80
1/24/66

IDO-14667

December 1965

MASTER

BURNUP DETERMINATION OF NUCLEAR FUELS
PROJECT REPORT
FOR THE QUARTER JULY 1-SEPTEMBER 30, 1965

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

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

**PHILLIPS
PETROLEUM
COMPANY**

ATOMIC ENERGY DIVISION

**NATIONAL REACTOR TESTING STATION
US ATOMIC ENERGY COMMISSION**

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

PRINTED IN USA. PRICE \$2.00. AVAILABLE FROM THE CLEARINGHOUSE FOR FEDERAL
SCIENTIFIC AND TECHNICAL INFORMATION, NATIONAL BUREAU OF STANDARDS
U. S. DEPARTMENT OF COMMERCE, SPRINGFIELD, VIRGINIA

LEGAL NOTICE

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

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

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

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

IDO-14667
AEC Research and Development Report

Chemistry
TID-4500 (46th Ed.)
Issued: December 1965

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

BURNUP DETERMINATION
OF NUCLEAR FUELS

PROJECT REPORT
FOR THE QUARTER JULY 1 - SEPTEMBER 30, 1965

Edited by

William J. Maeck and James E. Rein

PHILLIPS
PETROLEUM
COMPANY

Atomic Energy Division

Contract AT(10-1)-205

Idaho Operations Office

U. S. ATOMIC ENERGY COMMISSION

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

BURNUP DETERMINATION
OF NUCLEAR FUELS

PROJECT REPORT
FOR THE QUARTER JULY 1 - SEPTEMBER 30, 1965

Edited by

William J. Maeck and James E. Rein

Contributing Personnel

Analytical Development Section

R. E. Foster
M. E. Kussy
F. L. Lisman
S. F. Marsh

Mass Spectrometry Group

R. M. Abernathey
L. W. Buttars
L. L. Dickerson
R. E. McAtee
R. A. Nielsen
A. H. Toepfer
G. D. Workman

Operations Analysis (Statistics)

F. W. Spraktes

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

SUMMARY

In this continuing program to develop methods for the accurate determination of the burnup of nuclear fuels, the main emphasis this period was additional measurements of U-235 and U-233 thermal fission yields from the second irradiated capsules of U-235 and U-233. Two capsules, one containing a weighed quantity of highly enriched U-233, and the other highly enriched U-235, both irradiated to ~ 12 atom percent fission, were dissolved and analyzed for the number of atoms of each uranium isotope and for the number of atoms of the various stable and long-lived fission product nuclides. The absolute fission yield values determined on the second U-233 capsule are given below. Also reported is the weighted average of the absolute U-235 fission yields determined on the first and second U-235 capsules.

Mass Number	Element	Fission Yield, %	
		U-233	U-235
83	Kr	0.985±0.024	0.497±0.009
84	Kr	1.63±0.04	0.920±0.016
85(10.76y)	Kr	0.484±0.012	0.265±0.004
86	Kr	2.74±0.07	1.81±0.03
131	Xe	3.44±0.09	2.84±0.05
132	Xe	4.71±0.12	4.24±0.08
134	Xe	5.93±0.15	7.62±0.15
137(30.3y)	Cs	6.60±0.17	6.21±0.10
140	Ce	6.30±0.13	6.30±0.06
142	Ce	6.35±0.14	5.87±0.05
143	Nd	5.57±0.12	5.89±0.06
144	Nd	4.48±0.09	5.36±0.05
144	Ce	4.46±0.13	5.40±0.05
144	Nd+Ce	4.48±0.09	5.40±0.05
145	Nd	3.22±0.06	3.88±0.04
146	Nd	2.41±0.04	2.95±0.02
147	Sm	1.78±0.07	2.13±0.05
148	Nd	1.25±0.02	1.68±0.02
149	Sm	0.710±0.013	1.00±0.01
150	Nd	0.471±0.009	0.630±0.006
151	Sm	0.363±0.019	0.431±0.016
152	Sm	0.166±0.006	0.226±0.005
154	Sm	0.024±0.005	0.056±0.001

In addition the values for the thermal capture-to-fission ratio (α) of U-233 and U-235 were determined. These values are:

U-233; 0.0942±0.0016

U-235; 0.1715±0.0024

All of the above values are tentative and as additional capsules are analyzed refined data will be reported.

