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BURNUP DETERMINATION OF NUCLEAR FUELS
PROJECT REPORT
FOR THE QUARTER JANUARY 1-MARCH 31, 1965

Edited By
W. J. Maeck and J. E. Rein

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IN NUCLEAR SCIENCE ABSTRACTS

**PHILLIPS
PETROLEUM
COMPANY**

ATOMIC ENERGY DIVISION

NATIONAL REACTOR TESTING STATION
US ATOMIC ENERGY COMMISSION

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BURNUP DETERMINATION
OF NUCLEAR FUELS

PROJECT REPORT
FOR THE QUARTER JANUARY 1 - MARCH 31, 1965

Edited by

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SUMMARY

In this continuing program to develop methods for the accurate determination of the burnup of nuclear fuels, the main emphasis this period was the measurement of U-235 thermal fission yields for many stable and long-lived fission products. A capsule containing a weighed quantity of highly enriched U-235, irradiated to 11.5 atom percent fission, was dissolved and analyzed for the number of atoms of each uranium isotope and for the number of atoms of various stable and long-lived fission product nuclides. From these data, the capture-to-fission ratio (α) and absolute fission yields were determined. The value for α_0 is 0.1716 ± 0.0019 . The fission yield values are:

<u>Mass Number</u>	<u>Element</u>	<u>Fission Yield</u>	<u>Mass Number</u>	<u>Element</u>	<u>Fission Yield</u>
83	Kr	0.496 ± 0.015	144	Ce	5.40 ± 0.07
84	Kr	0.453 ± 0.023	144	Nd+Ce	5.36 ± 0.06
85	Kr(10.8y)	0.270 ± 0.006	145	Nd	3.87 ± 0.05
86	Kr	1.84 ± 0.05	146	Nd	2.94 ± 0.03
131	Xe	2.90 ± 0.07	147	Sm	2.03 ± 0.05
132	Xe	4.22 ± 0.10	148	Nd	1.68 ± 0.02
134	Xe	7.74 ± 0.19	149	Sm	1.01 ± 0.01
137(30.3y)	Cs	6.22 ± 0.14	150	Nd	0.634 ± 0.007
140	Ce	6.24 ± 0.07	151	Sm	0.398 ± 0.019
142	Ce	5.84 ± 0.07	152	Sm	0.236 ± 0.009
143	Nd	5.88 ± 0.07	154	Sm	0.050 ± 0.003
144	Nd	5.31 ± 0.06			

These values are tentative. Additional capsules will be similarly handled to produce refined data.

The conditions, established for the mass spectrometric analysis of krypton, xenon, and the lanthanides, to obtain the above fission yield data, are described. Also described is the preparation and standardization of the enriched isotope standard solutions that were used for the isotope dilution mass analysis of the lanthanides.

Separation methods were developed for the alkali metals, the alkaline earths, and ruthenium applicable to irradiated uranium and plutonium samples. These methods are designed for subsequent isotope dilution mass analysis to establish absolute fission yields. Also developed was a separation method for individual lanthanides, applicable to irradiated plutonium samples which will contain relatively high amounts of americium and curium.

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BURNUP DETERMINATION OF NUCLEAR FUELS

INTRODUCTION

The objective of this project is the development of methods for the accurate determination of burnup of nuclear fuels. The emphasis is on power reactor fuels having long residence time and high burnup rather than on short term, experimental irradiations. The principal measurement technique under study is chemical analysis for fission products. The use of long-lived, radiochemically-determined fission products is not applicable because the errors of the in-pile decay 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 analyses 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 atoms of the fission products 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 analysis methods for these nuclides. Presently, experiments are in progress to measure the absolute thermal fission yields for U-233, U-235, Pu-239, and Pu-241 to a high degree of accuracy. Later in the program it is planned to determine the fast fission yields for the heavy element isotopes, from Th-232 through Pu-242.

ABSOLUTE U-235 THERMAL FISSION YIELDS OF LONG-LIVED AND STABLE FISSION PRODUCTS

A major goal of the burnup project is the accurate determination of the thermal fission yields of long-lived and stable fission products. A series of capsules containing weighed amounts of the oxides of highly-enriched U-233, U-235, Pu-239, and Pu-241 was prepared and irradiated in the MTR(1). The first of these, a U-235 capsule, was dissolved and the absolute fission yields were measured for many lanthanides, krypton, xenon, and Cs-137. Also measured was α_0 , the thermal capture-to-fission ratio.

Irradiation History of Capsule

Standard burnup capsule CPP-6-4-1, which contained 115.2 mg of highly-enriched U_3O_8 , was irradiated for 3 cycles (1850 Mwd) in the MTR at an estimated unperturbed flux of 6×10^{13} n/cm² sec (accumulated nvt = 2.4×10^{20}). The cadmium ratio for cobalt for this irradiation was 15. The capsule was discharged from the reactor June 8, 1964, allowed to cool in the MTR canal, and was dissolved January 25, 1965.

Dissolution of Capsule, R. E. Foster, S. F. Marsh

The dissolution of the capsule was carried out in a 4-in. lead-shielded facility. A sketch of the dissolution apparatus is shown in Fig. 1. The dissolver flask and reflux condenser were fabricated from high-purity quartz. With borosilicate glassware, impurities would likely be leached into the dissolver solution. An acid mixture of 6M HCl-1.6M HNO_3 was used for the dissolution(1).

The dissolution proceeded smoothly and was complete in 3.5 hr. The fission gases, krypton and xenon, appeared 1.5 hr after the start of the dissolution and their evolution continued for 2 hr. The final solution (about 200 ml) was water clear and contained no residue.

After cooling, the dissolver solution was quantitatively transferred to a 250-ml, class "A" volumetric flask with the aid of 6M HCl-1.6M HNO_3 rinses. The solution was cooled and diluted to volume with quartz-distilled water. A Teflon-coated stirring bar was admitted and the contents were stirred for 1 hr.

Determination of Number of Fissions, R. M. Abernathey, L. W. Buttars, M. E. Kussy, W. J. Maeck, R. A. Nielsen, F. W. Spraktes

The accuracy of the absolute fission yield measurements directly depends on the accuracy of the determined number of fissions in the irradiated U-235 capsule. For this reason, a large effort was placed on the pre- and postirradiation analyses for the atoms of the various uranium isotopes from which the number of fissions was calculated.

