Ni carrier (10 mg/ml) and an equal volume of 10M NaOH .
16. Allow 5 min for complete precipitation. Centrifuge and discard the supernatant solution.
17. Dissolve the precipitate in an equal volume of QD 2M HCl .

18. Add 2 ml of QD water and 2 drops of slurried Dowex 50 X 8, 200-400 mesh, resin (NH_4^+ form).
19. Swirl for 1 min and let stand for 15 min.
20. Transfer the resin to the top of a cation exchange column. Take care to transfer as little liquid as possible. Prepare the column to a depth of 14 cm in an 8-mm glass tube, with Dowex 50 X 8, 200-400 mesh, resin in the NH_4^+ form.
21. Wash the column with 15 ml of 0.5M α -hydroxyisobutyric acid (pH = 3.50). Discard the effluent. Adjust to pH 3.50 with QD 6M NH_4OH .
22. Immediately continue the elution with six 1-ml portions of 0.5M α -hydroxyisobutyric acid at a pH of 3.90. Collect each of these 1-ml portions in a different 10-ml polypropylene tube and label as fraction 1 through 6. Add the first few portions carefully to avoid disturbing the top of the resin bed.
23. Continue the elution with nine 0.5-ml portions of the elutriant and collect each portion separately and label as fractions 7 through 15. Each volume of elutriant should be delivered with a drifilmed pipet.
24. Continue the elution with additional ten 1-ml portions of elutriant and collect each portion separately and label as fractions 16 through 25.
25. From each fraction, mount a 5- μ l aliquot for β -proportional counting.
26. Beta count each fraction and prepare a semilog plot of activity versus fraction or activity versus total volume of elutriant. A gamma scan of each fraction for Eu-152 and Eu-155 aids to locate the Eu peak.
27. The first major peak observed is due to Eu and the second to Pm. The fraction lying in the valley between the Eu and Pm peaks contains the Sm fraction

and the fraction immediately after the Pm peak contains the Nd fraction.

28. To the Nd and Sm fractions, add an equal volume of QD 2M HCl. Mix well. A Vortex mixer is satisfactory.
29. Transfer the Nd and Sm fractions, individually, to different cation exchange columns. Prepare the ion exchange columns in 8-mm polyethylene eye droppers from 0.5 ml of Dowex 50 X 8, 100-200 mesh, resin in the H^+ form. Wash the column with 3 ml of QD 10M HCl and then with 5 ml of QD water.
30. Wash the column with 4 ml of QD 2M HNO_3 and discard the effluent.
31. Elute the respective lanthanide with 4 ml of QD 6M HCl. Collect the effluent in a 10-ml polypropylene tube.
32. Evaporate the effluent to dryness and transfer the tube and residue to the mass spectrometry laboratory.

APPENDIX II

PROCEDURE FOR THE CHEMICAL SEPARATION OF FISSION
PRODUCT ZIRCONIUM FOR Pu-239 CAPSULES

W. A. Emel

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CHEMICAL SEPARATION

1. To a 50-ml Pyrex centrifuge tube, add the enriched Zr-90 spike, 1 mg of high-purity thorium carrier, 1 drop of 30% H_2O_2 , 2 ml of 15.3M HNO_3 , and 10 ml of 0.35M HIO_3 .

Distill all acids and use quartz-distilled water to prepare them.

Remove any Br_2 formed by air sparging.

$\text{Al}(\text{NO}_3)_3$ may precipitate in unspiked samples; if so, add QD water to redissolve.
2. Allow 5 min for thorium iodate to precipitate.
3. Centrifuge and discard the supernatant solution.
4. Dissolve the precipitate in a few drops of 10.3M HCl , add 1 drop of 30% H_2O_2 , and heat over an open flame for about 3 min until all the free iodine is expelled.

Test for the completeness of the evolution of free iodine with moistened starch paper held above the tube.
5. Add 2 ml of 15.3M HNO_3 , and heat to destroy the HCl .
6. Add 1 drop of 30% H_2O_2 and 10 ml of 0.35M HIO_3 .
7. Allow 5 min for thorium iodate to precipitate.
8. Centrifuge and discard the supernatant solution.
9. Dissolve the precipitate in approximately 0.5 ml of 10.3M HCl . Warm the sample to ensure complete dissolution.
10. Cool the sample and add 1 ml of quartz distilled 10.3M HCl .

About 80% of the initial plutonium is present at this point.
11. Pass the dissolved residue through a small Dowex 1X8 (100 to 200 mesh) column.

Fill a 2-ml polyethylene medicine dropper with approximately 1 in. of Dowex 1X8 resin. The column is preequilibrated with 10.3M HCl . Do not use glassware for the column nor in any subsequent steps. Polypropylene ware is

recommended for the subsequent steps. The plutonium is removed from the sample at this point.

12. Wash the column with three 2-ml portions of 10.3M HCl. Discard the washings.
13. Elute the adsorbed zirconium with three 1-ml portions of 8M HNO₃. Collect the effluent in a polypropylene tube.
14. Evaporate the effluent to approximately 0.1 ml and transfer the tube and residue to the mass laboratory.