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BURNUP DETERMINATION OF NUCLEAR FUELS
PROJECT REPORT
FOR THE QUARTER APRIL 1 - JUNE 30, 1968 AND FINAL REPORT

MASTER

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
IDAHO FALLS, IDAHO

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SUMMARY

In this final quarterly report for the program to develop methods for the accurate determination of the burnup of nuclear fuels, new measurements are reported for the thermal fission yields of Sb-125 and the stable isotopes of molybdenum for U-233; Ce-141, Sb-125, and the stable isotopes of molybdenum for U-235; Sb-125, the stable isotopes of molybdenum, the stable and long lived isotopes of ruthenium, the stable isotopes of samarium, and mass chains 95, 97, and 99 for Pu-239; Sb-125, the stable and long lived isotopes of ruthenium, and mass chains 95, 97, and 99 for Pu-241. Also reported are new measurements of the fast (EBR I) fission yields of Sb-125 and the stable isotopes of molybdenum for U-235; and of Sb-125 and the long lived and stable isotopes of Ba, Cd, Ce, Cs, Kr, Mo, Nd, Rb, Sm, Sr, Xe, and Zr for Pu-239.

A comprehensive listing is presented for all the fission yield data obtained in the program. Included are thermal fission yields for U-233, U-235, Pu-239, and Pu-241 and fast fission yields for U-235 and Pu-239.

Finally, various fission products are recommended as nuclear fuel burnup monitors for various fuel types. Monitors are proposed for the determination of total fissions in monoisotopic and polyisotopic fissioning fuels and for the relative fissions from the individual fissionable isotopes in several duoisotopic fuels.

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BURNUP DETERMINATION OF NUCLEAR FUELS

INTRODUCTION

The objective of this program 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 corrections 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.

Thermal fission yields have been measured accurately for U-233, U-235, Pu-239, and Pu-241. Also measured were a considerable number of fast (EBR I) fission yields for U-235 and Pu-239. These values have been presented in previous reports IDO-14660, IDO-14663, IDO-14667, IDO-14676, IDO-14678, IDO-14681, IN-1064, IN-1113, IN-1157 Rev, IN-1178, IN-1189, IN-1207, and IN-1215. The present report is the final one for the program and includes additional fast fission yield data for U-235 and Pu-239, additional thermal fission yields for U-233, U-235, Pu-239, and Pu-241, and a summary of all the fission yield data obtained in the program. Lastly, a selected listing of fission products is presented for consideration as burnup monitors as a function of reactor irradiation conditions.

FAST FISSION YIELDS OF THE STABLE AND LONG LIVED ISOTOPES OF Ba, Cd, Ce, Cs, Kr, Mo, Nd, Rb, Ru, Sm, Sr, Xe, and Zr FOR Pu-239

The last goal of this project is the measurement of the fast fission yields for stable and long lived fission products for Pu-239. An irradiated fuel rod from the Mark IV loading of EBR I was kindly provided by the Argonne National Laboratory for this purpose.

The burnup of this rod was only about 0.07% fissioning of the Pu-239. At this low burnup, an independent calculation of the number of fissions based on measured losses of Pu-239 atoms could not be done accurately. Thus direct measurements of absolute fission yields (the quotients obtained by dividing measured atoms of fission products by the number of fissions) were not possible. At a burnup of 0.07% fission, both the preirradiation and postirradiation number of Pu-239 atoms would have to be known (or determined) to 0.0005 relative percent to obtain the number of fissions to a precision of 1 relative percent. This obviously is impossible. Thus, relative fission yields of many stable and long lived fission products of the 13 elements listed in the title to this section were measured by determining the relative number of atoms, mainly by the technique of isotope dilution mass spectrometry, and in a few cases by gamma ray spectrometry.

Irradiation History of the Fuel Rod

The fuel rod (Argonne designation P-231) consisted of a Zircaloy-2 tube containing four slugs of 90 atom % Pu - 10 atom % Al alloy. The stated amount of plutonium in the rod was 88.07 g and the stated ²³⁹Pu isotope abundance was 95.1%. The rod had been exposed intermittently in the NaK-cooled EBR I for a total exposure of 5.8×10^5 kWh over a period of 1 year with nominal power operation approximately 80% of this time. Nominal operating conditions were 900 to 1100 kW power, 9.5×10^{13} n/cm²-sec flux at 950 kW, and 450° C fuel temperature.

The rod was cut open, and the four slugs were removed and stored for 4 years before dissolution.

Dissolution of the Fuel Slugs (W. A. Emel, R. E. Foster, Jr.).

The four slugs were dissolved as two pairs. The dissolution and krypton-xenon gas collection apparatus have been described previously (1, 2). Each pair was dissolved in ~500 ml of 7.5M HCl. To maintain control of the dissolution rate, the slugs were covered with water, heated to just below boiling, and the acid was added incrementally by a peristaltic pump. To achieve the complete dissolution of traces of the alloy, any traces of PuO₂, and to dissociate polymeric forms of plutonium, 0.5 ml of 27M HF was added near the end of the dissolution. Each dissolution took about 2 hours. The two dissolver solutions were combined in one polyethylene bottle and the final volume was measured to be 1371 ml.

Fission Product Analyses

The solution was analyzed for the stable and long-lived fission products of Ba, Cd, Ce, Cs, Mo, Nd, Rb, Ru, Sm, Sr, and Zr. The separation procedures except for an initial separation of plutonium, the mass spectrometric procedures, and the calculational procedures were similar to those used for the previous thermal fission yield measurements (3-6). For all the fission products except Mo and Cd, the sample aliquots were adjusted to 8M HNO₃, transferred to a small Dowex 1 X8 anion exchange column which retained the plutonium, and the fission products were eluted with 8M HNO₃. Plutonium was separated from molybdenum by passing a sample aliquot through a cation exchange column preequilibrated with 1M HCl. Molybdenum was eluted with 1M HCl. For Cd, no preseparation was necessary.

The atoms of the krypton and xenon isotopes that had been quantitatively trapped from the dissolution off-gases were determined by a combination of vapor phase chromatography and mass spectrometry as described previously (2).

The average number of determined atoms for each fission product in the rod is listed in Table I. The associated errors are in terms of the standard deviation of the average value.

Because of the very sporadic and lengthy irradiation period of the fuel rod, decay corrections for Sr-90, Cs-137, and 10.7-yr Kr-85 were made to the midtime of the irradiation period. Buildup and burnout corrections for neutron capture on fission products are unnecessary for this low burnup, fast spectrum irradiated fuel.

Partial loss of krypton and xenon was possible from the bare slugs during the 4-year storage period. Figure 1 is a plot of the ratios of the determined atoms of each mass chain from 83 to 88 relative to 88 for this rod and for previously determined thermal Pu-239 data (5). The fact that the curve drawn through the ratios at the mass chains 83, 84, and 86 (in which krypton was predominant at the time of dissolution) and mass chains 85 and 87 (in which rubidium was predominant at time of dissolution) is essentially parallel to the curve for thermal data indicates little loss of krypton. This treatment was not applied to the xenon data because its stable isotopes are in the mass region where significant fine structure is observed. Because the diffusion coefficient for krypton is larger than that for xenon, the loss of xenon also is believed to be small.

Figure 1. Determined Atoms of Mass Chains 83 to 88
Relative to Mass Chain 88 for EBR I Pu-239 Fuel

Table I

DETERMINED NUMBERS OF FISSION PRODUCT ATOMS IN EBR I Pu-239 FUEL

Nuclide ^[a]	Atoms	Nuclide ^[a]	Atoms
Kr-83 (1) ^[b]	$(7.3589 \pm 0.1100) \times 10^{17}$	Mo-100 (4)	$(1.4523 \pm 0.0078) \times 10^{19}$
Kr-84 (1) ^[b]	$(1.2011 \pm 0.0168) \times 10^{18}$	Ru-101 (4)	$(1.4774 \pm 0.0060) \times 10^{19}$
Kr-85 (1) + Rb-85 (4)	$(1.4439 \pm 0.0066) \times 10^{18}$	Ru-102 (4)	$(1.4968 \pm 0.0061) \times 10^{19}$
Kr-86 (1) ^[b]	$(1.3943 \pm 0.0265) \times 10^{18}$	Ru-104 (4)	$(1.4529 \pm 0.0049) \times 10^{19}$
Rb-87 (4)	$(2.4980 \pm 0.0117) \times 10^{18}$	Cd-111 (4)	$(8.0748 \pm 0.0283) \times 10^{17}$
Sr-88 (2)	$(3.3896 \pm 0.0259) \times 10^{18}$	Cd-112 (4)	$(4.4466 \pm 0.0169) \times 10^{17}$
Sr-90 (2)	$(4.8142 \pm 0.0616) \times 10^{18}$	Cd-113 (4)	$(2.8513 \pm 0.0120) \times 10^{17}$
Zr-91 (4)	$(5.5423 \pm 0.0227) \times 10^{18}$	Cd-114 (4)	$(2.1177 \pm 0.0123) \times 10^{17}$
Zr-92 (4)	$(6.7182 \pm 0.0457) \times 10^{18}$	Cd-116 (4)	$(1.3653 \pm 0.0141) \times 10^{17}$
Zr-93 (4)	$(8.4028 \pm 0.0304) \times 10^{18}$	Xe-131 (1) ^[b]	$(8.7113 \pm 0.0923) \times 10^{18}$
Zr-94 (4)	$(9.4216 \pm 0.0335) \times 10^{18}$	Xe-132 (1) ^[b]	$(1.1634 \pm 0.0123) \times 10^{19}$
Mo-95 (4)	$(1.0274 \pm 0.0039) \times 10^{19}$	Cs-133 (4)	$(1.4840 \pm 0.0028) \times 10^{19}$
Zr-96 (4)	$(1.0964 \pm 0.0068) \times 10^{19}$	Xe-134 (1) ^[b]	$(1.5790 \pm 0.0167) \times 10^{19}$
Mo-97 (4)	$(1.1739 \pm 0.0037) \times 10^{19}$	Cs-135 (4)	$(1.6201 \pm 0.0031) \times 10^{19}$
Mo-98 (4)	$(1.2483 \pm 0.0064) \times 10^{19}$	Xe-136 (1) ^[b]	$(1.4867 \pm 0.0157) \times 10^{19}$

Table I (Continued)

DETERMINED NUMBERS OF FISSION PRODUCT ATOMS IN EBR I Pu-239 FUEL

Cs-137 (4)	$(1.4124 \pm 0.0027) \times 10^{19}$	Nd-146 (15)	$(5.4108 \pm 0.0254) \times 10^{18}$
Ba-138 (4)	$(1.0674 \pm 0.0067) \times 10^{19}$	Nd-148 (15)	$(3.7108 \pm 0.0163) \times 10^{18}$
Ce-140 (3)	$(1.2013 \pm 0.0032) \times 10^{19}$	Sm-149 (4)	$(2.9226 \pm 0.0077) \times 10^{18}$
Ce-142 (3)	$(1.0636 \pm 0.0020) \times 10^{19}$	Nd-150 (15)	$(2.2767 \pm 0.0114) \times 10^{18}$
Nd-143 (15)	$(9.2330 \pm 0.0518) \times 10^{18}$	Sm-151 (4)	$(1.8018 \pm 0.0047) \times 10^{18}$
Nd-144(15) + Ce-144(3)	$(7.9064 \pm 0.0432) \times 10^{18}$	Sm-152 (4)	$(1.4663 \pm 0.0041) \times 10^{18}$
Nd-145 (15)	$(6.5343 \pm 0.0323) \times 10^{18}$	Sm-154 (4)	$(6.9510 \pm 0.0183) \times 10^{17}$

[a] The number of replicate determinations is given in parentheses.

[b] Only one sample of krypton and xenon is obtained for each dissolution. The associated error was obtained from measurements on standards in the same range as the fission product sample.

Fission Yield Values

The calculated fast fission yields relative to Nd-148 are listed in Table II. The associated errors are in terms of the standard deviation of the average value.

The absolute fission yields for the 119 to 160 mass region were calculated by normalizing the sum of the relative values to 100. In this calculation, quantitative recovery of xenon was assumed. With no measurements in the 119 to 124 and 126 to 130 regions, the relative fission yields for these regions were estimated using the experimental cadmium data to fix the depth of the valley in the 119 to 123 region, fitting a smooth curve between 123 and 125, then fitting an assumed smoothly rising curve between 125 and 131. For the mass region above 138 little fast fission yield data, relative or absolute, have been reported for masses 139, 141, 147, 153, and 155 through 160. For these masses, relative yields were estimated by interpolation and extrapolation of the experimental data assuming a smoothly varying mass yield curve in this region.

Absolute fast fission yields were calculated for the experimentally measured nuclides in the light mass peak by multiplying the determined relative (to Nd-148) yield by the now calculated absolute Nd-148 fast fission yield.

Table III lists the measured, as well as the estimated, Pu-239 fast fission yields. A relative standard deviation of $\pm 20\%$ was assigned to each estimated yield value. The standard deviation errors given for the measured values in Table III are propagated errors and include the 20% uncertainty in the estimated values. Details of the error calculation have been previously reported (6).

Table II

FAST FISSION YIELDS RELATIVE TO Nd-148 FOR Pu-239

<u>Mass No</u>	<u>Element</u>	<u>Relative Fission Yield</u>	<u>Mass No</u>	<u>Element</u>	<u>Relative Fission Yield</u>
83	Kr	0.2118±0.0031	116	Cd	0.0368±0.0004
84	Kr	0.3237±0.0047	125 ^[a]	Sb	0.0415±0.0050
85	Kr + Rb	0.3891±0.0025	131	Xe	2.347±0.027
86	Kr	0.5105±0.0075	132	Xe	3.135±0.036
87	Rb	0.6732±0.0043	133	Cs	3.999±0.019
88	Sr	0.8326±0.0079	134	Xe	4.255±0.049
90	Sr	1.297±0.017	135	Cs	4.366±0.021
91	Zr	1.494±0.009	136	Xe	4.006±0.046
92	Zr	1.810±0.015	137	Cs	3.806±0.017
93	Zr	2.264±0.013	138	Ba	2.876±0.022
94	Zr	2.539±0.014	140	Ce	3.237±0.017
95	Mo	2.769±0.015	142	Ce	2.866±0.014
96	Zr	2.955±0.022	143	Nd	2.488±0.018
97	Mo	3.163±0.016	144	Nd + Ce	2.131±0.015
98	Mo	3.364±0.022	145	Nd	1.761±0.012
100	Mo	3.914±0.026	146	Nd	1.458±0.009
101	Ru	3.981±0.024	148	Nd	1.000
102	Ru	4.034±0.024	149	Sm	0.7876±0.0040
104	Ru	3.915±0.022	150	Nd	0.6135±0.0041
111	Cd	0.2176±0.0012	151	Sm	0.4856±0.0025
112	Cd	0.1198±0.0007	152	Sm	0.3951±0.0021
113	Cd	0.0768±0.0005	154	Sm	0.1873±0.0010
114	Cd	0.0571±0.0004			

[a] Sb-125 determination described on page 15 of this report.