Figure 1. CAPSULE DISSOLUTION APPARATUS

The U_3O_8 (obtained from ORNL) that was weighed into the capsule was analyzed both for weight percent uranium content and isotopic distribution. For the former, samples of the U_3O_8 were weighed on a micro balance, dissolved in nitric acid, weight aliquots were mixed with weight aliquots of a standard U-233 solution, and the mixtures were analyzed by isotope dilution mass spectrometry. Isotopic distribution was determined on unspiked samples. The calculated atoms of each uranium isotope (in the capsule charge of 115.2 ± 0.1 mg U_3O_8) are summarized in Table I. Of interest was the finding that the uranium content of the U_3O_8 corresponded to 99.619% of theoretical.

The postirradiation atoms of each uranium isotope was determined in a similar manner with unspiked and U-233 spiked aliquots of the dissolver solution. The aliquots of the dissolver solution were delivered with a calibrated 1000- μ l pipet. The calculated atoms are presented in Table I.

To accurately compute the number of U-235 fissions by the relationship:

$$F_5 = U^0 - U$$

in which:

$$F_5 = \text{number of U-235 fissions}$$

$$U^0 = \text{total preirradiated atoms of uranium}$$

$$U = \text{total postirradiated atoms of uranium}$$

one must account for those atoms of U-236 and U-238 lost by capture and subsequent decay to non-uranium isotopes. This was done with data obtained from a computer program which simulated the history of the irradiation. The cross sections were selected from the latest literature. The buildup and burnout correction calculations are detailed in the Appendix. It is of interest to note that the U-236 and U-238 lost during the irradiation amounted to 0.28% and 0.29% respectively of that measured at the end of the irradiation. Though these losses are insignificant for the calculation of F_5 , the loss of U-236 is important for the calculation of α , the capture-to-fission ratio of U-235. In this case of highly enriched U-235, the loss of U-236 approximated the amount of U-236 originally present.

From the results:

$$U^0 = (2.4888 \pm 0.0026) \times 10^{20}$$

$$U = (2.2049 \pm 0.0018) \times 10^{20}$$

$$\text{The number of fissions} = F_5 = (2.839 \pm 0.032) \times 10^{19}$$

The propagated standard deviation error of 0.032×10^{19} for the computed number of fissions corresponds to a relative error of 1.13%. The atom

Table I. ANALYSIS RESULTS OF IRRADIATED U-235 CAPSULE
AND CALCULATED NUMBERS OF ATOMS

<u>Preirradiation Data</u>				
Initial Loading	115.2±0.1 mg U ₃ O ₈			
mg U-235/mg U ₃ O ₈	0.8361±0.0007			
mg U-235 in capsule	96.319±0.102(0.106%)			
	<u>U-234</u>	<u>U-235</u>	<u>U-236</u>	<u>U-238</u>
Isotopic distribution, wt%	0.4677	99.1633	0.0603	0.3087
std dev _{ind}	0.0025	0.0032	0.0015	0.0006
std dev _{avg} (n=3)	0.0015	0.0019	0.0009	0.0004
Number of atoms	1.1692x10 ¹⁸	2.4680x10 ²⁰	1.4948x10 ¹⁷	7.5883x10 ¹⁷
std dev	0.0040x10 ¹⁸	0.0026x10 ²⁰	0.0224x10 ¹⁷	0.0129x10 ¹⁷
Total atoms of uranium	(2.48877±0.00264)x10 ²⁰ (0.106%)			
<u>Postirradiation Data</u>				
mg U determined in dissolved sample (n=4)	86.053±0.071(0.082%)			
	<u>U-234</u>	<u>U-235</u>	<u>U-236</u>	<u>U-238</u>
Isotopic distribution, wt%	0.5138	96.7776	2.3508	0.3578
std dev _{ind}	0.0056	0.0074	0.0064	0.0039
std dev _{avg} (n=5)	0.0025	0.0033	0.0028	0.0017
Number of atoms	1.1378x10 ¹⁸	2.1339x10 ²⁰	5.1614x10 ¹⁸	7.7891x10 ¹⁷
std dev	0.0055x10 ¹⁸	0.0017x10 ²⁰	0.0062x10 ¹⁸	0.0370x10 ¹⁷
Total atoms of uranium	(2.2047±0.0018)x10 ²⁰ (0.082%)			
Number of atoms, corrected for capture loss of U-236 and U-238 and the capture of U-234 to U-235	1.1713x10 ¹⁸	2.1336x10 ²⁰	5.1758x10 ¹⁸	7.8114x10 ¹⁷
std dev	0.0055x10 ¹⁸	0.0017x10 ²⁰	0.0062x10 ¹⁸	0.037x10 ¹⁷
Total atoms of uranium, corrected for capture loss of U-236 and U-238	(2.20488±0.00181)x10 ²⁰ (0.082%)			

percent U-235 fissioned (defined as atoms of U-235 fissioned per 100 initial atoms of U-235) is:

$$\begin{aligned} \% F_5 &= 100 \left[\frac{(2.839 \pm 0.032) \times 10^{19}}{(2.4680 \pm 0.0026) \times 10^{20}} \right] \\ &= (11.50 \pm 0.13) \end{aligned}$$

in which the value of 0.13 is the propagated standard deviation error and corresponds to a relative error of 1.14%.

Capture-To-Fission Ratio, W. J. Maeck, F. W. Spratkes

The effective capture-to-fission ratio, $\hat{\alpha}$, is readily calculated from the data in Table I. The number of U-235 atoms captured to form U-236 equals the postirradiated number of U-236 atoms plus those lost by capture to U-237 (and β decay to Np-237) less those atoms of U-236 initially present in the loading. As stated in the previous section, the U-236 atoms lost by capture was obtained from a computer program, and is detailed in the Appendix.

Calculation of $\hat{\alpha}$

(1) atoms U_6 present at end of irradiation	=	$(5.1614 \pm 0.0062) \times 10^{18}$
(2) atoms U_6 lost by capture	=	0.0144×10^{18}
(3) (1) + (2)	=	$(5.1758 \pm 0.0062) \times 10^{18}$
(4) atoms U_6^0 , initial atoms loaded into capsule	=	$(0.1495 \pm 0.0022) \times 10^{18}$
(5) atoms U_5^0 captured to U_6 , (3) - (4)	=	$(5.0263 \pm 0.0066) \times 10^{18}$
(6) atoms U_5^0 fissioned (see previous section)	=	$(28.39 \pm 0.32) \times 10^{18}$
(7) $\hat{\alpha}$, (atoms U_5^0 captured/atoms U_5^0 fissioned)	=	$\frac{(5.0263 \pm 0.0066) \times 10^{18}}{(28.39 \pm 0.32) \times 10^{18}}$
	=	0.1770 ± 0.0019

The propagated standard deviation value of 0.0019 corresponds to a relative error of 1.10%.