Previous project reports are:

<u>Period</u>	<u>Report</u>
Annual - 1962	TID-17385
Annual - 1963	IDO-14649
Annual - 1964	IDO-14656
Quarter, January-March, 1965	IDO-14660
Quarter, April-June, 1965	IDO-14663

CONTENTS

	<u>Page</u>
SUMMARY	v
INTRODUCTION	1
ABSOLUTE U-235 THERMAL FISSION YIELDS OF LONG-LIVED AND STABLE FISSION PRODUCTS	2
<u>Irradiation History of U-235 Capsule</u>	2
<u>Dissolution of U-235 Capsule</u>	2
<u>Determination of Number of U-235 Fissions</u>	2
<u>Capture-to-Fission Ratio for U-235</u>	5
<u>Lanthanide Fission Yields of U-235</u>	6
<u>Cs-137 Fission Yield of U-235</u>	7
<u>Krypton and Xenon Fission Yields for U-235</u>	7
ABSOLUTE U-233 THERMAL FISSION YIELDS OF LONG-LIVED AND STABLE FISSION PRODUCTS	14
<u>Irradiation History of U-233 Capsule</u>	14
<u>Dissolution of U-233 Capsule</u>	14
<u>Determination of Number of U-233 Fissions</u>	14
<u>Capture-to-Fission Ratio for U-233</u>	17
<u>Lanthanide Fission Yields of U-233</u>	19
<u>Cs-137 Fission Yield of U-233</u>	19
<u>Krypton and Xenon Fission Yields for U-233</u>	24
SUMMARY OF DETERMINED U-233 AND U-235 THERMAL FISSION YIELDS AND CAPTURE-TO-FISSION RATIO.	26
CORRECTIONS FOR THE BUILDUP AND BURNOUT OF STABLE FISSION PRODUCTS.	30
REFERENCES	33
APPENDIX - COMPUTER PROGRAM SCHEMATICS FOR THE BUILDUP AND BURNOUT OF FISSION PRODUCTS AND SAMPLE CALCULATION OF THE CORRECTION FACTOR FOR Na-143	34

FIGURES

	<u>Page</u>
A-1. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT SAMARIUM	36
A-2. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT CERIUM AND NEODYMIUM	37
A-3. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT XENON.	38
A-4. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT KRYPTON	39

TABLES

I. ANALYSIS RESULTS OF IRRADIATED U-235 CAPSULE AND CALCULATED NUMBERS OF ATOMS	4
II. DETERMINED ATOMS OF EACH NEODYMIUM ISOTOPE AND CALCULATED NEODYMIUM THERMAL FISSION YIELDS FOR U-235	9
III. DETERMINED ATOMS OF EACH CERIUM ISOTOPE AND CALCULATED CERIUM THERMAL FISSION YIELDS FOR U-235	10
IV. DETERMINED ATOMS OF EACH SAMARIUM ISOTOPE AND CALCULATED SAMARIUM THERMAL FISSION YIELDS FOR U-235	11
V. DETERMINED ATOMS OF Cs-137 AND CALCULATED U-235 FISSION YIELD.	12
VI. U-235 THERMAL FISSION YIELDS OF STABLE KRYPTON AND XENON ISOTOPES	13
VII. ANALYSIS RESULTS OF IRRADIATED U-233 CAPSULE AND CALCULATED NUMBERS OF ATOMS	16
VIII. U-233 THERMAL FISSION YIELDS OF THE STABLE NEODYMIUM ISOTOPES	20
IX. U-233 THERMAL FISSION YIELDS OF LONG-LIVED AND STABLE CERIUM ISOTOPES	21
X. U-233 THERMAL FISSION YIELDS OF STABLE SAMARIUM ISOTOPES	22
XI. U-233 THERMAL FISSION YIELD OF Cs-137	23