Table III

FAST FISSION YIELDS FOR Pu-239

<u>Mass No</u>	<u>Element</u>	<u>Fission Yield</u>	<u>Mass No</u>	<u>Element</u>	<u>Fission Yield</u>
83	Kr	0.366±0.008	116	Cd	0.0636±0.0013
84	Kr	0.559±0.013	119 ^[a]	---	0.064
85	Rb + Kr	0.672±0.012	120 ^[a]	---	0.064
86	Kr	0.882±0.020	121 ^[a]	---	0.066
87	Rb	1.16±0.02	122 ^[a]	---	0.071
88	Sr	1.44±0.03	123 ^[a]	---	0.086
90	Sr	2.24±0.05	124 ^[a]	---	0.119
91	Zr	2.58±0.05	125 ^[b]	Sb	0.194±0.023
92	Zr	3.13±0.06	126 ^[a]	---	0.304
93	Zr	3.91±0.07	127 ^[a]	---	0.502
94	Zr	4.39±0.08	128 ^[a]	---	0.796
95	Mo	4.78±0.09	129 ^[a]	---	1.31
96	Zr	5.11±0.10	130 ^[a]	---	2.08
97	Mo	5.47±0.10	131	Xe	4.06±0.08
98	Mo	5.81±0.11	132	Xe	5.42±0.11
100	Mo	6.76±0.13	133	Cs	6.91±0.12
101	Ru	6.88±0.13	134	Xe	7.35±0.15
102	Ru	6.97±0.13	135	Cs	7.54±0.13
104	Ru	6.77±0.12	136	Xe	6.92±0.14
111	Cd	0.376±0.007	137	Cs	6.58±0.11
112	Cd	0.207±0.004	138	Ba	4.97±0.09
113	Cd	0.133±0.002	139 ^[a]	---	5.88
114	Cd	0.0987±0.0019	140	Ce	5.59±0.10

Table III (Continued)

FAST FISSION YIELDS FOR Pu-239

<u>Mass No</u>	<u>Element</u>	<u>Fission Yield</u>	<u>Mass No</u>	<u>Element</u>	<u>Fission Yield</u>
141 ^[a]	---	5.28	151	Sm	0.839±0.015
142	Ce	4.95±0.09	152	Sm	0.683±0.012
143	Nd	4.30±0.08	153 ^[a]	---	0.484
144	Ce + Nd	3.68±0.07	154	Sm	0.324±0.006
145	Nd	3.04±0.05	155 ^[a]	---	0.258
146	Nd	2.52±0.04	156 ^[a]	---	0.178
147 ^[a]	---	2.18	157 ^[a]	---	0.119
148	Nd	1.73±0.03	158 ^[a]	---	0.071
149	Sm	1.36±0.02	159 ^[a]	---	0.044
150	Nd	1.06±0.02	160 ^[a]	---	0.025

[a] Estimated yield.

[b] Sb-125 determination described on page 15 of this report.

FAST FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-235
(R. E. Foster, Jr., W. A. Emel, M. E. Kussy, L. W. Buttars,
R. E. McAtee, R. A. Nielsen, G. D. Workman)

The fast fission yields for the stable molybdenum isotopes were measured on a U-235 fuel rod from the MARK III loading of EBR I. Details on the composition, irradiation history, and dissolution of the rod have been previously reported (6). Duplicate aliquots of the dissolver solution spiked with known amounts of Mo-94 and duplicate unspiked aliquots were analyzed by the separation and mass spectrometric procedures (Appendix I) that previously were used for thermal fission yield measurements with the addition of an initial separation step for zirconium. Zirconium (from the Zircaloy-2 cladding) was absorbed on a Dowex 50 X8 cation exchange column pre-equilibrated with 0.1M HCl and the molybdenum was eluted with 0.1M HCl.

The number of fissions in the fuel rod could not be directly calculated (6). The molybdenum fission yields were, therefore, obtained relative to Nd-148 and the absolute yields were calculated by multiplying by the fast fission yield of Nd-148 that previously had been determined (6).

The analysis results, relative fission yields, and calculated fast fission yields are summarized in Table IV.

Table IV

FAST FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-235

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms ^{[a][b]}	$(1.3126 \pm 0.0047) \times 10^{20}$	$(1.2436 \pm 0.0043) \times 10^{20}$	$(1.2253 \pm 0.0048) \times 10^{20}$	$(1.2890 \pm 0.0065) \times 10^{20}$
Determined number of Nd-148 atoms ^[b]		$(3.5501 \pm 0.0316) \times 10^{19}$	(Ref 6)	
Relative fission yield to Nd-148	3.697 ± 0.035	3.503 ± 0.033	3.451 ± 0.034	3.631 ± 0.037
Nd-148 fast fission yield, %		1.75 ± 0.03	(Ref 6)	
Fast fission yield, %	6.47	6.13	6.04	6.35
Std dev	0.13	0.12	0.12	0.13

[a] Error is expressed as standard deviation of the reported average of duplicate determinations.

[b] The values for both the molybdenum isotopes and Nd-148 are expressed per total volume of the dissolved fuel solution.

FAST FISSION YIELDS OF Sb-125 FOR U-235 AND Pu-239

(R. E. Foster, Jr.)

The fast fission yields of Sb-125 for U-235 and Pu-239 were measured on the dissolved EBR I fuel rods. Four 1-ml aliquots of the dissolved U-235 solution and four 0.5-ml aliquots of the dissolved Pu-239 solution were analyzed for atom concentrations of Sb-125 as described in the next section for the thermal fission yield measurements of Sb-125 for U-233 and U-235. The fast fission yields of Sb-125 were calculated by taking these atom concentrations relative to the previously determined Nd-148 concentrations for these two solutions and multiplying by the previously determined Nd-148 fission yields. The experimental data and calculated fission yields are given in Table V.

Table V

FAST FISSION YIELDS OF Sb-125 FOR U-235 AND Pu-239

	<u>U-235</u>	<u>Pu-239</u>
Sb-125 activity [a], [b] (counts/min/ml)	$(3.059 \pm 0.114) \times 10^4$	$(1.838 \pm 0.038) \times 10^4$
Detector and nuclear parameters for Sb-125 [(TPA)(BR)(λ)]	$(6.06 \pm 0.69) \times 10^{-11}$	
Atoms Sb-125/ml	(1) $(5.048 \pm 0.605) \times 10^{14}$	$(3.033 \pm 0.351) \times 10^{14}$
Previously determined atoms Nd-148/ml	(2) $(1.216 \pm 0.011) \times 10^{16}$ (Ref 6)	$(2.707 \pm 0.012) \times 10^{15}$ [c]
Nd-148 fission yield, %	(3) 1.75 ± 0.03 (Ref 6)	1.73 ± 0.03 [c]
Sb-125 fission yield, %	0.0727 ± 0.0090	0.194 ± 0.023
$\frac{(1) (3)}{(2)}$		

[a] Four 1-ml aliquots of the dissolved U-235 solution were each counted for 4 min. Four 0.5-ml aliquots of the dissolved Pu-239 solution were each counted for 10 min.

[b] All activities were corrected for decay to the midtime of the reactor residence time of the fuel.

[c] This report, page 6.

THERMAL FISSION YIELDS OF Sb-125 FOR U-233, U-235, Pu-239, AND Pu-241

(R. E. Foster, Jr.)

The thermal fission yields of Sb-125 for U-233 and U-235 were measured on the dissolved, standard burnup capsules CPP-6-4-2 and CPP-6-2-4 for U-233 (7, 8) and CPP-6-4-1 and CPP-6-6-2 for U-235 (2, 9). The antimony was quantitatively separated by a carrier-free distillation of the volatile bromide (APPENDIX II) on four 1-ml aliquots from each of the four dissolved capsule solutions. The distillate, in a 50-ml volumetric flask, was counted with a Ge(Li) detector and pulse height analyzer system. The detector parameters at the 428-keV photopeak of Sb-125 were obtained by cross calibrating a pure Sb-125 source with a 3- x 3-in. NaI(Tl) detector with known parameters. The branching ratio for the 428-keV gamma transition used was 0.284 ± 0.031 (10). The fission yield was calculated by dividing the determined atoms of Sb-125 by the previously determined number of fissions for each of the four capsules (2, 7, 8, 9). The experimental data and calculated fission yields are given in Table VI for U-233 and in Table VII for U-235.

The thermal fission yields of Sb-125 for Pu-239 and Pu-241 were measured on 10-hr irradiated samples at a flux of 10^{14} n/cm²-sec in a highly thermalized section (VH-1) of the Materials Testing Reactor. Two flame sealed quartz ampoules containing 2 to 3 mg of each isotope were irradiated. The plutonium materials were 99% Pu-239 as the dioxide and 95% Pu-241 as the dioxide. The Pu-241 material was purified of Am-241 by anion exchange from an 8M HNO₃ medium just prior to irradiation. After irradiation, each quartz ampoule was individually placed in a stainless steel tube which had one crimped end. The tubes were individually suspended in the neck of a round bottom quartz flask. The neck of the flask had a standard taper joint to accommodate a water-cooled reflex condenser. The ampoule was shattered with a tungsten rod and the quartz fragments and irradiated PuO₂ were quantitatively transferred to the flask with 7M HCl - 1M HF washings. The PuO₂ was then dissolved by boiling under total reflux in a mixture of 7M HCl - 1M HF. Four 10-ml aliquots of each of the four solutions were analyzed for Sb-125 as described above and for Cs-137 (1). The fission yields of Sb-125 were calculated by multiplying the determined atom ratio of Sb-125/Cs-137 by the previously determined fission yields of Cs-137 for Pu-239 and Pu-241. The experimental data and calculated fission yields are given in Table VIII for Pu-239 and in Table IX for Pu-241.

Table VI

THERMAL FISSION YIELD OF Sb-125 FOR U-233

Sb-125 activity ^[a] (counts/min/ml)	<u>Standard Capsule 6-2-4</u>		<u>Standard Capsule 6-4-2</u>	
	(1)	(1.065±0.018)x10 ⁴	(1.177±0.023)x10 ⁴	
Detector parameters and nuclear constants for Sb-125 [(λ)(TPA)(BR)] ^[b]			(6.06±0.69)x10 ⁻¹¹	
Total volume of dissolver solution (ml)	(3)	242.7±0.1	254.2±0.1	
Determined number of fissions	(4)	(3.786±0.059)x10 ¹⁹	(Ref_8)	(4.138±0.045)x10 ¹⁹ (Ref_7)
Fission yield, %		0.113±0.013	0.119±0.013	
$\frac{(1)(3)}{(2)(4)} (100)$				
Avg fission yield, %			0.116±0.013	

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

[b] The half life value used to calculate λ was 2.81±0.05 yr (13).

Table VII

THERMAL FISSION YIELD OF Sb-125 FOR U-235

		<u>Standard Capsule 6-4-1</u>	<u>Standard Capsule 6-6-2</u>
Sb-125 activity ^[a] (counts/min/ml)	(1)	$(2.011 \pm 0.035) \times 10^3$	$(1.609 \pm 0.026) \times 10^3$
Detector parameters and nuclear constants for Sb-125 $[(\lambda)(TPA)(BR)]$ ^[b]	(2)		$(6.06 \pm 0.69) \times 10^{11}$
Total volume of dissolver solution (ml)	(3)	243.7 ± 0.1	247.4 ± 0.1
Determined number of fissions	(4)	$(2.839 \pm 0.032) \times 10^{19}$ (Ref 2)	$(2.204 \pm 0.030) \times 10^{19}$ (Ref 9)
Fission yield %		0.0285 ± 0.0033	0.0298 ± 0.0034
$\frac{(1)(3)}{(2)(4)}$ (100)			
Avg fission yield, %			0.0291 ± 0.0033

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

[b] The half life value used to calculate λ was 2.81 ± 0.05 yr (13).

Table VIII

THERMAL FISSION YIELD OF Sb-125 FOR Pu-239

	<u>Capsule 423</u>	<u>Capsule 424</u>
Sb-127 activity ^{[a][b]} (counts/min/ml)	10.63±0.10	9.00±0.15
Cs-137 activity ^[b] (counts/min/ml)	353.8±3.4	312.9±0.6
Sb-125/Cs-137 activity ratio	(2.922±0.039)×10 ⁻²	(2.876±0.048)×10 ⁻²
Avg Sb-125/Cs-137 activity ratio (1)	(2.904±0.030)×10 ⁻²	
Detector and nuclear parameters for Cs-137 [(TPA)(BR)(λ)] (2)	(3.608±0.108)×10 ⁻¹¹	
Detector and nuclear parameters for Sb-125 [(TPA)(BR)(λ)][c] (3)	(6.06±0.69)×10 ⁻¹¹	
Cs-137 fission yield, % (4)	6.74±0.14	
Sb-125 fission yield, %	0.116±0.014	
$\frac{(1)(2)(4)}{(3)}$		

[a] Four 10-ml aliquots of the sample were each counted for 40 min.

[b] Corrected for in-pile and out-of-pile decay.

[c] The half life value used to calculate λ was 2.81±0.05 yr (13).

Table IX

THERMAL FISSION YIELD OF Sb-125 FOR Pu-241

	<u>Capsule 421</u>	<u>Capsule 422</u>
Sb-125 activity ^{[a][b]} (counts/min/ml)	3.016±0.079	3.036±0.015
Cs-137 activity ^[b] (counts/min/ml)	(2.838±0.036)×10 ²	(2.869±0.044)×10 ²
Sb-125/Cs-137 activity ratio	(1.063±0.031)×10 ⁻²	(1.058±0.017)×10 ⁻²
Avg Sb-125/Cs-137 activity ratio	(1) (1.059±0.015)×10 ⁻²	
Detector and nuclear parameters for Cs-137 [(TPA)(BR)(λ)]	(2) (3.608±0.108)×10 ⁻¹¹	
Detector and nuclear parameters for Sb-125 [(TPA)(BR)(λ)][c]	(3) (6.06±0.69)×10 ⁻¹¹	
Cs-137 fission yield, %	(4) 6.60±0.17 (Ref 11)	
Fission yield, %	0.0416±0.0050	
$\frac{(1)(2)(4)}{(3)}$		

- [a] Four 10-ml aliquots of sample 421 were each counted for 40 min.
Four 10-ml aliquots of sample 422 were each counted for 1000 min.
- [b] Corrected for in-pile and out-of-pile decay.
- [c] The half life value used to calculate λ was 2.81±0.05 yr (13).

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM

FOR U-233 AND U-235

The thermal fission yields of the molybdenum isotopes were determined on four previously dissolved U-233 capsules and on three previously dissolved U-235 capsules. The irradiation histories, capsule contents, and burnup for each capsule are given in Table X. Four aliquots spiked with a known amount of Mo-94 and two without spike, from each dissolved capsule solution, were chemically separated and then analyzed by mass spectrometry.

The analysis results and calculated absolute thermal fission yields are summarized in Tables XI through XIV for U-233 and Tables XV through XVII for U-235.

The accurate determination of absolute thermal 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. 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 (9).

For the molybdenum isotopes, corrections for buildup and burnout by neutron capture are small being less than 0.2% (relative). Figure 2 is a schematic diagram of the buildup and burnout program for molybdenum.

Chemical Separation (R. E. Foster, Jr., M. E. Kussy, F. L. Lisman)

The procedure developed for the carrier-free separation of molybdenum prior to mass spectrometric analysis is rapid and simple. Ammonium hydroxide is added to precipitate aluminum hydroxide which carries hydrolyzable fission products, the supernatant solution is evaporated to dryness, the residue is dissolved in 0.1M HCl and the solution is transferred to an anion column, and after washing with 0.1M HCl, molybdenum is eluted with 5M HNO₃.

As K₂O interferes with the Mo-94 spike peak in the mass spectrometric analysis, the final sample must be free from potassium. For this reason, only clean polypropylene ware is used in handling the sample after it is eluted from the anion exchange column. As a general practice, only clean plastic ware and quartz-distilled reagents are used throughout the separation procedure.

The separation procedure and the preparation and standardization of the Mo-94 spike are detailed in APPENDIX I.

Mass Spectrometric Analysis (R. M. Abernathey, R. E. McAtee,
G. D. Workman)

The mass spectrometer used for this work was a Nuclide Corporation TDD-1, tandem, 12-in., 90° instrument. For the molybdenum analysis, an electron multiplier, electrometer, strip chart detection system was used. Details of this instrument and the detection system have been previously reported (3). An aqueous chloride solution of the separated molybdenum was mounted on a single rhenium filament.

The procedure for the mass spectrometric analysis is detailed in APPENDIX I. Typical data showing the reproducibility of the isotope dilution analysis, obtained on four aliquots of a dissolved, irradiated U-235 sample, are summarized in Table XVIII.