The capsule was irradiated in the MTR in a location near the beryllium reflector where the cadmium ratio for cobalt is 15. The Wescott "r" value corresponding to $R_{Cd}^{Co} = 15$ is 0.031, computed from the relationship(2):

$$R_{Cd}^{Co} = \frac{g + rs}{r \left(s + \frac{1}{K} \sqrt{\frac{T}{T_0}} \right)}$$

in which:

$$g = 1$$

$$T = 313^{\circ}\text{K}$$

$$^{\text{s}}\text{Co} = 1.795 \text{ (ref. 2)}$$

$$k = 2.2931 \text{ (for 0.040 in. of Cd) (ref. 3).}$$

To convert $\hat{\alpha}$ to α_o , the 2200 m/sec value, the relationship:

$$1 + \alpha_o = (1 + \hat{\alpha}) (g_f + rs_f)/(g_a + rs_a)$$

applies(4). For a neutron temperature of 40°C (313°K), the values are:

$$g_f = 0.0735 \text{ (ref. 5)} \quad s_a = 0.1501 \text{ (ref. 5)}$$

$$s_f = -0.0316 \text{ (ref. 5)} \quad r = 0.031$$

$$g_a = 0.9723 \text{ (ref. 5)} \quad \hat{\alpha} = 0.1770$$

The computed value of α_o is 0.1716 ± 0.0019 . The error (standard deviation) 0.0019 was computed assuming no uncertainty in the r , g , and s factors. This value of α_o agrees closely with the value of 0.1718 recently published by Lounsbury et al(6).

Lanthanide Fission Yields, S. F. Marsh, W. J. Maeck, F. W. Spraktes

The absolute fission yields of neodymium and samarium isotopes were determined on the same sample aliquots. The cerium fission yields were determined on different sample aliquots.

For neodymium and samarium, four 1000- μl aliquots of the irradiated U-235 dissolver solution were spiked with known amounts of Nd-150 and Sm-144. The preparation of the spike solutions has been described previously(1). After pre separation of the lanthanides (see section, Lanthanides, Method Applicable to Trivalent Actinide Samples), the neodymium and samarium were separated by an α -hydroxyisobutyric acid chromatographic cation exchange procedure(1) and were analyzed by isotope dilution mass spectrometry. The analysis results for neodymium and samarium are summarized in Tables II and IV respectively.

Ordinarily, cerium is selectively separated from the trivalent lanthanides prior to the chromatographic separation of the trivalent lanthanides. In this case, however, the neodymium and samarium separations were made before the cerium standard spike solution was available. For this reason, four separate dissolver solution aliquots were analyzed for cerium after the separation of the neodymium and samarium fractions. The procedure was slightly different from that normally used. The cerium was

coprecipitated with nickel hydroxide, dissolved in 8M HNO_3 -0.1M KBrO_3 , and retained on a D2EHP column (see section, Lanthanides, Method Applicable to Trivalent Actinide Samples). Cerium was then eluted with 2M HCl -1% H_2O_2 , separated from accompanying salts on a small cation exchange column, and mass analyzed.

To accurately calculate the fission yields of the lanthanides isotopes, the atoms of each nuclide must be corrected for buildup and burnout due to neutron capture. These corrections were obtained from a computer program which simulated the actual irradiation conditions of the experimental capsule.

For cerium and neodymium, these corrections were 3% or less for each isotope. The determined number of atoms of each isotope in the capsule, the computer-corrected number of atoms, and the calculated fission yields for neodymium and cerium are summarized in Tables III and VII respectively.

The yield of the mass 144 chain was calculated three different ways; from the neodymium analysis, the cerium analysis, and the combination of the two. In the case of the neodymium analysis, the atoms of undecayed Ce-144, calculated from the computer program, were added to the determined atoms of Nd-144 giving a percent thermal fission yield of 5.31 ± 0.06 . The yield of the 144 chain calculated only from the mass spectrometrically determined number of atoms of Ce-144 with the appropriate decay corrections applied was 5.40 ± 0.07 . A statistical "t" test of these two values showed no significant difference. The most accurate fission yield value is considered to be the sum of the determined atoms of Nd-144 and Ce-144. This value is 5.36 ± 0.06 .

The determined number of atoms of each samarium isotope in the capsule, the computer-corrected number of atoms, and the calculated fission yields are summarized in Table V. Due to the large capture cross sections on some of the samarium isotopes, corrections for neutron capture buildup and burnout are much greater than for cerium and neodymium and hence the errors associated with the fission yields are larger.

Most of the Sm-149, due to its very large cross section, is converted to Sm-150. Rather than apply a large correction factor, it is possible to sum the determined atoms of Sm-149 and Sm-150 because Sm-150 is a shielded nuclide and not directly formed in fission. Small corrections were required, however, for the loss of Sm-150 by neutron capture. This correction factor was determined from the computer program.

Cs-137 Fission Yield, M. E. Kussy, W. J. Maeck

The number of atoms of Cs-137 in the capsule dissolver solution was determined by gamma counting after radiochemical separation. An additional measurement of the number of atoms of Cs-137 by isotope dilution mass spectrometry is planned.

Three 1000- μ l aliquots of the dissolved capsule solution were diluted to 50.00-ml volumes. Duplicate 1000- μ l aliquots then were analyzed by a modified ASTM cesium perchlorate procedure(1). Two different gamma counting techniques were used. One was based on using the 3 x 3 in. NaI(Tl) crystal as the standard with the crystal parameters recommended by Heath(7). The other was a comparison method in which the area under the 661-kev photopeak, after correction for Cs-134, was compared to that of a certified standard (RC-137-ASTM, Nuclear Chicago Corp.). This certified standard contained $(1.72 \pm 0.05) \times 10^{14}$ atoms Cs-137/ml on August 8, 1962. The details of the analyses are given in Table VIII. The computation of the fission yield is given in Table IX. The results of the two counting techniques agreed to within 1%.

Krypton and Xenon Fission Yields, R. E. Foster, W. J. Maeck, F. W. Sprakles

These two gases were trapped from the capsule dissolver off-gas on Molecular Sieve 5A at -190°C . The concentration of each gas was determined by temperature-programmed gas chromatography. The separated gas fractions from the chromatographic column were collected on charcoal at -80°C and were transferred to the mass laboratory for isotopic distribution analysis. Details of the apparatus and the procedure for the collection and analysis have been reported(1).

The computed fission yields of the stable krypton and xenon isotopes and of 10.76-yr Kr-85 are given in Table X. These values are the average of three irradiated U-235 capsules; one the present capsule and two earlier capsules. For the two earlier capsules, the yields were computed relative to Cs-137, using 6.22% as the fission yield of Cs-137 (see previous section). For the present capsule, the yields were computed with respect to the determined number of fissions and hence are absolute. The three values were averaged by weighting the reciprocals of the variances. The error associated with each value is the standard deviation of the mean.

Table II. FISSION PRODUCT NEODYMIUM ANALYSIS RESULTS

<u>Total Neodymium</u>							
<u>Sample</u>	<u>μg Nd/ml</u>						
1	4.863						
2	4.831						
3	4.830						
4	4.859						
$\bar{X}$	4.846						
std dev _{avg} (measurement error only)	$\pm 0.009(0.18\%)$						
std dev _{avg} (including spike uncertainty)	$\pm 0.019(0.40\%)$						
Total micrograms neodymium in postirradiated sample							
$= (4.846 \pm 0.019 \text{ μg Nd/ml})(250 \pm 0.06 \text{ ml}) = 1211.50 \pm 4.85(0.40\%)$							
<u>Isotopic Analysis</u>							
atom % ^a							
<u>Sample</u>	<u>Nd-142</u>	<u>Nd-143</u>	<u>Nd-144</u>	<u>Nd-145</u>	<u>Nd-146</u>	<u>Nd-148</u>	<u>Nd-150</u>
1	0.010	32.57	16.03	21.66	16.70	9.46	3.56
2	0.010	32.48	15.98	21.73	16.75	9.52	3.53
3	0.010	32.51	16.01	21.62	16.75	9.50	3.60
4	0.010	32.55	16.02	21.68	16.69	9.47	3.58
$\bar{X}$	0.010	32.58	16.01	21.67	16.72	9.49	3.57
std dev _{avg}	0.000	0.02	0.02	0.03	0.02	0.014	0.015
% std dev _{avg}		0.06	0.06	0.14	0.12	0.15	0.42

a

All isotopic data corrected for natural contamination by that amount of Nd-142 in excess of 0.010%. Uncorrected Nd-142 usually was about 0.3% in raw sample.

Table III. U-235 THERMAL FISSION YIELDS OF THE STABLE NEODYMIUM ISOTOPES

$$\text{Number of Fissions} = (2.839 \pm 0.032) \times 10^{19} \text{ (1.13\%)}$$

	Nd-143	Nd-144	Nd-145	Nd-146	Nd-148	Nd-150
Determined number of atoms	1.6416×10^{18}	8.0670×10^{17}	1.0919×10^{18}	8.4248×10^{17}	4.7819×10^{17}	1.7988×10^{17}
std dev	0.0066×10^{18}	0.0326×10^{17}	0.0046×10^{18}	0.0352×10^{17}	0.0204×10^{17}	0.0104×10^{17}
Atoms after correction for neutron capture	1.6703×10^{18}	1.5068×10^{18a}	1.0986×10^{18}	8.3604×10^{17}	4.7664×10^{17}	1.7988×10^{17}
std dev	0.0067×10^{18}	0.0076×10^{18}	0.0047×10^{18}	0.0349×10^{17}	0.0204×10^{17}	0.0104×10^{17}
std dev, %	0.40	0.50	0.42	0.42	0.43	0.58
Percent fission yield	5.88	5.31	3.87	2.94	1.68	0.634
Std dev	0.07	0.06	0.05	0.03	0.02	0.007
Std dev, %	1.20	1.23	1.21	1.21	1.21	1.27

^a

Includes correction for holdup by Ce-144.

Table IV. FISSION PRODUCT SAMARIUM ANALYSIS RESULTS

<u>Total Samarium</u>	
<u>Sample</u>	<u>µg Sm/ml</u>
1	0.5830
2	0.5837
3	0.5918
4	0.5727
$\bar{X}$	0.5828
std dev _{avg} (measurement error only)	0.0039(0.67%)
std dev _{avg} (including spike uncertainty)	0.0041(0.70%)
Total micrograms samarium in postirradiated sample	
= (0.5828±0.0041 µg Sm/ml)(250±0.06 ml) = 145.7±1.0(0.70%)	

<u>Isotopic Analysis</u>							
weight % ^a							
<u>Sample</u>	<u>Sm-147</u>	<u>Sm-148</u>	<u>Sm-149</u>	<u>Sm-150</u>	<u>Sm-151</u>	<u>Sm-152</u>	<u>Sm-154</u>
1	17.37	1.00	4.89	43.70	7.00	22.86	3.17
2	17.22	1.00	4.77	43.60	7.05	23.36	3.00
3	17.32	1.00	4.85	44.03	7.08	23.24	2.49
4	17.35	1.00	4.84	43.91	7.05	23.09	2.76
$\bar{X}_4$	17.31	1.00	4.84	43.81	7.05	23.14	2.86
std dev _{avg}	0.03	0.00	0.02	0.10	0.02	0.11	0.15
% std dev _{avg}	0.02	---	0.52	0.22	0.23	0.46	5.17

a

All isotopic data corrected for natural contamination by that amount of Sm-148 in excess of 1.00%. Uncorrected Sm-148 ranged from 1.40 to 1.50% in raw sample.

Table V. U-235 THERMAL FISSION YIELDS OF STABLE SAMARIUM ISOTOPES

$$\text{No. of fissions} = (2.839 \pm 0.032) \times 10^{19} \text{ (1.13\%)}$$

	Sm-147	Sm-148	Sm-149	Sm-150	Sm-151	Sm-152	Sm-154
Determined number of atoms	1.0339×10^{17}	0.0593×10^{17}	0.2852×10^{17}	2.5642×10^{17}	0.4100×10^{17}	1.3365×10^{17}	0.1630×10^{17}
std dev	0.0072×10^{17}	0.0004×10^{17}	0.0025×10^{17}	0.0187×10^{17}	0.0030×10^{17}	0.0119×10^{17}	0.0073×10^{17}
Atoms after correction for neutron capture	$5.7716 \times 10^{17}{}^a$	None	2.8597×10^{17}	None	1.1327×10^{17}	0.6709×10^{17}	0.1413×10^{17}
std dev	0.1212×10^{17}		0.0189		0.0543	0.0245	0.0073×10^{17}
Percent fission yield	2.03		1.01		0.398	0.236	0.050
std dev	0.05		0.01		0.019	0.009	0.003
std dev, %	2.4		1.3		5.1	3.7	5.3

a

Includes correction for holdup by Pm-147.