Figure 2. COMPUTER PROGRAM SCHEMATIC FOR
BUILDUP AND BURNOUT OF FISSION
PRODUCT MOLYBDENUM

Table X

COMPOSITION AND IRRADIATION HISTORY OF STANDARD BURNUP CAPSULES

Capsule	Fissionable Isotope	Enrichment, Wt %	mg U ₃ O ₈ in Capsule	MTR Cycles	Burnup ^[a]	Determined Number of Fissions
6-2-4	U-233	99.908	58.8±0.2	8	32.40±0.37	(4.138±0.045)×10 ¹⁹ (Ref 8)
6-4-2	U-233	99.908	152.4±0.2	3	11.44±0.18	(3.786±0.059)×10 ¹⁹ (Ref 7)
6-4-3	U-233	99.908	126.0±0.2	3	12.39±0.23	(3.390±0.062)×10 ¹⁹ (Ref 9)
6-5-1	U-233	99.908	144.4±0.2	2	8.33±0.17	(2.614±0.055)×10 ¹⁹ (Ref 8)
6-4-1	U-235	99.163	115.2±0.1	3	11.50±0.13	(2.839±0.032)×10 ¹⁹ (Ref 2)
6-6-2	U-235	99.163	81.9±0.1	3	12.54±0.18	(2.201±0.030)×10 ¹⁹ (Ref 9)
6-2-2	U-235	99.163	61.2±0.1	8	31.57±0.19	(4.142±0.022)×10 ¹⁹ (Ref 12)

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

Table XI

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-233 FROM CAPSULE 6-2-4

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms	2.7188×10^{18}	2.3835×10^{18}	2.2086×10^{18}	1.9104×10^{18}
Std dev _{avg} (n=4)	0.0250×10^{18}	0.0510×10^{18}	0.0210×10^{18}	0.0183×10^{18}
Atoms after correction for neutron capture	2.7242×10^{18}	2.3854×10^{18}	2.2069×10^{18}	1.9105×10^{18}
Std dev, % (includes estimated uncertainties in corrections for neutron capture)	0.92	2.14	0.95	0.96
Number of fissions	$(4.138 \pm 0.045) \times 10^{19}$ (Ref 8)			
Percent fission yield	6.58	5.76	5.33	4.62
Std dev	0.09	0.14	0.08	0.07
Std dev, %	1.4	2.4	1.51	1.44

Table XII

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-233 FROM CAPSULE 6-4-2

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms	2.3734×10^{18}	2.0616×10^{18}	1.9619×10^{18}	1.6782×10^{18}
Std dev avg (n=3) ^[a]	0.0069×10^{18}	0.0062×10^{18}	0.0057×10^{18}	0.0049×10^{18}
Atoms after correction for neutron capture	2.3726×10^{18}	2.0620×10^{18}	1.9615×10^{18}	1.6782×10^{18}
Std dev, % (includes estimated uncertainties in corrections for neutron capture)	0.29	0.30	0.29	0.29
Number of fissions		$(3.786 \pm 0.059) \times 10^{19}$	(Ref 7)	
Percent fission yield	6.27	5.45	5.18	4.43
Std dev	0.10	0.09	0.08	0.07
Std dev, %	1.6	1.6	1.6	1.6

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

Table XIII

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-235 FROM CAPSULE 6-4-3

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms	2.0754×10^{18}	1.7807×10^{18}	1.6910×10^{18}	1.4382×10^{18}
Std dev avg (n=4)	0.0058×10^{18}	0.0053×10^{18}	0.0051×10^{18}	0.0046×10^{18}
Atoms after correction for neutron capture	2.0762×10^{18}	1.7811×10^{18}	1.6906×10^{18}	1.4382×10^{18}
Std dev, % (includes estimated uncertainties in corrections for neutron capture)	0.28	0.30	0.30	0.32
Number of fissions		$(3.390 \pm 0.062) \times 10^{19}$	(Ref 9)	
Percent fission yield	6.12	5.25	4.99	4.24
Std dev	0.11	0.10	0.09	0.08
Std dev, %	1.8	1.8	1.8	1.8

Table XIV

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-233 FROM CAPSULE 6-5-1

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms	1.7175×10^{18}	1.4789×10^{18}	1.4049×10^{18}	1.1952×10^{18}
Std dev avg (n=4)	0.0052×10^{18}	0.0044×10^{18}	0.0041×10^{18}	0.0038×10^{18}
Atoms after correction for neutron capture	1.7178×10^{18}	1.4791×10^{18}	1.4047×10^{18}	1.1952×10^{18}
Std dev, % (includes estimated uncertainties in corrections for neutron capture)	0.30	0.30	0.29	0.32
Number of fissions		$(2.614 \pm 0.055) \times 10^{19}$	(Ref 8)	
Percent fission yield	6.57	5.66	5.37	4.57
Std dev	0.14	0.12	0.11	0.10
Std dev, %	2.1	2.1	2.1	2.1

Table XV

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-235 FROM CAPSULE 6-2-2

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms	2.6614×10^{18}	2.4169×10^{18}	2.3812×10^{18}	2.5767×10^{18}
Std dev avg (n=4)	0.0071×10^{18}	0.0066×10^{18}	0.0054×10^{18}	0.0069×10^{18}
Atoms after correction for neutron capture	2.6665×10^{18}	2.4186×10^{18}	2.3793×10^{18}	2.5768×10^{18}
Std dev, % (includes estimated uncertainties in corrections for neutron capture)	0.27	0.27	0.27	0.27
Number of fissions	$(4.142 \pm 0.022) \times 10^{19}$ (Ref 12)			
Percent fission yield	6.44	5.84	5.74	6.22
Std dev	0.04	0.03	0.03	0.04
Std dev, %	0.6	0.6	0.6	0.6

Table XVI

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-235 FROM CAPSULE '6-4-1

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms	1.8363×10^{18}	1.6723×10^{18}	1.6451×10^{18}	1.7757×10^{18}
Std dev _{avg} (n=4)	0.0048×10^{18}	0.0043×10^{18}	0.0043×10^{18}	0.0046×10^{18}
Atoms after correction for neutron capture	1.8356×10^{18}	1.6726×10^{18}	1.6448×10^{18}	1.7755×10^{18}
Std dev, % (includes estimated uncertainties in corrections for neutron capture)	0.26	0.26	0.26	0.26
Number of fissions	$(2.839 \pm 0.032) \times 10^{19}$ (Ref 2)			
Percent fission yield	6.46	5.89	5.79	6.25
Std dev	0.07	0.07	0.07	0.07
Std dev, %	1.2	1.2	1.2	1.2

Table XVII

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF MOLYBDENUM FOR U-235 FROM CAPSULE 6-6-2

	Mo-95	Mo-97	Mo-98	Mo-100
Determined number of atoms	1.4385×10^{18}	1.3139×10^{18}	1.2966×10^{18}	1.4000×10^{18}
Std dev avg (n=4)	0.0039×10^{18}	0.0034×10^{18}	0.0034×10^{18}	0.0036×10^{18}
Atoms after correction for neutron capture	1.4391×10^{18}	1.3142×10^{18}	1.2963×10^{18}	1.3999×10^{18}
Std dev, % (includes estimated uncertainties in corrections for neutron capture)	0.27	0.26	0.26	0.26
Number of fissions		$(2.201 \pm 0.030) \times 10^{19}$	(Feb 9)	
Percent fission yield	6.54	5.97	5.89	6.36
Std dev	0.09	0.08	0.08	0.09
Std dev, %	1.4	1.4	1.4	1.4

Table XVIII

REPRODUCIBILITY OF THE MASS SPECTROMETRIC PROCEDURE FOR MOLYBDENUM

<u>Aliquot</u>	<u>Molybdenum Isotope (Atom/ml x 10⁻¹⁵)</u>			
	<u>95</u>	<u>97</u>	<u>98</u>	<u>100</u>
1	9.643	8.732	8.596	9.289
2	9.591	8.774	8.610	9.342
3	9.640	8.730	8.639	9.350
4	9.655	8.754	8.627	9.322
<hr/>				
Avg	9.632	8.747	8.618	9.326
Std dev ^[a]	0.028	0.021	0.019	0.027
Std dev, %	0.29	0.24	0.22	0.29

[a] Standard deviation for an individual result.

THERMAL FISSION YIELDS OF THE 95, 97, AND 99 MASS CHAINS

FOR Pu-239 AND Pu-241

(R. E. Foster, Jr.)

The thermal fission yields of the 95, 97, and 99 mass chains for Pu-239 and Pu-241 were obtained from the analysis of two series of 10-min irradiations of U-233, U-235, Pu-239, and Pu-241. The irradiations were at a flux of 10^{14} n/cm²-sec in the VH-1 position of the Materials Testing Reactor. Each series consisted of individual quartz-encapsulated samples of U-233 (8 mg as U₃O₈), U-235 (4 mg as U₃O₈), Pu-239 (3 mg as PuO₂), and Pu-241 (1.5 mg as PuO₂).

After irradiation, the individual capsules were scanned at numerous time intervals ranging from 1.5 to 100 days with a Ge(Li) detector and a 4096 pulse height analyzer system. At each time interval, all four capsules of a series were scanned. The activities of Zr-95, Nb-95, Zr-97, and Tc-99m were measured relative to Y-91m, La-140, Ba-140, and Ce-143 whose fission yields had previously been measured (Table XXXIX). The fission yield of the desired mass chain was calculated by ratioing the activity ratio of two selected photopeaks from the plutonium samples with the activity ratio of the two same photopeaks from the U-233 or U-235 sample. This technique, illustrated on the next page for the fission yield of Zr-95 for Pu-239, circumvents the uncertainties of absolute counting.

The data and calculated fission yields of the 95, 97, and 99 mass chains are given in Tables XIX, XX, and XXI for Pu-239 and in Tables XXII, XXIII, and XXIV for Pu-241.

$$FY_{95}^{239} = FY_{95}^{235} \left(\frac{Zr-95^{239}}{La-140^{239}} \right) \left(\frac{La-140^{235}}{Zr-95^{235}} \right) \left(\frac{FY_{140}^{239}}{FY_{140}^{235}} \right)$$

in which:

FY_{95}^{239} = fission yield of Zr-95 for Pu-239

FY_{95}^{235} = fission yield (known) of the 95 mass chain for U-235

$\frac{Zr-95^{239}}{La-140^{239}}$ = measured activity ratio of Zr-95 to La-140 for Pu-239 sample

$\frac{La-140^{235}}{Zr-95^{235}}$ = measured activity ratio of La-140 to Zr-95 for U-235 sample

FY_{140}^{239} = fission yield (known) of the 140 mass chain for Pu-239

FY_{140}^{235} = fission yield (known) of the 140 mass chain for U-235

Table XIX

THERMAL FISSION YIELD OF THE 95 MASS CHAIN FOR Pu-239

$\left(\frac{\text{Zr-95}^{239}}{\text{La-140}^{239}} \right) \left(\frac{\text{La-140}^{235}}{\text{Zr-95}^{235}} \right)$	$\frac{^{239}\text{[b]}}{\text{FY}_{140}}$	$\frac{^{235}\text{[b]}}{\text{FY}_{95}}$	$\frac{^{239}}{\text{FY}_{95}}$
0.8848	0.889±0.019	6.45±0.03	5.068
0.8285	0.889±0.019	6.45±0.03	4.745
0.8196	0.889±0.019	6.45±0.03	4.694
$\left(\frac{\text{Zr-95}^{239}}{\text{Ba-140}^{239}} \right) \left(\frac{\text{Ba-140}^{235}}{\text{Zr-95}^{235}} \right)$			
0.8721	0.889±0.019	6.45±0.03	4.994
0.8517	0.889±0.019	6.45±0.03	4.878
$\left(\frac{\text{Nb-95}^{239}}{\text{La-140}^{239}} \right) \left(\frac{\text{La-140}^{235}}{\text{Nb-95}^{235}} \right)$			
0.8551	0.889±0.019	6.45±0.03	4.898
$\left(\frac{\text{Nb-95}^{239}}{\text{Ba-140}^{239}} \right) \left(\frac{\text{Ba-140}^{235}}{\text{Nb-95}^{235}} \right)$			
0.8721	0.889±0.019	6.45±0.03	4.994
$\left(\frac{\text{Zr-95}^{239}}{\text{La-140}^{239}} \right) \left(\frac{\text{La-140}^{233}}{\text{Zr-95}^{233}} \right)$	$\frac{^{239}\text{[b]}}{\text{FY}_{140}}$	$\frac{^{233}\text{[b]}}{\text{FY}_{95}}$	
0.8919	0.859±0.019	6.40±0.05	4.898
0.8931	0.859±0.019	6.40±0.05	4.905
0.8455	0.859±0.019	6.40±0.05	4.643

Table XIX (Continued)

THERMAL FISSION YIELD OF THE 95 MASS CHAIN FOR Pu-239

$\left(\frac{\text{Zr-95}^{239}}{\text{Ba-140}^{239}} \right) \left(\frac{\text{Ba-140}^{233}}{\text{Zr-95}^{233}} \right)$	$\frac{\text{FY}_{140}^{239}[\text{b}]}{\text{FY}_{140}^{233}}$	$\frac{\text{FY}_{95}^{233}[\text{b}]}{\text{FY}_{95}^{239}}$	$\frac{\text{FY}_{95}^{239}}{\text{FY}_{95}^{233}}$
0.8721	0.859±0.019	6.40±0.05	4.789
0.9231	0.859±0.019	6.40±0.05	5.070
$\left(\frac{\text{Nb-95}^{239}}{\text{Ba-140}^{239}} \right) \left(\frac{\text{Ba-140}^{233}}{\text{Nb-95}^{233}} \right)$			
0.8973	0.859±0.019	6.40±0.05	4.927
$\left(\frac{\text{Nb-95}^{239}}{\text{La-140}^{239}} \right) \left(\frac{\text{La-140}^{233}}{\text{Nb-95}^{233}} \right)$			
0.8237	0.859±0.019	6.40±0.05	4.524

$$\text{Avg FY}_{95}^{239} = 4.86 \pm 0.11^{[\text{a}]}$$

[a] The errors assigned to the average value were obtained by assigning the variances in the activity ratios as random errors and the variances of the known fission yields as systematic errors.

[b] See Table XXXIX, this report, for a listing of fission yields.

Table XX

THERMAL FISSION YIELD OF THE 97 MASS CHAIN FOR Pu-239

36	$\left(\frac{\text{Zr-97}^{239}}{\text{Y-91m}^{239}} \right) \left(\frac{\text{Y-91m}^{235}}{\text{Zr-97}^{235}} \right)$	$\frac{\text{FY}_{91}^{239[b]}}{\text{FY}_{91}^{235}}$	$\frac{\text{FY}_{97}^{235[b]}}{\text{FY}_{97}^{235}}$	$\frac{\text{FY}_{97}^{239}}{\text{FY}_{97}^{235}}$
	2.3377	0.427±0.009	5.86±0.03	5.859
	$\left(\frac{\text{Zr-97}^{239}}{\text{Ce-143}^{239}} \right) \left(\frac{\text{Ce-143}^{235}}{\text{Zr-97}^{235}} \right)$	$\frac{\text{FY}_{143}^{239[b]}}{\text{FY}_{143}^{235}}$		
	1.1921	0.759±0.017	5.86±0.03	5.317
	1.2596	0.759±0.017	5.86±0.03	5.619
	$\left(\frac{\text{Zr-97}^{239}}{\text{Ce-143}^{239}} \right) \left(\frac{\text{Ce-143}^{233}}{\text{Zr-97}^{233}} \right)$	$\frac{\text{FY}_{143}^{239[b]}}{\text{FY}_{143}^{233}}$	$\frac{\text{FY}_{97}^{233[b]}}{\text{FY}_{97}^{233}}$	
	1.3690	0.766±0.018	5.52±0.05	5.801

$$\text{Avg FY}_{97}^{239} = 5.64 \pm 0.16^{[a]}$$

[a] The errors assigned to the average values were obtained by assigning the variances in the activity ratios as random errors and the variances of the known fission yields as systematic errors.

[b] See Table XXXIX, this report, for a listing of fission yields.

Table XXI

THERMAL FISSION YIELD OF THE 99 MASS CHAIN FOR Pu-239

$\left(\frac{\text{Tc-99m}^{239}}{\text{Y-91m}^{239}} \right) \left(\frac{\text{Y-91m}^{235}}{\text{Tc-99m}^{235}} \right)$	$\frac{{}^{239}_{91}\text{FY}}{\text{FY}_{91}^{235}}$		
2.6435	0.427±0.009	$\text{FY}_{99}^{235}[\text{b}]$ 6.14±0.10	FY_{99}^{239} 6.941
$\left(\frac{\text{Tc-99m}^{239}}{\text{Ce-143}^{239}} \right) \left(\frac{\text{Ce-143}^{235}}{\text{Tc-99m}^{235}} \right)$	$\frac{{}^{239}_{143}\text{FY}}{\text{FY}_{143}^{235}}$		
1.3212	0.759±0.017	6.14±0.10	6.175
1.3438	0.759±0.017	6.14±0.10	6.280
1.2596	0.759±0.017	6.14±0.10	5.886
1.4076	0.759±0.017	6.14±0.10	6.579
$\left(\frac{\text{Tc-99m}^{239}}{\text{La-140}^{239}} \right) \left(\frac{\text{La-140}^{235}}{\text{Tc-99m}^{235}} \right)$	$\frac{{}^{239}_{140}\text{FY}}{\text{FY}_{140}^{235}}$		
1.1855	0.889±0.019	6.14±0.10	6.463
1.3021	0.889±0.019	6.14±0.10	7.099
1.1258	0.889±0.019	6.14±0.10	6.138
$\left(\frac{\text{Tc-99m}^{239}}{\text{Ba-140}^{239}} \right) \left(\frac{\text{Ba-140}^{235}}{\text{Tc-99m}^{235}} \right)$			
1.2342	0.889±0.019	6.14±0.10	6.729
1.1701	0.889±0.019	6.14±0.10	6.380

Table XXI (Continued)

THERMAL FISSION YIELD OF THE 99 MASS CHAIN FOR Pu-239

$\left(\frac{\text{Tc-99m}^{239}}{\text{Ce-143}^{239}}\right)\left(\frac{\text{Ce-143}^{233}}{\text{Tc-99m}^{233}}\right)$	$\frac{\text{FY}_{143}^{239[b]}}{\text{FY}_{143}^{233}}$	$\frac{\text{FY}_{99}^{233[b]}}{\text{FY}_{99}^{239}}$	$\frac{\text{FY}_{99}^{239}}{\text{FY}_{99}^{233}}$
1.8148	0.766±0.018	5.16±0.07	7.188
1.6290	0.766±0.018	5.16±0.07	6.824
$\left(\frac{\text{Tc-99m}^{239}}{\text{La-140}^{239}}\right)\left(\frac{\text{La-140}^{233}}{\text{Tc-99m}^{233}}\right)$	$\frac{\text{FY}_{140}^{239[b]}}{\text{FY}_{140}^{233}}$	$\frac{\text{FY}_{99}^{233[b]}}{\text{FY}_{99}^{239}}$	$\frac{\text{FY}_{99}^{239}}{\text{FY}_{99}^{233}}$
1.5063	0.859±0.019	5.16±0.07	6.669
1.4936	0.859±0.019	5.16±0.07	6.614
$\left(\frac{\text{Tc-99m}^{239}}{\text{Ba-140}^{239}}\right)\left(\frac{\text{Ba-140}^{233}}{\text{Tc-99m}^{233}}\right)$			
1.6269	0.859±0.019	5.16±0.07	7.204
Avg $\text{FY}_{99}^{239} = 6.59 \pm 0.18^{[a]}$			

[a] The errors assigned to the average values were obtained by assigning the variances in the activity ratios as random errors and the variances of the known fission yields as systematic errors.

[b] See Table XXXIX, this report, for a listing of fission yields.

Table XXII

THERMAL FISSION YIELD OF THE 95 MASS CHAIN FOR Pu-241

$\left(\frac{\text{Zr-95}^{241}}{\text{La-140}^{241}} \right) \left(\frac{\text{La-140}^{235}}{\text{Zr-95}^{235}} \right)$	$\begin{matrix} {}^{241}[\text{b}] \\ \text{FY}_{140} \\ \hline {}^{235} \\ \text{FY}_{140} \end{matrix}$	$\begin{matrix} {}^{235}[\text{b}] \\ \text{FY}_{95} \end{matrix}$	FY_{95}^{241}
0.6673	0.929±0.026	6.45±0.03	3.997
0.6622	0.929±0.026	6.45±0.03	3.967
0.6584	0.929±0.026	6.45±0.03	3.944
$\left(\frac{\text{Zr-95}^{241}}{\text{Ba-140}^{241}} \right) \left(\frac{\text{Ba-140}^{235}}{\text{Zr-95}^{235}} \right)$			
0.6619	0.929±0.026	6.45±0.03	3.965
0.6516	0.929±0.026	6.45±0.03	3.903
$\left(\frac{\text{Nb-95}^{241}}{\text{La-140}^{241}} \right) \left(\frac{\text{La-140}^{235}}{\text{Nb-95}^{235}} \right)$			
0.6588	0.929±0.026	6.45±0.03	3.946
$\left(\frac{\text{Nb-95}^{241}}{\text{Ba-140}^{241}} \right) \left(\frac{\text{Ba-140}^{235}}{\text{Nb-95}^{235}} \right)$			
0.6535	0.929±0.026	6.45±0.03	3.914

Table XXII (Continued)

THERMAL FISSION YIELD OF THE 95 MASS CHAIN FOR Pu-241

07	$\left(\frac{\text{Zr-95}^{241}}{\text{La-140}^{241}} \right) \left(\frac{\text{La-140}^{233}}{\text{Zr-95}^{233}} \right)$	$\frac{\text{FY}_{140}^{241[b]}}{\text{FY}_{140}^{233}}$	$\frac{\text{FY}_{95}^{233[b]}}{\text{FY}_{95}^{241}}$	$\frac{\text{FY}_{95}^{241}}{\text{FY}_{95}^{233}}$
	0.6726	0.897±0.025	6.40±0.05	3.863
	0.7139	0.897±0.025	6.40±0.05	4.100
	0.6793	0.897±0.025	6.40±0.05	3.901
	$\left(\frac{\text{Zr-95}^{241}}{\text{Ba-140}^{241}} \right) \left(\frac{\text{Ba-140}^{233}}{\text{Zr-95}^{233}} \right)$	$\frac{\text{FY}_{140}^{241[b]}}{\text{FY}_{140}^{233}}$	$\frac{\text{FY}_{95}^{233[b]}}{\text{FY}_{95}^{241}}$	$\frac{\text{FY}_{95}^{241}}{\text{FY}_{95}^{233}}$
	0.6811	0.897±0.025	6.40±0.05	3.912
	0.7062	0.897±0.025	6.40±0.05	4.056
	$\left(\frac{\text{Nb-95}^{241}}{\text{La-140}^{241}} \right) \left(\frac{\text{La-140}^{233}}{\text{Nb-95}^{233}} \right)$			
	0.6346	0.897±0.025	6.40±0.05	3.645
	$\left(\frac{\text{Nb-95}^{241}}{\text{Ba-140}^{241}} \right) \left(\frac{\text{Ba-140}^{233}}{\text{Nb-95}^{233}} \right)$			
	0.6426	0.897±0.025	6.40±0.05	3.691
	$\text{Avg FY}_{95}^{241} = 3.92 \pm 0.09 [a]$			

[a] The errors assigned to the average values were obtained by assigning the variances in the activity ratios as random errors and the variances of the known fission yields as systematic errors.

[b] See Table XXXIX, this report, for a listing of fission yields.

Table XXIII

THERMAL FISSION YIELD OF THE 97 MASS CHAIN FOR Pu-241

14	$\left(\frac{\text{Zr-97}^{241}}{\text{Y-91m}^{241}} \right) \left(\frac{\text{Y-91m}^{235}}{\text{Zr-97}^{235}} \right)$	$\frac{\text{FY}_{91}^{241[b]}}{\text{FY}_{91}^{235}}$	$\frac{\text{FY}_{97}^{235[b]}}{\text{FY}_{97}^{241}}$	
	2.6205	0.308±0.009	5.86±0.03	4.737
	$\left(\frac{\text{Zr-97}^{241}}{\text{Ce-143}^{241}} \right) \left(\frac{\text{Ce-143}^{235}}{\text{Zr-97}^{235}} \right)$	$\frac{\text{FY}_{143}^{241[b]}}{\text{FY}_{143}^{235}}$		
	1.0455 1.0275	0.759±0.021 0.759±0.021	5.86±0.03 5.86±0.03	4.652 4.572
	$\left(\frac{\text{Zr-97}^{241}}{\text{Ce-143}^{241}} \right) \left(\frac{\text{Ce-143}^{233}}{\text{Zr-97}^{233}} \right)$	$\frac{\text{FY}_{143}^{241[b]}}{\text{FY}_{143}^{233}}$	$\frac{\text{FY}_{97}^{233[b]}}{\text{FY}_{97}^{241}}$	
	1.2006	0.766±0.022	5.52±0.05	5.075
<hr/>		$\text{Avg FY}_{97}^{241} = 4.76 \pm 0.14^{[a]}$		

[a] The errors assigned to the average values were obtained by assigning the variances in the activity ratios as random errors and the variances of the known fission yields as systematic errors.

[b] See Table XXXIX, this report, for a listing of fission yields.

Table XXIV

THERMAL FISSION YIELDS OF THE 99 MASS CHAIN FOR Pa-241

$\frac{\left(\frac{\text{Tc-99m}^{241}}{\text{Y-91m}^{241}}\right)\left(\frac{\text{Y-91m}^{235}}{\text{Tc-99m}^{235}}\right)}{3.1638}$	$\frac{\frac{241[\text{b}]}{\text{FY}_{91}}}{\frac{235}{\text{FY}_{91}}}$	$\frac{235[\text{b}]}{\text{FY}_{99}}$	$\frac{\text{FY}_{99}^{241}}{\text{FY}_{99}}$
	0.308±0.009	6.14±0.10	5.993
$\frac{\left(\frac{\text{Tc-99m}^{241}}{\text{Ce-143}^{241}}\right)\left(\frac{\text{Ce-143}^{235}}{\text{Tc-99m}^{235}}\right)}{1.3575}$	$\frac{\frac{241[\text{b}]}{\text{FY}_{143}}}{\frac{235}{\text{FY}_{143}}}$		
1.2249	0.759±0.021	6.14±0.10	6.329
1.2404	0.759±0.021	6.14±0.10	5.711
1.3301	0.759±0.021	6.14±0.10	5.783
	0.759±0.021	6.14±0.10	6.201
$\frac{\left(\frac{\text{Tc-99m}^{241}}{\text{La-140}^{241}}\right)\left(\frac{\text{La-140}^{235}}{\text{Tc-99m}^{235}}\right)}{1.1033}$	$\frac{\frac{241[\text{b}]}{\text{FY}_{140}}}{\frac{235}{\text{FY}_{140}}}$		
1.1084	0.929±0.026	6.14±0.10	6.291
1.0169	0.929±0.026	6.14±0.10	6.320
	0.929±0.026	6.14±0.10	5.799
$\frac{\left(\frac{\text{Tc-99m}^{241}}{\text{Ba-140}^{241}}\right)\left(\frac{\text{Ba-140}^{235}}{\text{Tc-99m}^{235}}\right)}{1.0528}$			
1.0062	0.929±0.026	6.14±0.10	6.003
	0.929±0.026	6.14±0.10	5.738

Table XXIV (Continued)

THERMAL FISSION YIELDS OF THE 99 MASS CHAIN FOR Pu-241

$\left(\frac{\text{Tc-99m}^{241}}{\text{Ce-143}^{241}} \right) \left(\frac{\text{Ce-143}^{233}}{\text{Tc-99m}^{233}} \right)$	$\frac{^{241}\text{[b]}}{\text{FY}_{143}}$	$\frac{\text{FY}_{143}^{233}}{\text{FY}_{143}}$	$\frac{\text{FY}_{99}^{233}\text{[b]}}{\text{FY}_{99}}$	$\frac{\text{FY}_{99}^{241}}{\text{FY}_{99}}$
1.8646	0.766±0.022	5.16±0.07	7.368	
1.4849	0.766±0.022	5.16±0.07	5.868	
$\left(\frac{\text{Tc-99m}^{241}}{\text{La-140}^{241}} \right) \left(\frac{\text{La-140}^{233}}{\text{Tc-99m}^{233}} \right)$	$\frac{^{241}\text{[b]}}{\text{FY}_{140}}$			
1.4019	0.897±0.025	5.16±0.07	6.492	
1.3491	0.897±0.025	5.16±0.07	6.247	
$\left(\frac{\text{Tc-99m}^{241}}{\text{Ba-140}^{241}} \right) \left(\frac{\text{Ba-140}^{233}}{\text{Tc-99m}^{233}} \right)$				
1.3989	0.897±0.025	5.16±0.07	6.478	

$$\text{Avg FY}_{99}^{241} = 6.17 \pm 0.16^{[a]}$$

[a] The errors assigned to the average values were obtained by assigning the variances in the activity ratios as random errors and the variances of the known fission yields as systematic errors.

[b] See Table XXXIX, this report, for a listing of fission yields.

THERMAL FISSION YIELDS OF THE STABLE AND LONG LIVED ISOTOPES
OF RUTHENIUM FOR Pu-239 AND Pu-241

During the dissolution of the standard Pu-239 and Pu-241 burnup capsules, ruthenium partially precipitated from solution. This prevented obtaining ruthenium fission yields from these capsules. However, sufficient ruthenium remained in solution to measure the relative isotopic distributions of the ruthenium isotopes. These measurements were combined with radiometric measurements of the Ru-106 fission yields for Pu-239 and Pu-241 to calculate the fission yields of Ru-101, -102, and -104 for both Pu-239 and Pu-241.

Mass Spectrometric Determined Ratios (R. E. Foster, Jr., R. M. Abernathey, R. E. McAtee, G. D. Workman)

From aliquots of the dissolver solutions of standard burnup capsules CPP-6-2-5, 6-2-6, and 6-5-5 for Pu-239 and capsules CPP-6-4-6 and 6-5-6 for Pu-241, ruthenium was separated by distillation (3) and mass spectrometrically analyzed for the relative isotope abundances. The measured ratios of Ru-101/Ru-106, Ru-102/Ru-106, and Ru-104/Ru-106 with the Ru-106 value first corrected for in-pile and out-of-pile decay are listed in Table XXV for Pu-239 and Table XXVI for Pu-241. The average values and their associated errors were obtained by weighting the results from the individual capsules as the reciprocals of their variances.

Radiometric Determined Ru-106 Fission Yields for Pu-239 and Pu-241
(R. E. Foster, Jr., M. E. Kussy)

Aliquots of the dissolved irradiated Pu-239 and Pu-241 samples used for the Sb-125 measurements (see page 15) were analyzed for Ru-106 and Cs-137. The separation procedure for ruthenium and the radiometric procedure for Cs-137 have been described (1, 3).

For the Pu-241 samples, the aliquots were taken shortly after irradiation and the Ru-106 and Cs-137 activities were measured with a Ge(Li) detector-pulse height analyzer system. The Ru-106 activity was measured relative to a Ru-106 sample that had been calibrated for atom concentration to $\pm 0.46\%$ by isotope dilution mass spectrometry giving a direct measure of Ru-106 atoms independent of detector parameters. The measured Cs-137 activities on the Ge(Li) system were converted to atoms by direct comparison to a pure Cs-137 sample that had been calibrated on a 3- x 3-in. NaI(Tl) crystal.