Table VI. FISSION PRODUCT CERIUM ANALYSIS RESULTS

<u>Total Cerium</u>	
<u>Sample</u>	<u>μg Ce/ml</u>
1	3.781
2	3.826
3	3.790
4	3.829
$\bar{X}$	3.807
std dev _{avg} (measurement error only)	0.012(0.32%)
std dev _{avg} (includes spike uncertainty)	0.014(0.36%)
Total micrograms cerium in postirradiation sample	
= (3.807±0.014 μg Ce/ml)(250±0.06 ml) = 951.6±3.5(0.37%)	

<u>Isotopic Analysis</u>			
Weight %			
<u>Sample</u>	<u>Ce-140</u>	<u>Ce-142</u>	<u>Ce-144</u>
1	43.449	41.058	15.492
2	43.346	41.215	15.439
3	43.032	41.193	15.736
4	43.250	41.189	15.472
5	43.083	41.088	15.802
6	43.443	41.029	15.494
$\bar{X}_6$	43.267	41.129	15.573
std dev _{avg}	0.044	0.033	0.025
% std dev _{avg}	0.10	0.08	0.16

Table VII. U-235 THERMAL FISSION YIELDS OF LONG-LIVED AND STABLE CERIUM ISOTOPES

$$\text{No. of fissions} = (2.839 \pm 0.032) \times 10^{19} \text{ (1.13\%)}$$

	Ce-140	Ce-142	Ce-144
Determined number of atoms	1.7723×10^{18}	1.6609×10^{18}	6.2017×10^{17}
std dev	0.0067×10^{18}	0.0063×10^{18}	0.0236×10^{17}
Atoms after correction for neutron capture and decay	1.7703×10^{18}	1.6566×10^{18}	1.5338×10^{18a}
std dev	0.0067×10^{18}	0.0063×10^{18}	0.0075×10^{18}
Percent fission yield	6.24	5.84	5.40
std dev	0.07	0.07	0.07
std dev, %	1.2	1.2	1.2

a

Includes in-pile and out-pile decay corrections.

Table VIII. SUMMARY OF Cs-137 ANALYSIS DATA

Constants for Cs-137 counting

Source = 3/4-in. disk

Distance = 10 cm

T = detector efficiency = 0.02012

P = photopeak efficiency = 0.536

A = absorber (1.18 mg/cm² Be = 0.934)

q = γ/dis = 0.857 $\pm$ 0.010

$t_{1/2}$ = 30.3 $\pm$ 0.5 yr (1.7%)

Cooling time = 240 days

} Estimated std dev = 1.5%

Results based on 3 x 3 in. NaI(Tl) crystal as the standard

cpm under 661-keV photopeak (corrected for chemical yield)

$\bar{X}_6$ = 52,225

std dev_{ind} = 647 (1.24%)

std dev_{avg} = $647/\sqrt{6}$ = 264 (0.51%) measurement error only

$\bar{X}_6$ = 52,225

std dev_{avg} = 339 (0.65%) includes uncertainty in carrier value

atoms Cs-137 capsule = (1.767 $\pm$ 0.045) $\times 10^{18}$ (2.56%)

Results from comparison method

ratio, $\frac{\text{sample activity}}{\text{standard activity}}$ = 8.535 $\pm$ 0.069(0.81%)

atoms Cs-137 per capsule = (1.763 $\pm$ 0.055) $\times 10^{18}$ (3.1%) includes 3% uncertainty in certified value of the standard

Table IX. U-235 THERMAL FISSION YIELD OF Cs-137

Atoms Cs-137 using crystal as standard

$$= (1.767 \pm 0.045) \times 10^{18} \quad (2.56\%)$$

Atoms Cs-137 by comparison method

$$= (1.763 \pm 0.055) \times 10^{18} \quad (3.1\%)$$

Weighted value, atoms Cs-137^a

$$= 1.765 \pm 0.035 \quad (1.98\%)$$

Number of fissions

$$= (2.839 \pm 0.032) \times 10^{19} \quad (1.13\%)$$

Fission yield Cs-137

$$= 6.22 \pm 0.14 \quad (2.25\%)$$

^a

Values were averaged by weighting the reciprocals of the variances.

Table X. U-235 THERMAL FISSION YIELDS OF
STABLE KRYPTON AND XENON ISOTOPES

<u>Isotope</u>	<u>Percent Fission Yield</u>
Kr-83	0.496 $\pm$ 0.015
Kr-84	0.953 $\pm$ 0.023
Kr-85 (10.76 yr)	0.270 $\pm$ 0.006
Kr-86	1.84 $\pm$ 0.05
Xe-131	2.90 $\pm$ 0.07
Xe-132	4.22 $\pm$ 0.10
Xe-134	7.74 $\pm$ 0.19

SUMMARY OF FISSION YIELDS

Summarized in Table XI from the preceding sections are the U-235 thermal fission yield values for the stable and long-lived isotopes of krypton, xenon, Cs-137, neodymium, cerium, and samarium. For comparison purposes, literature values for U-235 thermal fission are also given. Additional capsules containing U-235 have been irradiated over a range of burnup levels and also will be analyzed for fission yield data to produce refined data.