For the Pu-239 sample, aliquots were taken about one year after irradiation, to allow Ru-103 to decay, and the Ru-106 and Cs-137 activities were determined with a NaI(Tl)-pulse height analyzer system. As with the Pu-241 samples, the Ru-106 activities were measured relative to a Ru-106 mass spectrometric standardized sample to provide results directly in atoms. During the dissolution of the Pu-239 samples, some ruthenium plated or precipitated from solution. The relative amount of ruthenium that deposited was measured by counting the Ru-106 activity in the dissolver flask plus contents, decanting the liquid, and counting the "empty" flask. The fission yield of Ru-106 was calculated by multiplying the atom ratio of Ru-106/Cs-137 by the previously determined Cs-137 fission yield (Table XXXIX). The experimental data and calculated fission yields are given in Table XXVII for Pu-239 and Table XXVIII for Pu-241. To confirm the Ru-106 fission yields, data obtained from the ratio method described on page 32 were used to calculate the Ru-106 fission yield for Pu-239 and Pu-241. The ratio Ru-106/Ce-144 was used and a fission yield value for Ru-106 of 4.92 ± 0.23 and 6.29 ± 0.23 for Pu-239 and Pu-241 respectively were obtained. This compares with values of 4.55 ± 0.19 and 6.08 ± 0.25 obtained by the present chemical separation method.

The fission yield for Ru-101, -102, and -104 were calculated by multiplying the mass spectrometrically determined Ru-101/Ru-106, Ru-102/Ru-106, and Ru-104/Ru-106 ratios by the now determined Ru-106 fission yield. These fission yields are given in Table XXIX for Pu-239 and Table XXX for Pu-241.

Table XXV

THERMAL IRRADIATION RATIOS OF RU-101, -102, AND -104 TO Ru-106 FOR Pu-239^[a]

<u>Capsule</u>	<u>Ru-101/Ru-106</u>	<u>Ru-102/Ru-106</u>	<u>Ru-104/Ru-106</u>
6-2-5	1.386±0.047	1.427±0.049	1.418±0.048
6-2-6	1.414±0.026	1.454±0.027	1.444±0.027
6-5-5	<u>1.451±0.025</u>	<u>1.479±0.025</u>	<u>1.469±0.025</u>
$\bar{X}$	1.428	1.462	1.452
$\sigma\bar{X}$	0.017	0.017	0.017
$\% \sigma\bar{X}$	1.2	1.2	1.2

[a] The error associated with each capsule is the standard deviation of the mean value for four determinations and includes the uncertainty in the Ru-106 decay correction.

Table XXVI

THERMAL IRRADIATION RATIOS OF Ru-101, -102, AND -106 TO Ru-106 FOR Pu-241^[a]

<u>Capsule</u>	<u>Ru-101/Ru-106</u>	<u>Ru-102/Ru-106</u>	<u>Ru-104/Ru-106</u>
6-4-6	0.974±0.025	1.040±0.027	1.117±0.029
6-5-6	0.981±0.022	1.040±0.023	1.119±0.025
$\bar{x}$	0.978	1.040	1.118
$\sigma\bar{x}$	0.017	0.018	0.019
$\%\sigma\bar{x}$	1.7	1.7	1.7

[a] The error associated with each capsule is the standard deviation of the mean value for four determinations and includes the uncertainty in the Ru-106 decay correction.

Table XXVII

THERMAL FISSION YIELD OF Ru-106 FOR Pu-239

	<u>Capsule 423</u>	<u>Capsule 424</u>
Ru-106, c/m/ml ^[a]	$(6.907 \pm 0.202) \times 10^3$	$(5.945 \pm 0.242) \times 10^3$
Ru-106 std, atom/c	$(1.013 \pm 0.011) \times 10^9$	
Ru-106, atom/ml	$(6.997 \pm 0.218) \times 10^{12}$	$(6.022 \pm 0.253) \times 10^{12}$
Cs-137, c/m/ml	$(3.927 \pm 0.035) \times 10^3$	$(3.298 \pm 0.082) \times 10^3$
Cs-137 detector and nuclear constants [(TPA)(λ)(BR)]	$(3.761 \pm 0.082) \times 10^{-10}$	
Cs-137, atom/ml	$(1.044 \pm 0.024) \times 10^{13}$	$(0.8769 \pm 0.0290) \times 10^{13}$
Ru-106/Cs-137 atom ratio	0.670 ± 0.026	0.687 ± 0.037
Avg Ru-106/Cs-137 atom ratio ^[b]	0.675 ± 0.024	
Cs-137 FY, %	6.74 ± 0.14 ^[c]	
Ru-106 FY, %	4.55 ± 0.19	

[a] Corrected for in-pile and out-of-pile decay and any ruthenium deposited during the dissolution.

[b] Individual ratios were weighted as the reciprocals of their random variances.

[c] This report, Table XXXIX.

Table XXVIII

THERMAL FISSION YIELD OF Ru-106 FOR Pu-241

	<u>Capsule 421</u>	<u>Capsule 422</u>
Ru-106 c/m/ml ^[a]	223.0±2.5	234.6±2.4
Ru-106 std, atom/c	(3.189±0.015)×10 ¹⁰	
Ru-106, atom/ml	(7.111±0.086)×10 ¹²	(7.481±0.084)×10 ¹²
Cs-137, c/m/ml ^[a]	283.8±3.6	286.9±4.4
Cs-137 detector and nuclear constants [(TPA)(λ)(BR)]	(3.61±0.11)×10 ⁻¹¹	
Cs-137, atom/ml	(7.861±0.259)×10 ¹²	(7.947±0.271)×10 ¹²
Ru-106/Cs-137 atom ratio	0.905±0.032	0.941±0.033
Avg Ru-106/Cs-137 atom ratio ^[b]	0.921±0.030	
Cs-137 FY, %	6.60±0.17 ^[c]	
Ru-106 FY, %	6.08±0.25	

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

[b] Individual ratios were weighted as the reciprocals of their random variances.

[c] This report Table XXXIX.

Table XXIX

THERMAL FISSION YIELDS OF THE STABLE RUTHENIUM ISOTOPES FOR Pu-239

	<u>Ru-101</u>	<u>Ru-102</u>	<u>Ru-104</u>
Ratio to Ru-106	1.428±0.017	1.462±0.017	1.452±0.017
Ru-106 fission yield (%)		4.55±0.19	
Fission yield (%)	6.53±0.28	6.65±0.29	6.61±0.29

Table XXX

THERMAL FISSION YIELDS OF THE STABLE RUTHENIUM ISOTOPES FOR Pu-241

	<u>Ru-101</u>	<u>Ru-102</u>	<u>Ru-104</u>
Ratio to Ru-106	0.977±0.017	1.040±0.018	1.118±0.019
Ru-106 fission yield (%)		6.08±0.25	
Fission yield (%)	5.94±0.26	6.32±0.28	6.80±0.30

THERMAL FISSION YIELD OF Ce-141 FOR U-235
(R. E. Foster, Jr.)

The thermal fission yield of Ce-141 was measured on four quartz encapsulated U-235 foils which had been irradiated similarly to the Pu-239 and Pu-241 capsules described in a previous section on the thermal fission yields of Sb-125. The uranium capsules were treated in the same manner as the Pu-239 and Pu-241 capsules (see page 15) except that a mixture of 5M HCl-1M HNO₃-1M HF was used for the dissolution. Cesium and cerium were individually separated (1, 14) in quadruplicate for each dissolved U-235 foil. The separated samples were gamma counted with a Ge(Li) detector-pulse height analyzer system and the numbers of atoms of Ce-141/ml and Cs-137/ml were determined for each of the dissolved U-235 foils. The Ce-141 fission yield was calculated by multiplying the Ce-141/Cs-137 atom ratio by the known fission yield of Cs-137. The detector parameters at the 145-keV photopeak of Ce-141 were obtained by cross calibrating the Ge(Li) detector with a 3- x 3-in. NaI(Tl) crystal with known parameters. The branching ratio used for the 145-keV transition was $0.454 \pm 0.023(15)$. The detector parameters for Cs-137 were obtained as described in the previous section (page 44) on the Ru-106 fission yields. The experimental data and calculated fission yield of Ce-141 for U-235 are given in Table XXXI.

Table XXXI

THERMAL FISSION YIELD OF Ce-141 FOR U-235

	<u>Capsule 387</u>	<u>Capsule 388</u>	<u>Capsule 398</u>	<u>Capsule 399</u>
Ce-141 activity (counts/ min/ml)[a]	$(1.146 \pm 0.006) \times 10^5$	$(8.766 \pm 0.087) \times 10^4$	$(1.045 \pm 0.010) \times 10^5$	$(1.191 \pm 0.004) \times 10^5$
Ce-141 detector parameters and nuclear constants [(λ)(TPA)(BR)]		$(9.447 \pm 0.514) \times 10^{-9}$		
Atoms of Ce-141/ml	$(1.213 \pm 0.066) \times 10^{13}$	$(9.279 \pm 0.513) \times 10^{12}$	$(1.106 \pm 0.061) \times 10^{13}$	$(1.261 \pm 0.069) \times 10^{13}$
Cs-137 activity (counts/ min/ml)[a]	$(4.988 \pm 0.011) \times 10^2$	$(4.009 \pm 0.020) \times 10^2$	$(4.642 \pm 0.012) \times 10^2$	$(5.032 \pm 0.032) \times 10^2$
Cs-137 detector parameters and nuclear constants [(λ)(TPA)(BR)]		$(3.608 \pm 0.108) \times 10^{-11}$		
Atoms of Cs-137/ml	$(1.382 \pm 0.013) \times 10^{13}$	$(1.111 \pm 0.034) \times 10^{13}$	$(1.287 \pm 0.039) \times 10^{13}$	$(1.395 \pm 0.043) \times 10^{13}$
<u>Atoms Ce-141</u> <u>Atoms Cs-137</u>	0.878 ± 0.055	0.835 ± 0.053	0.859 ± 0.054	0.904 ± 0.057
Avg <u>atoms Ce-141</u> ^[b] <u>atoms Cs-137</u>		0.875 ± 0.054		
Fission yield of Cs-137, %		6.28 ± 0.03 ^[c]		
Fission yield of Ce-141, %		5.50 ± 0.34		

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

[b] The average value (and associated standard deviation) was obtained by error propagation in which the individual values were weighted as the reciprocals of the random variances and the uncertainties in the detector parameters and nuclear constants were systematic.

[c] This report Table XXXIX.

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF SAMARIUM
FOR Pu-239

The thermal fission yields of the stable isotopes of samarium were measured on three thermal Pu-239 burnup capsules. The irradiation histories, dissolution procedures, and calculated numbers of fissions for these capsules have been previously reported (5). The chemical separation and mass spectrometric procedures were the same as previously reported (5).

The experimental data and the calculated fission yields are summarized in Tables XXXII, XXXIII, and XXXIV. The average values and associated errors were obtained by weighting the individual values as the reciprocals of their variances. For comparison purposes, the mass spectrometric determined Pu-239 fission yield values of Fickel and Tomlinson (16) have been included in Table XXXV.

The large capture cross sections on certain samarium isotopes required corrections for buildup and burnout. These corrections were made with the aid of a computer calculation which simulated the irradiation histories of the capsules. A detailed discussion of these calculations has been previously reported (9). Corrections also were necessary for fission product contributions from Pu-241. These corrections were obtained from the calculated fission ratio of Pu-241 to Pu-239 and the reported heavy mass Pu-241 fission yields of Farrar et al (17).

Most of the Sm-149 is converted to Sm-150 during the irradiation. Rather than apply large corrections, the measured atoms of Sm-149 and Sm-150 have been summed because Sm-150 is a shielded nuclide not directly formed in fission. Small corrections were required, however, for the loss of Sm-150 by neutron capture.

Table XXXII

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF SAMARIUM FOR Pu-239 FROM CAPSULE CPP 6-2-5

Number of Fissions = $(4.001 \pm 0.066) \times 10^{19}$ (Ref 5)

	Sm-147	Sm-149	Sm-150	Sm-151	Sm-152	Sm-154
Determined number of atoms	3.7019×10^{17}	1.7330×10^{16}	4.6946×10^{17}	3.6670×10^{16}	4.5153×10^{17}	1.0497×10^{17}
Std dev avg (n = 3)	0.0185×10^{17}	0.0199×10^{16}	0.0333×10^{17}	0.0271×10^{16}	0.0384×10^{17}	0.0109×10^{17}
Atoms after correction for neutron capture and radioactive decay	8.4707×10^{17}	4.8109×10^{17}	None	3.3890×10^{17}	2.2316×10^{17}	1.0514×10^{17}
Std dev, % (includes estimated uncertainties in corrections for neutron capture and radioactive decay)	3.04	0.68		5.05	5.07	1.04
Calculated fission yield, %	2.12	1.21		0.847	0.558	0.262
<u>Pu-241 fissions</u> <u>Pu-239 fissions</u>			0.0166			
Assumed Pu-241 fission yield, %	2.33	1.55		0.959	0.757	0.408
Pu-239 fission yield, %	2.08	1.18		0.831	0.545	0.255
Std dev	0.07	0.02		0.044	0.029	0.005

Table XXXIII

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF SAMARIUM FOR Pu-239 FROM CAPSULE CPP 6-2-6

Number of Fissions = $(4.448 \pm 0.067) \times 10^{19}$ (Ref 5)

	Sm-147	Sm-149	Sm-150	Sm-151	Sm-152	Sm-154
Determined number of atoms	4.3347×10^{17}	1.9292×10^{16}	5.3736×10^{17}	4.2418×10^{16}	5.1736×10^{17}	1.2081×10^{17}
Std dev avg (n = 3)	0.0221×10^{17}	0.0228×10^{16}	0.0559×10^{17}	0.0433×10^{16}	0.0662×10^{17}	0.0104×10^{17}
Atoms after correction for neutron capture and radioactive decay	9.6820×10^{17}	5.5387×10^{17}	None	3.7101×10^{17}	2.5578×10^{17}	1.2100×10^{17}
Std dev, % (includes estimated uncertainties in corrections for neutron capture and radioactive decay)	3.04	1.00		5.10	5.16	0.86
Calculated fission yield, %	2.18	1.24		0.834	0.575	0.272
<u>Pu-241 fissions</u> Pu-239 fissions			0.0158			
Assumed Pu-241 fission yield, %	2.33	1.55		0.959	0.757	0.408
Pu-239 fission yield, %	2.14	1.22		0.819	0.563	0.266
Std dev	0.07	0.02		0.043	0.030	0.005

Table XXXIV

THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF SAMARIUM FOR Pu-239 FROM CAPSULE CPP 6-5-5

Number of Fissions = $(2.065 \pm 0.046) \times 10^{19}$ (Ref 5)

	Sm-147	Sm-149	Sm-150	Sm-151	Sm-152	Sm-154
Determined number of atoms	1.8938×10^{17}	4.3182×10^{16}	2.0515×10^{17}	8.1177×10^{16}	1.8974×10^{17}	5.5887×10^{16}
Std dev avg (n = 4)	0.0161×10^{17}	0.0121×10^{16}	0.0049×10^{17}	0.0203×10^{16}	0.0045×10^{17}	0.0654×10^{16}
Atoms after correction for neutron capture and radioactive decay	4.1968×10^{17}	2.4873×10^{17}	None	1.5454×10^{17}	1.1876×10^{17}	5.5901×10^{16}
Std dev, % (includes estimated uncertainties in corrections for neutron capture and radioactive decay)	3.12	0.20		5.01	5.01	1.17
Calculated fission yield, %	2.03	1.20		0.743	0.575	0.271
<u>Pu-241 fissions</u> Pu-239 fissions			0.0011			
Assumed Pu-241 fission yield, %	2.33	1.55		0.959	0.757	0.408
Pu-239 fission yield, %	2.03	1.20		0.747	0.574	0.270
Std dev	0.08	0.03		0.041	0.032	0.007

Table XXXV.

AVERAGE THERMAL FISSION YIELDS OF THE STABLE ISOTOPES OF SAMARIUM
FOR Pu-239

<u>Isotope</u>	<u>Fission Yield, %</u>	
	<u>This Work</u>	<u>Fickel and Tomlinson</u> ^[a]
Sm-147	2.09±0.04	1.99
Sm-149	1.20±0.01	1.30
Sm-151	0.797±0.024	0.802
Sm-152	0.560±0.017	0.616
Sm-154	0.262±0.003	0.293

[a] Reported uncertainty of 2% (relative) on each value.

NORMALIZED THERMAL FISSION YIELDS FOR Pu-239

Our previously reported thermal fission yields for Pu-239 had been determined on an absolute basis on three irradiated capsules. The atoms of individual nuclides had been measured on the dissolver solutions, usually by isotope dilution mass spectrometry. The number of fissions had been measured by the loss of plutonium as the difference in the measured loaded atoms and the measured atoms in the dissolver solutions, supplemented by computer-calculated buildup and burnout corrections. The fission yields then were directly calculated as the ratios of these independent experimental measurements.

The values so obtained are an average 2 relative percent lower than those values reported by others (16, 18, 19) and show a definite low correlation when compared to our measured fast fission yields for Pu-239. A thorough investigation of the experimental procedures and data has been made with no apparent cause evident. The most likely cause is the experimentally measured preirradiation numbers of atoms in the capsules because this phase of the experiment had the greatest uncertainty.

For this reason, the normalization technique that had proven so successful for calculating the fast fission yields for U-235 and Pu-239 was applied to the Pu-239 thermal data. As reported in the previous section, on the fast fission yields for Pu-239, the fission yields relative to Nd-148 are computed and the heavy mass peak is normalized to 100%. For those mass chains for which experimental data were not obtained, literature (19) values were used each with an assigned uncertainty of 20 relative percent. About 80% of the normalization was with experimental values and 20% with literature values.

The fission yields obtained by this normalization technique are 2 relative percent higher and are recommended over our previously reported values.

The relative (to Nd-148) fission yields are summarized in Table XXXVI and the normalized fission yields are summarized in Table XXXVII. The uncertainties assigned to each normalized fission yield value are in terms of the standard deviation obtained by error propagation in the same way (6) as for the fast U-235 and Pu-239 data.

Table XXXVI

THERMAL FISSION YIELDS RELATIVE TO Nd-148 FOR Pu-239

Nuclide	Standard Capsule CPP-6-2-5	Standard Capsule CPP-6-2-6	Standard Capsule CPP-6-5-5	Average Value ^[a]
Kr-83	0.1731±0.0035	0.1753±0.0037	0.1841±0.0038	0.1772±0.0021
Kr-84	0.2769±0.0057	0.2859±0.0060	0.3002±0.0062	0.2870±0.0034
Rb + Kr-85	0.3360±0.0038	0.3354±0.0030	0.3441±0.0035	0.3383±0.0020
Kr-86	0.4377±0.0089	0.4546±0.0095	0.4701±0.0097	0.4532±0.0054
Rb-87	0.5892±0.0054	0.5857±0.0051	0.5907±0.0052	0.5885±0.0030
Sr-88	0.7974±0.0091	0.7882±0.0105	[b]	0.7935±0.0069
Sr-90	1.2344±0.0094	1.2308±0.0110	[b]	1.2329±0.0071
Zr-91	1.4767±0.0086	1.4769±0.0102	1.4933±0.0094	1.4822±0.0054
Zr-92	1.7107±0.0099	1.8106±0.0126	1.8393±0.0116	1.7769±0.0065
Zr-93	2.3243±0.0135	2.3048±0.0160	2.3405±0.0148	2.3241±0.0085
Zr-94	2.6359±0.0153	2.6391±0.0183	2.6792±0.0169	2.6509±0.0096
Zr-96	3.0012±0.0174	2.9976±0.0208	3.0455±0.0192	3.0146±0.0110
118-130				3.74±0.75 ^[c]
Xe-131	2.1095±0.0431	2.1278±0.0446	2.1307±0.0438	2.1225±0.0253
Xe-132	3.0312±0.0619	3.0646±0.0642	2.9197±0.0600	3.0019±0.0358

Table XXXVI (Continued)

THERMAL FISSION YIELDS RELATIVE TO Nd-148 FOR Pu-239

Nuclide	Standard Capsule CPP-6-2-5	Standard Capsule CPP-6-2-6	Standard Capsule CPP-6-5-5	Average Value ^[a]
Cs-133	4.1481±0.0305	4.1923±0.0356	4.2865±0.0222	4.2292±0.0160
Xe-134	4.2454±0.0867	4.2805±0.0896	4.1950±0.0862	4.2393±0.0505
Xe-135 + 136	8.2411±0.1682	8.3000±0.1738	8.0413±0.1652	8.1902±0.0975
Cs-137	3.9596±0.0272	3.9290±0.0261	4.0001±0.0201	3.9700±0.0137
Ba-138	3.2129±0.0184	3.1006±0.0238	3.2024±0.0213	3.1809±0.0120
139				3.37±0.67 ^[c]
Ce-140	3.2643±0.0270	3.2575±0.0232	3.3545±0.0137	3.3046±0.0128
141				3.42±0.68 ^[c]
Ce-142	2.9425±0.0188	2.9307±0.0202	3.0141±0.0155	2.9710±0.0103
Nd-143	2.6371±0.0305	2.6302±0.0344	2.6467±0.0176	2.6420±0.0139
Ce + Nd-144	2.1995±0.0250	2.1953±0.0273	2.2443±0.0149	2.2261±0.0116
Nd-145	1.7803±0.0114	1.7846±0.0155	1.7970±0.0119	1.7875±0.0073
Nd-146	1.4633±0.0093	1.4645±0.0130	1.4767±0.0098	1.4686±0.0060
Sm-147	1.2729±0.0389	1.2663±0.0393	1.2454±0.0388	1.2643±0.0383
Nd-148	1.000	1.000	1.000	1.000

Table XXXVI (Continued)

THERMAL FISSION YIELDS RELATIVE TO Nd-148 FOR Pu-239

<u>Nuclide</u>	<u>Standard Capsule CPP-6-2-5</u>	<u>Standard Capsule CPP-6-2-6</u>	<u>Standard Capsule CPP-6-5-5</u>	<u>Average Value^[a]</u>
Sm-149	0.7221±0.0057	0.7219±0.0084	0.7362±0.0038	0.7306±0.0030
Nd-150	0.5630±0.0031	0.5674±0.0049	0.5760±0.0034	0.5686±0.0021
Sm-151	0.5086±0.0258	0.4846±0.0249	0.4583±0.0231	0.4776±0.0240
Sm-152	0.3335±0.0170	0.3331±0.0173	0.3521±0.0178	0.3423±0.0172
153				0.2143±0.0429 ^[c]
Sm-154	0.1561±0.0017	0.1574±0.0017	0.1656±0.0020	0.1591±0.0010
155-160				0.285±0.057

[a] Individual values are weighted as the reciprocals of their variances.

[b] Strontium was not determined on standard capsule CPP-6-5-5.

[c] Estimated yield.

Table XXXVII

NORMALIZED THERMAL FISSION YIELDS FOR Pu-239

<u>Mass No</u>	<u>Fission Yield, %</u>	<u>Mass No</u>	<u>Fission Yield, %</u>
83	0.301±0.006	138	5.40±0.11
84	0.487±0.010	139	5.73±1.08 ^[a]
85	0.574±0.011	140	5.61±0.12
86	0.770±0.016	141	5.81±1.10 ^[a]
87	1.00±0.02	142	5.04±0.11
88	1.35±0.03	143	4.48±0.10
90	2.09±0.04	144	3.78±0.08
91	2.52±0.04	145	3.03±0.06
92	3.02±0.05	146	2.49±0.05
93	3.95±0.07	147	2.15±0.08
94	4.50±0.08	148	1.70±0.03
96	5.12±0.09	149	1.24±0.03
118-130	6.35±1.19 ^[a]	150	0.965±0.020
131	3.60±0.09	151	0.811±0.044
132	5.09±0.12	152	0.581±0.031
133	7.18±0.15	153	0.364±0.073 ^[a]
134	7.20±0.17	154	0.270±0.006
135-136	13.90±0.32	155-160	0.483±0.097 ^[a]
137	6.71±0.14		

[a] Estimated yield.

SUMMARY OF THE FAST FISSION YIELDS OF 45 MASS CHAINS
FOR U-235 AND Pu-239

The fission yields measured in this program for 45 mass chains for the EBR I U-235 and Pu-239 irradiated fuel rods are listed in Table XXXVIII. The error associated with each value is in terms of the standard deviation and was calculated by error propagation techniques. Except for the 125 mass chain, the measurement technique for the atoms of the fission products was isotope dilution mass spectrometry.

Table XXXVIII

SUMMARY OF FAST FISSION CHAIN YIELDS FOR U-235 AND Pu-239

<u>Mass No</u>	<u>Fission Yield, %</u>		<u>Mass No</u>	<u>Fission Yield, %</u>	
	<u>U-235</u>	<u>Pu-239</u>		<u>U-235</u>	<u>Pu-239</u>
83	0.615±0.18	0.366±0.008	111	0.0456±0.0009	0.376±0.007
84	1.07±0.01	0.559±0.013	112	0.0391±0.0008	0.207±0.004
85	1.49±0.03	0.672±0.012	113	0.0342±0.0007	0.133±0.002
86	1.93±0.05	0.882±0.020	114	0.0342±0.0007	0.099±0.002
87	2.66±0.06	1.16±0.02	116	0.0359±0.0007	0.064±0.001
88	3.63±0.08	1.44±0.03	125	0.073±0.009	0.19±0.02
90	5.48±0.16	2.24±0.05	131	3.15±0.08	4.06±0.08
91	[a]	2.58±0.05	132	4.45±0.12	5.42±0.11
92	[a]	3.13±0.06	133	6.69±0.13	6.91±0.12
93	[a]	3.91±0.07	134	7.09±0.19	7.35±0.15
94	[a]	4.39±0.08	135	6.54±0.13	7.54±0.13
95	6.47±0.13	4.78±0.09	136	5.93±0.16	6.92±0.14
96	[a]	5.11±0.10	137	6.20±0.16	6.58±0.11
97	6.13±0.12	5.47±0.10	138	6.60±0.12	4.97±0.09
98	6.04±0.12	5.81±0.11	140	6.21±0.11	5.59±0.10
99	[b]	[b]	141	[d]	[d]
100	6.35±0.13	6.76±0.13	142	5.82±0.11	4.95±0.09
101	5.46±0.11	6.88±0.13	143	5.80±0.12	4.30±0.08
102	4.65±0.10	6.97±0.13	144	5.26±0.11	3.68±0.07
103	[c]	[c]	145	3.85±0.08	3.04±0.05
104	2.35±0.05	6.77±0.12	146	3.00±0.06	2.52±0.04

Table XXXVIII(Continued)

SUMMARY OF FAST FISSION CHAIN YIELDS FOR U-235 AND Pu-239

<u>Mass No</u>	<u>Fission Yield, %</u>		<u>Mass No</u>	<u>Fission Yield, %</u>	
	<u>U-235</u>	<u>Pu-239</u>		<u>U-235</u>	<u>Pu-239</u>
147	[e]	[e]	151	0.438±0.009	0.839±0.015
148	1.75±0.03	1.73±0.03	152	0.309±0.006	0.683±0.012
149	1.09±0.02	1.36±0.02	154	0.098±0.003	0.324±0.006
150	0.832±0.030	1.06±0.02			

- [a] Not measured, end member of chain is a zirconium isotope and the fuel was clad in Zircaloy-2.
- [b] Not measured, end member of chain is Tc-99 which is suspected to have plated out during dissolution.
- [c] Not measured, end member of chain is Rh-103 and rhodium was not determined in the program.
- [d] Not measured, end member of chain is Pr-141 and praseodymium isotopes were not determined in the program.
- [e] Not measured, intermediate member of chain is 2.5-yr Pm-147. The long and intermittent irradiation exposure causes large uncertainties in the decay correction calculation for this chain.

SUMMARY OF THE THERMAL FISSION YIELDS OF 43 MASS CHAINS
FOR U-233, U-235, Pu-239, AND Pu-241

The fission yields measured in this program for 43 mass chains for specially prepared samples irradiated in the Materials Testing Reactor at a position where the cadmium ratio for cobalt was 15 $\pm$ 1.5 are listed in Table XXXIX. The error associated with each value is in terms of the standard deviation and was calculated by error propagation techniques. Except where noted, the measurement technique for the atoms of the fission products was isotope dilution mass spectrometry.

Table XXXIX

SUMMARY OF THERMAL CHAIN YIELDS FOR
U-233, U-235, Pu-239, AND Pu-241

Mass Number	Fission Yield, %			
	U-233	U-235	Pu-239	Pu-241
83[a]	1.03±0.01	0.497±0.009	0.301±0.006	0.200±0.006
84[a]	1.73±0.02	0.930±0.015	0.487±0.010	0.353±0.010
85[a]	2.22±0.01	1.30±0.01	0.574±0.011	0.387±0.010
86[a]	2.90±0.03	1.81±0.03	0.770±0.016	0.601±0.018
87	4.06±0.03	2.54±0.01	1.00±0.02	0.741±0.020
88	5.57±0.04	3.61±0.02	1.35±0.03	0.954±0.025
90	6.96±0.06	5.90±0.04	2.09±0.04	1.53±0.04
91	6.60±0.05	5.90±0.03	2.52±0.04	1.82±0.05
92	6.69±0.05	5.95±0.03	3.02±0.05	2.23±0.06
93	7.09±0.06	6.34±0.04	3.95±0.07	2.90±0.08
94	6.91±0.05	6.41±0.04	4.50±0.08	3.33±0.09
95	6.40±0.05	6.45±0.03	4.86±0.11[b]	3.92±0.09[b]
96	5.84±0.05	6.23±0.04	5.12±0.09	4.33±0.12
97	5.52±0.05	5.86±0.03	5.64±0.16[b]	4.76±0.14[b]
98	5.22±0.04	5.77±0.03	NM[c]	NM[c]
99	5.16±0.07	6.14±0.10	6.59±0.18[b]	6.17±0.16[b]
100	4.46±0.04	6.24±0.03	NM[c]	NM[c]
101	3.27±0.02	5.03±0.04	6.50±0.28[b]	5.94±0.26[b]
102	2.48±0.02	4.19±0.04	6.65±0.29[b]	6.32±0.28[b]
103	NM[d]	NM[d]	NM[d]	NM[d]
104	1.04±0.01	1.82±0.01	6.61±0.29[b]	6.80±0.30[b]
106	0.262±0.002	0.389±0.004	4.55±0.19[b]	6.08±0.25[b]
111	[e]	[e]	[e]	[e]
112	[e]	[e]	[e]	[e]
113	[e]	[e]	[e]	[e]
114	[e]	[e]	[e]	[e]
116	[e]	[e]	[e]	[e]
125	0.116±0.013[b]	0.0291±0.0033[b]	0.116±0.014[b]	0.0416±0.0050[b]
131	3.51±0.04	2.79±0.04	3.60±0.09	3.15±0.09
132	4.88±0.06	4.16±0.06	5.09±0.12	4.64±0.13
133	6.06±0.05	6.73±0.04	7.18±0.15	6.71±0.18
134	6.13±0.07	7.51±0.11	7.20±0.17	8.06±0.22
135	[f]	[f]	[f]	[f]
136	[f]	[f]	[f]	[f]
137	6.93±0.06	6.28±0.03	6.74±0.14	6.60±0.17
138	5.97±0.05	6.80±0.03	5.40±0.11	6.37±0.17
140	6.53±0.05	6.31±0.03	5.61±0.12	5.86±0.16
141	[g]	5.50±0.34[b]	[g]	[g]
142	6.71±0.05	5.88±0.03	5.04±0.11	4.80±0.13

Table XXXIX

SUMMARY OF THERMAL CHAIN YIELDS FOR

U-233, U-235, Pu-239, AND Pu-241

Mass Number	Fission Yield, %			
	U-233	U-235	Pu-239	Pu-241
143	5.85±0.05	5.90±0.03	4.48±0.10	4.48±0.12
144	4.67±0.04	5.42±0.02	3.78±0.08	4.13±0.11
145	3.37±0.03	3.86±0.02	3.03±0.06	3.19±0.08
146	2.53±0.02	2.95±0.01	2.49±0.05	2.68±0.07
147	1.78±0.04	2.12±0.04	2.15±0.08	2.22±0.06
148	1.30±0.01	1.69±0.01	1.70±0.03	1.89±0.05
149	0.773±0.007	1.00±0.01	1.24±0.03	1.43±0.04
150	0.500±0.004	0.638±0.004	0.965±0.020	1.16±0.03
151	0.365±0.011	0.408±0.012	0.811±0.044	[h]
152	0.186±0.004	0.212±0.006	0.581±0.031	0.725±0.034
154	0.0458±0.0006	0.0561±0.0009	0.270±0.006	0.370±0.010

- [a] Krypton and xenon were quantitatively recovered from the off-gas during dissolution of the samples and the atoms of each isotope were determined by a combination of vapor phase chromatography and gas mass spectrometry.
- [b] The measurement technique for these mass chains was gamma-ray spectrometry with either a Ge(Li) or NaI(Tl) detector-pulse height analyzer system. These measurements are reported in earlier sections of this report.
- [c] Not measured, end members of chains are Mo-98 and Mo-100 and molybdenum plated out during dissolution.
- [d] Not measured due to the short half life of Ru-103.
- [e] Not measured, end member of chain is a cadmium isotope and a mass spectrometer suitable for the measurement was not available earlier in the program when the samples were analyzed.
- [f] Not measured, thermal cross section of Xe-135 is greater than 10^6 barns.
- [g] Not measured, experimental phase of program was terminated before the measurements could be completed.
- [h] Not reported, unexplained disagreement in experimental results of the two irradiated capsules.