Table XI. SUMMARY OF THE U-235 THERMAL FISSION YIELDS OF THE STABLE AND LONG-LIVED ISOTOPES OF KRYPTON, XENON, CERIUM, NEODYMIUM, SAMARIUM, AND Cesium-137

Mass No.	Element	Katcoff(8)	Farrar et al(9,10)	This Work
83	Kr	0.544	0.548	0.496±0.015
84	Kr	1.00	1.01	0.953±0.023
85(10.76y)	Kr	0.293		0.270±0.006
86	Kr	2.02	2.04	1.84±0.05
131	Xe	2.93	2.93	2.90±0.07
132	Xe	4.38	4.38	4.22±0.10
134	Xe	8.06	8.06	7.74±0.19
137(30.3y)	Cs	6.15	6.17	6.22±0.14
140	Ce	6.44	6.25	6.24±0.07
142	Ce	6.01	5.80	5.84±0.07
143	Nd	6.03	5.71	5.88±0.07
144	Nd	5.62	5.30	5.31±0.06
144	Ce			5.40±0.07
144	Nd+Ce			5.36±0.06
145	Nd	3.98	3.80	3.87±0.05
146	Nd	3.07	2.89	2.94±0.03
147	Sm	2.36	2.16	2.03±0.05
148	Nd	1.71	1.61	1.68±0.02
149	Sm	1.13	1.02	1.01±0.01
150	Nd	0.67	0.628	0.634±0.007
151	Sm	0.44	0.399	0.398±0.019
152	Sm	0.281	0.260	0.236±0.009
154	Sm	0.077	0.072	0.050±0.003

MASS SPECTROMETRY METHODS FOR STABLE AND LONG-LIVED FISSION PRODUCTS

R. M. Abernathey

Lanthanides, R. E. McAtee, G. D. Workman

The instrument used was a Nuclide Corp. TDD-1 with a triple filament source. The filaments were 1-x 30-mil rhenium, prebaked for 2 hr at $< 10^{-6}$ torr with a current of 3 amp.

The separated neodymium, samarium, and cerium fractions containing about 3 μg of the lanthanide in 0.1M HNO_3 were evaporated on the two sample filaments. The filaments were outgassed in the spectrometer when the pressure reached 2×10^{-6} torr. The scanning was started when the pressure decreased to 8×10^{-7} torr. The pressure usually decreased to 2×10^{-7} torr during the analysis. With currents of 1.3 amp through the sample filaments and 4.0 amp through the ionization filament, a lanthanide ion current of 3×10^{-12} amp was easily obtained.

For neodymium, interference was observed at the start of the scan period from natural barium oxide, cerium, and samarium. After a few minutes, the barium oxide and samarium beams disappeared. The correction made for cerium was small. For samarium, interference was initially observed from natural barium oxide, barium fluoride, and neodymium which disappeared after a few minutes. For cerium, little interference was observed and the ion beams were very stable.

Krypton and Xenon, L. L. Dickerson

The separated krypton and xenon fractions were analyzed with a Consolidated Electrodynamics Corp. Model 21-103 mass spectrometer. Voltage scanning with a magnet field strength of 3200 gauss was used. To establish voltage discrimination, samples of natural krypton and xenon were analyzed with both magnetic and voltage scanning. None was observed and voltage scanning was therefore used on the samples. The ionization current was adjusted to give near full-scale deflection on one of the galvanometers for the major isotope in each sample. For example, 50 μa was required for a 0.03 std cc sample of krypton.

Standardization of Isotope Spike Solutions, M. E. Kussy, R. E. McAtee, G. D. Workman, S. S. Yamamura

The primary measurement technique to determine the atoms of the various fission product nuclides for fission yield determinations is isotope dilution mass spectrometry. Because the aliquots of the dissolved capsule solutions that are analyzed contain microgram amounts of fission products, standard solutions of the enriched isotope spikes at the 1 $\mu\text{g/g}$ level are required. Chemical methods of analysis are not

sufficiently reliable at this level; hence, these solutions are standardized by isotope dilution mass spectrometry using normal element standards as the spike.

The adopted procedure is to prepare a 1 mg/g solution of the natural element, starting, if possible, with a known weight of a stoichiometric form and then standardizing it gravimetrically or volumetrically. This solution is then accurately diluted 1000-fold on a weight basis. The enriched isotope spike solution is prepared from ORNL-separated material using the most highly enriched material available. The spike solution is prepared at the 1 $\mu\text{g/g}$ level.

To standardize the spike solutions on a weight basis, weighed amounts of the natural and spike solutions are well mixed, then analyzed by isotope-dilution mass spectrometry. For storage, weight aliquots (1 to 3 g) of the standardized spike solution are sealed in corrosion-resistant glass ampoules with the intent being that the entire contents of an ampoule will serve for a single analysis. Ampoules of the 1 mg/g and 1 $\mu\text{g/g}$ natural element standards, of the 1 $\mu\text{g/g}$ enriched element standardized solution, and of weight mixtures of the 1 $\mu\text{g/g}$ solutions are also prepared for future calibration use.

Three enriched isotope spike solutions, Ce-136, Nd-150, and Sm-144 have been standardized. The enrichment values, and the concentration values with an associated standard deviation are:

<u>Isotope</u>	<u>Enrichment</u>	<u>Concentration $\pm$ std dev.</u>
Ce-136	29.97%	0.2898 $\pm$ 0.0009 μg Ce-136/g soln. (0.31%)
Nd-150	95.65%	0.9000 $\pm$ 0.0032 μg Nd-150/g soln. (0.36%)
Sm-144	91.51%	0.8203 $\pm$ 0.0017 μg Sm-144/g soln. (0.21%)

DEVELOPMENT OF SEPARATION METHODS FOR STABLE
AND LONG-LIVED FISSION PRODUCT NUCLIDES

Ruthenium, R. E. Foster

The stable isotopes of ruthenium comprise about 11% of the atoms formed in U-235 thermal fission. Measuring their fission yields is therefore necessary to define the mass-yield curve. Also, because the ruthenium isotopes are in the mass region where the yields for Pu-239 are about ten-fold greater than for U-235, they may be useful as monitors to differentiate the source of fission.

A method was developed for the carrier-free separation of stable fission product ruthenium for subsequent isotope dilution mass analysis. The method is applicable to aqua regia dissolver solutions of irradiated uranium-aluminum capsules. Following a prefuming with sulfuric acid to remove nitrate and chloride, the ruthenium is quantitatively distilled from a sodium bismuthate-sulfuric acid medium and the RuO_4 distillate is absorbed in sulfurous acid at ice temperature. The quantitative distillation guarantees isotopic exchange of the spike and sample.

Prefuming with sulfuric acid removes chloride and nitrate. These must be removed to effect rapid quantitative distillation of the ruthenium(11). The prefuming also dissociates strong chloro and nitrate complexes of ruthenium. It is important that the chloride to nitrate ratio be high to prevent partial distillation of the ruthenium in the prefuming. For samples high in nitrate and low in chloride, the addition of hydrochloric acid is required. Aqua regia samples were fumed up to 30 min with no detectable loss of ruthenium.