RECOMMENDED FISSION PRODUCTS FOR BURNUP MONITORS

The final objective of this program is to recommend fission products as burnup monitors of total fissions and source of fissions.

Total Fissions by Measurement of One Fission Product

An ideal monitor for total fissions (a) has the same fission yield for all fissioning species which does not vary with neutron energy, (b) builds up linearly with fissions, (c) is stable (or very long lived relative to in-pile and out-of-pile decay), (d) does not migrate in fuel, and (e) is determinable with high accuracy and precision.

Unfortunately, an ideal monitor is not available. Fission products which may have equal thermal yields for different fissioning species in general display significant variations in yield with neutron energy and seldom display all the other desirable properties. Thus, compromises are necessary when selecting a fission product as a burnup monitor for total fissions.

In addition to the properties described above, the effect of natural contamination upon the analysis for the fission product must be considered. Possible sources of natural contamination are the fuel itself, reagents and apparatus used in fuel dissolution, reagents and apparatus used in the separation procedure of the analysis, and the filaments used in mass spectrometry.

Pyrometallurgical fuel processing, such as used in the Argonne National Laboratory's EBR II reactor, creates a unique problem. The highly volatile fission products are removed in the process and the less volatile fission products are recycled. This requires either sampling the fuel before the high temperature phase of the process and analyzing for the highly volatile fission products or analyzing for the less volatile fission products followed by computation corrections based on fuel recycle records.

U-235 and Pu-239 Mixed Fission Source

To guide the selection of fission products as monitors of total fissions for a fuel in which the sources of fissions are U-235 and Pu-239, Figures 3 through 6 plot in various ways the fission yield data obtained in this program. Figure 3 plots the U-235 to Pu-239 ratios of the thermal (cadmium ratio for cobalt of 15) fission yields vs mass number and Figure 4 plots the same information for fast (EBR I) data. Figure 5 plots the ratios of the fast to thermal fission yields of U-235 vs mass number and Figure 6 is the similar

plot for Pu-239. Table XL summarizes potentially useful monitors for U-235, Pu-239, and mixed U-235, Pu-239 fuels based on a consideration of these four figures and upon the values of the neutron capture cross sections of the various fission products. The symbol X indicates usefulness. Caution must be exercised if an X does not appear in the "unknown" neutron energy column because the fission yield of the particular fission product varies with neutron energy.

The only fission product which has general applicability as a monitor of total fissions in mixed U-235, Pu-239 fuels is Nd-148. However, its fission yield increases with neutron energy, being 4% higher for a metal U-235 fuel in liquid NaK (EBR I) than a light-water enriched-uranium fuel (MTR), and 2% higher for Pu-239 with comparable reactor conditions.

U-233 and U-235 Mixed Fission Source

A potentially useful breeder material is thorium. The likely initial source of fissions in a reactor containing thorium is U-235 giving rise then to the combined fissioning of U-233 and U-235. Referring to the summary tables of thermal fission yields (Table XXXIX) and fast fission yields (Table XXXVIII), the only two indicated useful monitors of total fissions are Mo-95 and Ce-140. Both these isotopes are centrally located in the peaks of the fission yield curve and should not be very sensitive to neutron energy changes.

Figure 3. Ratio of the Fission Yields of Thermal Fissioned U-235 to Pu-239

Figure 4. Ratio of the Fission Yields of Fast Fissioned U-235 to Pu-239

Figure 5. Ratio of the Fission Yields of Fast Fission U-235 to Thermal Fissioned U-235

Figure 6. Ratio of the Fission Yields of Fast Fissioned Pu-239 to Thermal Fissioned Pu-239

Table XL

USEFUL NUCLEAR FUEL BURNUP MONITORS OF TOTAL FISSION

Fission Product	<u>U-235</u> Neutron Energy			<u>Pu-239</u> Neutron Energy			<u>U-235 + Pu-239</u> Neutron Energy			Comments ^[a]
	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	
Sr-88	X	X	X	X	X					Easily determined by ID ^[b] (4). Natural Sr contamination ^[c] is a serious problem. Migration in fuel can be a problem.
Sr-90	X	X		X	X					As for Sr-88. Corrections required for in-pile and out- of-pile decays which adversely affect reliability.
Zr-92	X			X	X					Determined by ID (5,21) with some difficulty. Large amounts of natural contamination cause decreased reliability. ^[c]
Zr-93	X			X	X	X				As for Zr-92.
Zr-94	X			X	X	X				As for Zr-92.
Zr-96	X			X	X	X				As for Zr-92.
Mo-95	X	X	X	X	X	X				Determination by ID is difficult (this report page 93). MoO ₃ is volatile causing migration in UO ₂ and PuO ₂ fuels. Natural contamination either of Mo or Zr seriously affects reliability. ^[c]

Table XL (Continued)

USEFUL NUCLEAR FUEL BURNUP MONITORS OF TOTAL FISSION

Fission Product	<u>U-235</u> <u>Neutron Energy</u>			<u>Pu-239</u> <u>Neutron Energy</u>			<u>U-235 + Pu-239</u> <u>Neutron Energy</u>			Comments ^[a]
	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	
Mo-97	X	X		X	X	X				As for Mo-95.
Mo-98	X	X								As for Mo-95.
Mo-100	X	X	X							As for Mo-95.
Ru-101, -102, -104	X	X								Easily determined by ID (3).
76 Cs-133	X	X	X	X	X			X		Easily determined by ID (21). Natural contamination seriously affects reliability. ^[c] High volatility causes migration in fuels.
Cs-137	X	X		X	X					Easily determined both by ID (21) and by radiochemistry (1). High volatility causes migration in fuels. Correc- tions required for in-pile and out-of-pile decays which adversely affect reliability.

Table XL (Continued)

USEFUL NUCLEAR FUEL BURNUP MONITORS OF TOTAL FISSION

Fission Product	<u>U-235</u> Neutron Energy			<u>Pu-239</u> Neutron Energy			<u>U-235 + Pu-239</u> Neutron Energy			Comments ^[a]
	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	<u>Thermal</u>	<u>Fast</u>	<u>Unknown</u>	
Ce-140	X	X	X	X	X	X				Easily determined by ID (1, 5).
Ce-142	X	X	X	X	X	X				As for Ce-140.
Nd-148	X	X	(X)?	X	X	X	X	X	X	Easily determined by ID (1, 5). The fission yield values vary 4% between the U-235 thermal and EBR I fast fission yield values.

[a] A parenthesis is the reference to the determination procedure.

[b] ID denotes determination by the technique of isotope dilution mass spectrometry, usually after a chemical separation.

[c] All sources of natural contamination must be considered including the fuel, dissolution reagents and apparatus, chemical separation procedure, and the filaments used in mass spectrometry.

Total Fissions by Measurement of the Ratio of Two Fission Product Isotopes of the Same Element

Early in this program, it was proposed (20) that the ratio of selected pairs of stable fission product isotopes of the same element differing by the mass of one neutron serve as a monitor of total fissions. The basis of the technique is that the lower mass isotope has a significant neutron capture cross section to produce the higher mass isotope which has a low capture cross section. The ratio then reflects the integrated flux and hence the total fissions. Two pairs that were proposed were $\frac{\text{Kr-84}}{\text{Kr-83}}$ and $\frac{\text{Xe-132}}{\text{Xe-131}}$. The experimental

data obtained in this program for these pairs are plotted in Figures 7 and 8 for thermal and fast fissioned U-235 and Pu-239. The fast data assume a constant ratio with fissions in view of the low capture cross sections characteristic of fast neutrons.

These figures show that the technique is useful for a highly thermalized reactor in which the fissions are essentially from one fissioning isotope. The fact that the ratio of two isotopes of the same element can be determined very accurately by mass spectrometry contributes greatly to this technique. For example, the $\frac{\text{Kr-84}}{\text{Kr-83}}$ ratio

is determinable to 0.2% which from Figure 7 indicates that percent fissions is determinable to within 1 percentage point for either a U-235 or a Pu-239 fissioning source.

An advantage of using these krypton and xenon isotope pairs, in addition to dispensing with the need for quantitative sampling, is that the sample can be the off-gas from the fuel dissolution process.

Measurement of Relative Fissions from Multifissioning Sources

An evident technique to measure the relative fissions for a source with more than one fissionable isotope is to measure several fission products with differing fission yields. Highest leverage would be obtained for a duo fissionable source by using the ratio of two fission products which have reversed relative fission yields for the two fissionable isotopes. Maximum changes of this nature occur at the sides of the fission yield maxima where, unfortunately, the fission yield values are sensitive to neutron energy. Thus these regions provide usefulness only for fuels with well defined neutron spectra. Another factor that must be considered is the capture cross sections. The values must be small (or fortuitously equal) for the neutron spectrum of the irradiation. As suggested early in the program (20), the ratios of two isotopes of the same element provide the simplest and most reliable measurement.

U-235 and Pu-239 Mixed Fission Source

From an examination of Figures 3 through 6, single element isotope pairs that have promise for mixed U-235 and Pu-239 fissions are Nd-148, Nd-146, and Ru-101. The three ratio values as a function

Nd-150 Nd-148 Ru-104

of percent fissions for thermal and fast fissioned U-235 and Pu-239 are plotted in Figures 9, 10, and 11. These same three ratio values are plotted in Figures 12, 13, and 14, as a function of fractional fissions of U-235 and Pu-239. All three ratios are useful with sensitivities in decreasing order of Ru-101, Nd-148, and Nd-146.

Ru-104 Nd-150 Nd-148

The last three figures show that the reliability of the technique is markedly sensitive to the neutron energy of the irradiation. The reliability of the technique, indicated by Figure 14 for the Ru-101 ratio, based on a reliability of 0.2% in the mass spectro-
Ru-104

metrically determined ratio is, for example, ± 0.005 at 0.8 fraction fissions of U-235 for a thermal neutron irradiation.

In addition to the two neodymium isotope ratios and the one ruthenium isotope ratio, zirconium isotopes appear promising. However, no data are presented in this report because fission yield data were not obtained for zirconium isotopes in the program for fast fissioned U-235. The reason, as explained on page 65, is that the U-235 (EBR I) fuel was clad in Zircaloy which was dissolved. The fission yield data of the zirconium isotopes for thermal and fast fissioned Pu-239 agree closely (Table XXXVIII and XXXIX) as do the data for Mo-95. If this also is the case for U-235, ratios of zirconium isotopes should be useful.

U-233 and Pu-239 Mixed Fission Source

For mixed U-233 and U-235 fuels, the present data do not indicate any isotope pairs useful for the measurement of relative fissions. Based on the thermal fission yield data for U-233 and U-235 (Table XXXIX), the ratios Mo-95 and Ce-140 have promise. Confirmation

Mo-100 Ce-142

requires fast U-233 fission yield data.

Figure 7. Ratio of Kr-84 to Kr-83 for Thermal and Fast Fissioned U-235 and Pu-239

Figure 8. Ratio of Xe-132 to Xe-131 for Thermal and Fast Fissioned U-235 and Pu-239

Figure 9. Ratio of Nd-148 to Nd-150 for Thermal and Fast Fissioned U-235 and Pu-239

Nd-146/Nd-148 RATIO

Figure 10. Ratio of Nd-146 to Nd-148 for Thermal and Fast Fissioned U-235 and Pu-239.

Figure 11. Ratio of Ru-101 to Ru-104 for Thermal and Fast Fissioned U-235 and Pu-239

Figure 12. Fractional Fissions of U-235 and Pu-239 as Function of Nd-148 to Nd-150 Ratio in Duo Fissioning Thermal and Fast Fuels

Figure 13. Fractional Fissions of U-235 and Pu-239 as Function of Nd-146 to Nd-148 Ratio in Duo Fissioning Thermal and Fast Fuels

Figure 14. Fractional Fissions of U-235 and Pu-239 as Function of Ru-101 to Ru-104 Ratio in Duo Fissioning Thermal and Fast Fuels

FUTURE STUDIES

In the completed program, extensive fission yield data reliable to 1 to 2% were obtained for U-233, U-235, Pu-239, and Pu-241 at a highly thermalized (cadmium ratio for cobalt of 15) neutron spectrum and for U-235 and Pu-239 at a fast (EBR I reactor of metal fuel in liquid NaK) neutron spectrum. These neutron spectra conditions represent the extremes anticipated for operational reactors. The yield values obtained for these two conditions are significantly different to warrant future programs at the intermediate neutron energy levels that represent the present power reactors and the next generation of fast oxide breeder reactors. A large fraction of the neutrons in the present power reactors is in the resonance region. The expected mean energy of the fast oxide breeder reactor is about 100 keV compared to 500 keV for EBR I. Without these additional fission yield data, determination of fissions with fission product monitors will remain questionable for power reactor and the fast oxide breeder reactors.

Plans are being formulated by the authors' company, Idaho Nuclear Corporation, to measure fission yields of the long-lived and stable fission products for purposes of burnup monitors at the 100-keV mean neutron energy. These plans include (a) irradiations in a specially fabricated shielded assembly being designed for the Advanced Test Reactor operated by Idaho Nuclear Corporation, (b) analysis of specimens now being irradiated by various organizations in the EBR II, and (c) special irradiations in EBR II by Idaho Nuclear Corporation.

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APPENDIX I

SEPARATION AND MASS SPECTROMETRIC PROCEDURES FOR MOLYBDENUM

(R. M. Abernathey, L. W. Buttars, R. E. Foster, Jr., M. E. Kussy,
F. L. Lisman, R. E. McAtee, R. A. Nielsen, G. D. Workman)

PREPARATION OF Mo-94 SPIKE

A 1-mg sample of isotopically enriched Mo-94, obtained from ORNL as the metal, was dissolved in aqua regia and diluted to 1 liter with quartz distilled 4M HCl. Weighed 3-ml aliquots were transferred to 10-ml polypropylene tubes which were stoppered with a polypropylene cork.