After prefuming, the sample is cooled and diluted with water to 2.2M H_2SO_4 and a slurry of 0.5g of sodium bismuthate is added. Complete distillation of the ruthenium is obtained in 10 min. This was confirmed by following the sulfuric acid-bismuthate distillation with repeated perchloric acid distillations. No additional ruthenium was observed.

The distillate is collected in chilled sulfurous acid (SO_2 -saturated quartz distilled water) in preference to the usually used sodium hydroxide. No salts are introduced which interfere in the subsequent surface ionization mass spectrometric analysis. Cooling the sulfurous acid in an ice bath is probably not required but is a precautionary measure. No losses were observed in the collection of the ruthenium in the sulfurous acid at room temperature; however, greater than 90% was lost at 80°C. This indicates that the reduction of RuO_4 is not instantaneous. No loss of ruthenium by plateout in the sulfurous acid was observed. Of particular interest is that the ruthenium-sulfurous acid mixture can be evaporated to near dryness with only 1% loss of ruthenium. This is important because the sample must be concentrated before loading onto the surface ionization sample filaments of the mass spectrometer.

The conditions selected for the distillation are; 2 to 5 μg of total ruthenium (fission product plus added Ru-96 spike), a distillation medium of 80 ml of 2.2M H_2SO_4 containing 0.5g of sodium bismuthate, and a distillate absorbant of 5 ml of one part SO_2 -saturated water and one part water.

Based on a series of 12 distilled aliquots of a dissolver solution and gamma counting of Ru-106 in the distillate, this procedure gives quantitative recovery with a relative standard deviation for a single determination of 1.3%. A sketch of the distillation apparatus is shown in Figure 2.

Lanthanides, Method Applicable to Trivalent Actinide Samples, S. F. Marsh

A three-column sequential procedure has been developed for the separation of individual lanthanides which is applicable to dissolver samples of both irradiated uranium and plutonium. With the first column, based on the Talspeak process(12), the lanthanide group is separated from the actinides and gross fission products. Cerium is separated from the other lanthanides on the second column. On the third column, the lanthanides are chromatographically separated with α -hydroxyisobutyric acid at controlled pH as the elutriant. All three columns are operated at room temperature.

The basis of the first (Talspeak) column is the retention of the lanthanides on the column as strong monoacid organophosphate complexes while the actinides and gross fission products are eluted with an aminocarboxylic acid complexing agent. The organophosphate is 0.3M di-(2-ethylhexyl)phosphoric acid (D2EHP)-0.06M tri-2 ethylhexylphosphate (T2EHP) in diisopropylbenzene and the complexing elutriant is glycine-1M HNO_3 -0.05M pentasodium diethylenetriaminepentaacetate (Na_5DTPA) at the unadjusted pH value of 1.78. The column material is prepared by mixing two parts of acid-washed DMCS chromosorb W, 100-120 mesh (obtained from Wilkens Instrument and Research, Walnut Creek, Calif.), one part of the organophosphate extractant, with sufficient acetone to saturate the chromosorb. The acetone is slowly evaporated over a steam bath, followed by air drying until the resulting column material is visually dry. This is packed into a 5- by 70-mm glass column with tamping to give a flow rate of 0.3 ml/min.

The separation of cerium on the second column is based on its oxidation to the (IV) valence state with bromate and the selective retention of cerium(IV) on a column of 20 parts Chromosorb, 4 parts D2EHP, and 1 part T2EHP.

The α -hydroxyisobutyric acid chromatographic column has been described previously(1).

The steps in the overall procedure are:

- (1) Add enriched isotope spikes to an aliquot of the sample containing about 5 μg of each of the lanthanides and mix well.

Figure 2. RUTHENIUM DISTILLATION APPARATUS

- (2) Coprecipitate the lanthanides with nickel hydroxide (100 μ g nickel carrier) from 6M NaOH.
- (3) Wash the precipitate with water.
- (4) Dissolve the precipitate with the complexing elutriant mixture (1M glycine-1M HNO_3 - 0.05M Na_5DTPA) and transfer to the first column.
- (5) Remove the actinides and gross fission products with six 1-ml volumes of the elutriant. Wash with 2 ml of water to remove the elutriant.
- (6) Elute the lanthanides with 3 ml of freshly prepared 8M HNO_3 - 0.1M KBrO_3 and transfer them to the second column. (The second column has the same dimensions as the first and is prepared in a similar manner with acetone. It is pre-equilibrated with 8M HNO_3 - 0.1M KBrO_3).
- (7) Elute the trivalent lanthanides with an additional 2 ml of the 8M HNO_3 - 0.1M KBrO_3 mixture. (Tetravalent cerium, which is retained on the column, may be eluted separately with 2M HCl -1% H_2O_2).
- (8) Coprecipitate the lanthanides with nickel hydroxide from 1M NaOH.
- (9) Dissolve the precipitate with 0.1M HCl and transfer to the Dowex 50-X4, 325-400 mesh column for the chromatographic separation.

The recovery of the lanthanides in this procedure is near quantitative and the decontamination of americium and curium is greater than 99.3%. The decontamination from gross fission products (Figure 3) and the subsequent separation of the individual lanthanides are excellent (Figure 4).

The isolation of the lanthanide group from all other fission products, actinides, and cerium requires less than 1 hr and easily adapts to remote operation. The overall time to separate the neodymium fraction from gross fission products is less than 4 hr.

Alkali Metals and Alkaline Earths

Previous studies(1) have shown that the two fission product alkali metals, rubidium and cesium, can be mass analyzed with one filament loading and the two alkaline earths, strontium and barium, with another single loading. Thus, only group separations from the heavy elements and other fission products are required. A single-column, ion exchange procedure has been developed to accomplish this.

Figure 3. DISTRIBUTION OF FISSION PRODUCTS IN TALSPEAK SEPARATION METHOD

Figure 4. SEPARATION OF INDIVIDUAL LANTHANIDES BY THE RECOMMENDED PROCEDURE

Olsen and Sobel(13) reported that monovalent cations remain on a tetramethylammonium form Dowex 50-X8 resin column while multivalent cations are eluted with 0.068M ethylenediaminetetraacetic acid (EDTA) adjusted to pH 8.0 with tetramethylammonium hydroxide. Decreasing the pH to 6.0 was found to also retain divalent cations on the resin. Sequential elution, first of the alkaline earths at pH 8.0 and then the alkali metals with dilute hydrochloric acid, proved feasible. The separated fractions, however, contained undesirably high amounts of organic salts for surface ionization mass spectrometry.

Korkisch(14) showed that alkaline earths formed anionic chloride complexes in a 90% ethanol-10% 6M HCl medium but not in a 90% methanol - 6M HCl medium. Uranium(VI) (and presumably PuO_2^{++}) formed anionic complexes with both media and the alkali metal ions were not complexed with either.

Based on the above, the following procedure was developed for the sequential group separation.

- (1) Add the spikes to an aliquot of the dissolved fuel solution, and mix well.
- (2) Transfer the mixture, in a pH 6.0, 0.068M EDTA medium to a Dowex 50-X8 column in the pH 6.0 tetramethylammonium form.
- (3) Thoroughly wash the column with the same medium.
- (4) Rinse with water.
- (5) Elute the residual uranium and plutonium with 90% methanol-10% 6M HCl.
- (6) Elute the alkaline earths (strontium and barium) with 90% ethanol-10% 6M HCl.
- (7) Elute the alkali metals (rubidium and cesium) with 3M HCl.

Final evaluation of the procedure is delayed until higher purity filament material for mass spectrometry is obtained.

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APPENDIX
CORRECTIONS FOR THE BURNOUT AND BUILDUP
OF URANIUM ISOTOPES

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APPENDIX

Corrections for the Burnout and Buildup of Uranium Isotopes

The relationship $F_5 = U^0 - U$, which is used to calculate the absolute number of fissions, assumes no loss of uranium other than by fission. Because some U-236 and U-238 is lost by neutron capture to non-uranium isotopes, corrections must be applied. Similarly, to accurately calculate the capture-to-fission ratio, corrections are required for the loss of U-236 by neutron capture and the production of U-236 by double neutron capture on U-234.

To obtain these corrections, the irradiation history of the analyzed capsule was simulated with a computer program. The buildup and burnout schematic is shown in Figure A-1. The effective cross sections for this program were obtained from the thermal neutron cross sections and the resonance integral tabulation of Stoughton and Halperin(15). For this irradiation, the cadmium ratio for cobalt was 15, corresponding to a $\phi_{\text{res}} = \frac{1}{28.6} \phi_{\text{th}}$.

The computer program used the experimentally determined preirradiation uranium concentrations, the estimated flux, and the reactor residence time of the analyzed capsule. The postirradiation uranium isotopic composition calculated from the computer output agreed to within 2% of the experimentally determined postirradiation uranium isotopic composition. It is therefore believed that the corrections applied to the experimental data are valid to within 10%. The correction factors and the corrected number of atoms are given in Table A-I.

BUILDUP AND BURNOUT OF URANIUM ISOTOPES

$$\text{FLUX} = 6 \times 10^{13} \text{ n/cm}^2 \text{ sec}$$

$$nvt = 2.4 \times 10^{20}$$

$$R_{Cd}^{Co} = 15; \quad \phi_{res} = \frac{1}{28} \phi_{th}$$

Figure A-1. BUILDUP AND BURNOUT OF URANIUM ISOTOPES

Table A-I. CORRECTIONS FOR THE BURNOUT AND BUILDUP OF URANIUM ISOTOPES

Determined Postirradiation Atoms of Uranium

$$\text{U-235} = (2.1339 \pm 0.0017) \times 10^{20}$$

$$\text{U-234} = (1.1378 \pm 0.005) \times 10^{18}$$

$$\text{U-236} = (5.1614 \pm 0.0062) \times 10^{18}$$

$$\text{U-238} = (7.7891 \pm 0.037) \times 10^{17}$$

U-236 Corrections

$$\text{Measured postirradiation atoms U-236 } (5.1614 \pm 0.0062) \times 10^{18}$$

Original U-236 lost by various processes	{	U-236 → U-237 → Np-237	0.000696
		U-236 → U-237 → f	0.000005
		U-236 → U-237 → Np-237 → C	0.000011
		U-236 → U-237 → U-238	<u>0.000003</u>
			$(5.162115 \pm 0.0062) \times 10^{18}$
U-236 formed from U-235 & lost	{	U-235 → U-236 → U-237 → Np-237	0.013950
		U-235 → U-236 → U-237 → f	0.000095
		U-235 → U-236 → U-237 → Np-237 → C	0.000008
		U-235 → U-236 → U-237 → U-238	<u>0.000047</u>
			$(5.176215 \pm 0.0062) \times 10^{18}$

$$\text{less U-234} \rightarrow \text{U-235} \rightarrow \text{U-236} \quad \underline{0.000400}$$

$$\text{corrected atoms U-236} \quad (5.1758 \pm 0.0062) \times 10^{18}$$

U-238 Corrections

$$\text{Measured postirradiation atoms U-238} \quad (7.7891 \pm 0.037) \times 10^{17}$$

$$\text{plus U-238 lost in irradiation} \quad \underline{0.0228}$$

$$(7.8119 \pm 0.037) \times 10^{17}$$

$$\text{less U-236} \rightarrow \text{U-237} \rightarrow \text{U-238} \quad 0.00003$$

$$\text{and U-235} \rightarrow \text{U-236} \rightarrow \text{U-237} \rightarrow \text{U-238} \quad \underline{0.00047}$$

$$\text{Corrected atoms U-238} \quad (7.8114 \pm 0.037) \times 10^{17}$$

U-235 Corrections

Measured postirradiation atoms U-235	$(2.1339 \pm 0.0017) \times 10^{20}$
less U-234 $\rightarrow$ U-235	<u>0.0003</u>
Corrected atoms U-235	$(2.1336 \pm 0.0017) \times 10^{20}$

U-234 Corrections

Measured postirradiation atoms U-234	$(1.1378 \pm 0.0055) \times 10^{18}$
plus U-234 lost during irradiation	<u>0.0335</u>
Corrected atoms U-234	$(1.1713 \pm 0.0055) \times 10^{18}$

Summary of Corrected Postirradiation Uranium Atoms

U-235	$(2.1336 \pm 0.0017) \times 10^{20}$
U-234	$(1.1713 \pm 0.0055) \times 10^{18}$
U-236	$(5.1758 \pm 0.0062) \times 10^{18}$
U-238	$(7.8114 \pm 0.037) \times 10^{17}$

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