CALIBRATION OF Mo-94 SPIKE

The calibration technique was isotope dilution mass spectrometry with natural molybdenum as the primary standard. Approximately 1.5 g of "specpure" molybdenum trioxide was dissolved in conc H₂SO₄, the solution was evaporated to dryness, the residue was dissolved in 10 ml of quartz distilled 10M HCl, and diluted to approximately 1 liter with quartz distilled 1M HCl. Weighed 25-ml aliquots were gravimetrically analyzed as lead molybdate. The determined concentration of this stock solution was 0.9796 mg Mo/g of solution with a relative standard deviation, for the average of four determinations, of 0.18%.

A weighed aliquot of the stock solution was diluted 1000 fold by weight from which eight weighed approximately 2-ml aliquots were individually transferred to eight of the polypropylene tubes that contained the Mo-94 spike solution. These mixtures were analyzed by isotope dilution mass spectrometry. The measured concentration of Mo-94 in the spike solution was 5.390×10^{15} atoms per gram with a relative standard deviation for the mean of 0.32%.

CHEMICAL SEPARATION

Note: All acids were purified by distillation in quartz and diluted to the desired concentration with quartz distilled water. The ammonium hydroxide was prepared by bubbling ammonia into quartz distilled water in a polyethylene apparatus.

1. Add a sample aliquot containing 1 to 5 μg of fission product molybdenum to a 10-ml polypropylene tube containing the Mo-94 .
Use a calibrated pipet.
2. Add 4 drops of 30% H_2O_2 and stir the solution.
A Vortex mixer is satisfactory.
3. Add NH_4OH to a pH of 9.
pH paper is satisfactory. Aluminum, if present, precipitates. If none is present, add aluminum nitrate to give a final molarity of 1M.
4. Centrifuge and decant the supernatant solution containing the molybdenum into a clean 10-ml polypropylene tube.
5. Add 1 ml of 15M HNO_3 and evaporate to dryness under a heat lamp.
During the evaporation, NH_4Cl may precipitate. If it does, add 1 drop each of conc HNO_3 and conc HCl and continue the evaporation.
6. Dissolve the residue in about 1 ml of 0.1M HCl .
7. Place the solution on an anion exchange column and wash with five 1-ml portions of 0.1M HCl .
To a 2-ml polyethylene medicine dropper add Dowex 1 X8 (100-200 mesh) resin in the chloride form to a height of 1 in. Wash the column several times with 0.1M HCl .
8. Elute the molybdenum with 4 ml of 5M HNO_3 into a 10-ml polypropylene tube and add 4 drops of 10M HCl .
9. Evaporate the solution to dryness under a heat lamp.

MASS SPECTROMETRIC ANALYSIS

1. Prepare a canoe-shaped filament from high purity, 1-x30-mil rhenium ribbon, and vacuum bake it for 1 hr at 4 A at a pressure of 1×10^{-6} torr. Assemble the sample filament in the center (ionizing) position. Surface molybdenum impurities are removed.
2. Using a new, freshly cleaned, disposable glass pipet, add a few drops of 1M HCl to the residue from Step 10 (previous section). Transfer the solution to the filament. Evaporate the solution by passing 1.5 A through the filament.
3. Mount the filament holder on the ion source carriage and insert the carriage into the vacuum lock. Allow about 1-min pumping time at each lock stage.
4. Adjust the filament current to 3.0 to 3.5 A and locate the major isotope beam by reference to a mass-vs DVM (accelerating voltage) curve for the magnetic field in use. Adjust the source voltage divider controls for the maximum beam. Make a rapid voltage scan in both directions to verify the mass identification. Use the electron multiplier. Keep the J2 setting low, the J10 setting high, and the J4-J5 setting near center. The source may need to be rotated to get J4-J5 centered with the maximum beam.
5. Increase the filament current to obtain a multiplier output current of 3×10^{-9} A. Scan the spectrum magnetically in both directions, recording the sensitivity settings used to give near full scale peak heights. Obtain 3 charts of 2 1/2 double scans. Filament current is about 3.2 A. If the filament current exceeds 4 A, residual molybdenum impurity in the filament material will interfere.

6. Graphically interpolate by drawing lines on the chart which connect the tops of the successive peaks of each isotope. Draw vertical lines on the chart at each turn-around in the scans, and take peak readings at the intersection of these lines with those connecting the peak tops. Scan the readings for each isotope.

7. Correct for mass discrimination. Obtain correction factors from comparison between multiplier and Faraday cage data obtained from the same molybdenum sample.

If the atom percent of an individual isotope is desired, it is given by:

$$\text{atom \% Mo}^A = \frac{100 \cdot \sum \text{Corr peak heights}^A}{\sum (\sum \text{Corr peak heights})}$$

CALCULATIONS

Using the spiked fission product samples, define:

A_i = atom percent of ith molybdenum isotope.

k_i = relative amount of ith molybdenum isotope in Mo-94 "spike".

S = relative amount of "spike" in sample aliquot.

l_i = relative amount of ith molybdenum isotope in "natural" molybdenum.

N = relative amount of "natural" molybdenum in sample aliquot.

x_i = relative amount of ith molybdenum isotope in fission product molybdenum.

F = relative amount of fission product molybdenum in sample.

Q = atoms of Mo-94 in "spike" added to sample aliquot.

Z_i = atoms of ith fission product in sample aliquot.

V = volume of dissolver solution.

Y = volume of sample aliquot.

Consider the Mo-94 and Mo-96 isotopes:

$$A_4 = k_4 S + l_4 N \quad (1)$$

$$A_6 = k_6 S + l_6 N \quad (2)$$

Solving (1) and (2) for N and S yields:

$$N = \frac{A_6 - k_6 \left[\frac{A_4 - \frac{l_4 A_6}{l_6}}{k_4 - \frac{l_4 k_6}{l_6}} \right]}{l_6} \quad (3)$$

$$S = \frac{A_4 - \frac{l_4 A_6}{l_6}}{k_4 - \frac{l_4 k_6}{l_6}} \quad (4)$$

For an individual fission product molybdenum isotope:

$$A_i = k_i S + l_i N + X_i F \quad (5)$$

Using equations (3), (4), and (5) above:

$$X_i = A_i - \frac{l_i}{l_6} A_6 + \left[\frac{l_i k_6}{l_6} - k_i \right] \left[\frac{A_4 - \frac{l_4 A_6}{l_6}}{k_4 - \frac{l_4 k_6}{l_6}} \right] \quad (6)$$

and

$$\frac{X_i F Q}{k_4 S} = Z_i \quad (7)$$

or

$$Z_i = \frac{1}{k_4} \left[\frac{\left(A_i - \frac{l_i A_6}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_4 - \frac{l_4 A_6}{l_6}} + \frac{l_i k_6}{l_6} - k_i \right] Q \quad (8)$$

Then total atoms of the i th fission product in the dissolver solution (Z_i) is given by:

$$Z_i = \frac{VQ}{Yk_4} \left[\frac{\left(A_i - \frac{l_i A_6}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_4 - \frac{l_4 A_6}{l_6}} + \frac{l_i k_6}{l_6} - k_i \right] \quad (9)$$

ERROR PROPAGATION FOR MOLYBDENUM ISOTOPES

The number of fission product atoms of the i th molybdenum isotope, by equation (9) of the previous section is:

$$Z_i = \frac{VQ}{Yk_4} \left[\frac{\left(A_i - \frac{l_i A_6}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_4 - \frac{l_4 A_6}{l_6}} + \frac{l_i k_6}{l_6} - k_i \right]$$

Reported values are the average of n determinations:

$$\bar{Z}_i = \frac{1}{n} \sum_j Z_{ij}$$

V and usually Y are the same for all samples and:

$$\bar{Z}_i = \frac{V}{nY} \sum_j \left[\frac{Q_j}{k_4} \left(\frac{\left(A_{ij} - \frac{l_i A_{6j}}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} + \frac{l_i k_6}{l_6} - k_i \right) \right]$$

The variance of $\bar{Z}_i$ is then given approximately by:

$$\begin{aligned}
 \sigma_{\bar{Z}_i}^2 = & \left(\frac{\partial \bar{Z}_i}{\partial V} \right)^2 \sigma_V^2 + \left(\frac{\partial \bar{Z}_i}{\partial Y} \right)^2 \sigma_Y^2 + \left(\frac{\partial \bar{Z}_i}{\partial k_4} \right)^2 \sigma_{k_4}^2 + \sum \left(\frac{\partial \bar{Z}_i}{\partial A_{ij}} \right)^2 \sigma_{A_{ij}}^2 \\
 & + \left(\frac{\partial \bar{Z}_i}{\partial l_{ij}} \right)^2 \sigma_{l_{ij}}^2 + \left(\frac{\partial \bar{Z}_i}{\partial l_6} \right)^2 \sigma_{l_6}^2 + \sum_j \left(\frac{\partial \bar{Z}_i}{\partial A_{6j}} \right)^2 \sigma_{A_{6j}}^2 \\
 & + \sum_j \left(\frac{\partial \bar{Z}_i}{\partial A_{4j}} \right)^2 \sigma_{A_{4j}}^2 + \left(\frac{\partial \bar{Z}_i}{\partial l_4} \right)^2 \sigma_{l_4}^2 + \left(\frac{\partial \bar{Z}_i}{\partial k_6} \right)^2 \sigma_{k_6}^2 \\
 & + \left(\frac{\partial \bar{Z}_i}{\partial k_i} \right)^2 \sigma_{k_i}^2 + \sum_j \left(\frac{\partial \bar{Z}_i}{\partial Q_j} \right)^2 \sigma_{Q_j}^2 + 2 \sum_{j=1}^{n-1} \sum_{k>j}^n \left(\frac{\partial \bar{Z}_i}{\partial Q_j} \right) \left(\frac{\partial \bar{Z}_i}{\partial Q_k} \right) \sigma_{Q_j k}^2 \\
 & + \text{(additional covariance terms)}
 \end{aligned}$$

It is assumed that the additional covariance terms are negligible and that

$$\begin{aligned}
 \frac{\sigma_{Q_{ij}}^2}{\sigma_{Q_j} \sigma_{Q_k}} &= 1 \\
 \text{and} \\
 \sigma_{\bar{Z}_i}^2 = & \frac{V^2}{n^2 Y^2} \left\{ \left[\sum_j \frac{Q_j}{k_4} \left[\frac{\left(A_{ij} - \frac{l_4 A_{6j}}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} + \frac{l_4 k_6}{l_6} - k_i \right] \right]^2 \right. \\
 & \left. + \left[\sum_j \frac{Q_j}{k_4} \left[\frac{\left(A_{ij} - \frac{l_4 A_{6j}}{l_6} \right) \left(1 - k_4 + \frac{l_4 k_6}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} \right] \right]^2 \right\} \sigma_{k_4}^2
 \end{aligned}$$

$$\begin{aligned}
& + \sum_j \left(\left[\frac{Q_j}{k_4} \left(\frac{k_4 - \frac{l_4 k_6}{l_6}}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} \right) \right]^2 \sigma_{A_{ij}}^2 \right) \\
& + \left[\sum_j \left(\frac{Q_j}{k_4} \left[\frac{k_6}{l_6} - \frac{\left(\frac{A_{6j}}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} \right] \right)^2 \sigma_{l_i}^2 \right] \\
& + \left[\sum_j \left(\frac{Q_j}{k_4 l_6^2} \left[\left(k_6 - \frac{A_{6j} \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} \right) \left(\frac{A_{ij} - \frac{l_i A_{6j}}{l_6} l_4}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} - l_i \right) \right] \right)^2 \sigma_{l_6}^2 \right] \\
& + \sum_j \left(\left[\frac{Q_j}{k_4 l_6} \left(\frac{k_4 - \frac{l_4 k_6}{l_6}}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} \right) \left(\frac{A_{ij} - \frac{l_i l_4}{l_6}}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} - l_i \right) \right]^2 \sigma_{A_{6j}}^2 \right) \\
& + \sum_j \left(\left[\frac{Q_j}{k_4} \left(\frac{\left(A_{ij} - \frac{l_i A_{6j}}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{\left(A_{4j} - \frac{l_4 A_{6j}}{l_6} \right)^2} \right) \right]^2 \sigma_{A_{4j}}^2 \right) \\
& + \left[\sum_j \left(\frac{Q_j}{k_4} \left[\frac{k_4 - \frac{l_4 k_6}{l_6}}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} \right] \left[\frac{\left(A_{ij} - \frac{l_i A_{6j}}{l_6} \right) \left(\frac{A_{6j}}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} - \frac{k_6}{k_4 - \frac{l_4 k_6}{l_6}} \right] \right)^2 \sigma_{l_4}^2 \right]
\end{aligned}$$

$$\begin{aligned}
& + \left[\sum_j \left(\frac{Q_j}{k_4} \left[\frac{l_i}{l_6} - \frac{A_{ij} - \frac{l_i A_{6j}}{l_6}}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} \right] \right) \right]^2 \sigma_{k_6}^2 \\
& + \sum_j \left[\left(\frac{1}{k_4} \left[\frac{\left(A_{ij} - \frac{l_i A_{6j}}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{\left(A_{4i} - \frac{l_4 A_{6j}}{l_6} \right)} + \frac{l_i k_6}{l_6} - k_i \right] \right)^2 \sigma_{Q_j}^2 \right] \\
& + 2 \left\{ \sum_{j=1}^{n-1} \sum_{k>i}^n K_j K_k \sigma_{Q_j} \sigma_{Q_k} \right\} \\
& + \left[\frac{1}{nY} \sum_j \left(\frac{Q_j}{k_4} \left[\frac{\left(A_{ij} - \frac{l_i A_{6j}}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} + \frac{l_i k_6}{l_6} - k_i \right] \right) \right]^2 \sigma_V^2
\end{aligned}$$

Where:

$$K = \frac{1}{k_4} \left(\frac{\left(A_{ij} - \frac{l_i A_{6j}}{l_6} \right) \left(k_4 - \frac{l_4 k_6}{l_6} \right)}{A_{4j} - \frac{l_4 A_{6j}}{l_6}} + \frac{l_i k_6}{l_6} - k_i \right)$$

APPENDIX II

SEPARATION PROCEDURE FOR ANTIMONY

(R. E. Foster, Jr.)

CHEMICAL SEPARATION

1. Accurately pipet an aliquot of the fission product solution containing 10^4 to 10^7 dpm of Sb-125 into an arsenic distillation flask. The maximum volume of the sample aliquot is 10 ml. The distillation flask is shown in Fig. A-1.
2. Attach the water-cooled condenser and immerse the outlet tip into 5 ml of 3M HCl contained in a 50-ml volumetric flask. Immerse the volumetric flask in an ice bath.
3. Add 25 ml of conc H_2SO_4 to the distillation flask.
4. Connect the reservoir to the distillation flask and the air hose to the reservoir.
5. Add 17 ml of conc HCl and 8 ml of conc HBr to the reservoir and mix.
6. Flow air through the distillation flask at a rate of 2 bubbles/sec.
7. Transfer 2 to 3 ml of the HCl:HBr mixture to the distillation flask.
8. Rapidly heat the distillation flask to between 200° and 220° C to distill antimony bromide. A heating mantle is recommended.
9. Adjust the flow rate of the HCl:HBr mixture into the distillation flask to about 1 ml/min.
10. Continue the distillation until all the acid mixture has been added to the distillation flask.

11. Cool the distillation flask and disconnect the condenser.
12. Wash the condenser and condenser tip with about 10 ml of water and transfer the washings to the 50-ml receiving flask.
13. Dilute the solution in the receiving flask to volume with water, stopper, and shake until homogeneous. Exercise caution when shaking the flask so as not to contaminate the exterior surface.

Figure A-1 Antimony Bromide Distillation Apparatus