TABLES (Continued)

	<u>Page</u>
XII. U-233 THERMAL FISSION YIELDS OF STABLE KRYPTON AND XENON ISOTOPES	25
XIII. SUMMARY OF U-235 FISSION YIELDS	27
XIV. SUMMARY OF U-233 FISSION YIELDS	28
XV. CAPTURE-TO-FISSION RATIO FOR U-233 AND U-235	29
XVI. CORRECTION FACTORS AND ESTIMATED ERRORS FOR THE BUILDUP AND BURNOUT OF FISSION PRODUCTS FOR U-235 AND U-233 CAPSULES	32

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.

Initial values for many U-235 and U-233 thermal fission yields have been reported in previous quarterly reports IDO-14660 and IDO-14663.

During this last quarter, a second set of values for the thermal fission yields of U-233 and U-235 was obtained and is given in this report.

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). During this quarter a second U-235 capsule was dissolved and the absolute fission yields were measured for many lanthanides, krypton, xenon, and Cs-137. Also measured was α_o^5 , the thermal capture-to-fission ratio for U-235.

Irradiation History of U-235 Capsule

Standard burnup capsule CPP-6-6-2, which contained 81.94 mg of highly-enriched U_3O_8 , was irradiated for three cycles (1908 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 September 15, 1964, allowed to cool in the MTR canal, and was dissolved June 30, 1965.

Dissolution of U-235 Capsule, R. E. Foster, S. F. Marsh

The dissolution of the capsule was carried out in a 4-in. lead-shielded facility. The dissolution apparatus has been previously described(1,2). The dissolver flask and reflux condenser were fabricated from high-purity quartz and an acid mixture of 6M HCl-1.6M HNO₃, prepared from quartz-distilled reagents, 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 HNO₃ 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 U-235 Fissions, R. M. Abernathey, L. W. Buttars, M. E. Kussy, F. L. Lisman, W. J. Maeck, R. A. Nielsen, F. W. Sprakles

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 post-irradiation analyses for the atoms of the various uranium isotopes from which the number of fissions was calculated.

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 81.94 ± 0.10 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 99.62% of theoretical.

The postirradiation atoms of each uranium isotope were 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 nonuranium isotopes. This was done with data obtained from a computer program which simulated the history of the irradiation. Discussion and details of the buildup and burnout calculations have been reported previously(2).

From the results:

$$U^0 = (1.77020 \pm 0.00261) \times 10^{20}$$

$$U = (1.55010 \pm 0.00155) \times 10^{20}$$

$$\text{The number of fissions} = F_5 = (2.2010 \pm 0.0305) \times 10^{19}$$

The propagated standard deviation error of 0.0305×10^{19} for the computed number of fissions corresponds to a relative error of 1.38%. The atom percent U-235 fissioned (defined as atoms of U-235 fissioned per 100 initial atoms of U-235) is:

$$\begin{aligned} \% F_5 &= 100 \frac{(2.2010 \pm 0.0305) \times 10^{19}}{(1.7554 \pm 0.0026) \times 10^{20}} \\ &= 12.54 \pm 0.18 \end{aligned}$$

in which the value of 0.18 is the propagated standard deviation error and corresponds to a relative error of 1.40%.

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

<u>Preirradiation Data</u>				
Initial Loading	81.94±0.10 mg U ₃ O ₈			
mg U-235/mg U ₃ O ₈	0.8361±0.0007			
mg U-235 in capsule	68.51±0.10 (0.148%)			
	<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	8.3164x10 ¹⁷	1.7554x10 ²⁰	1.0632x10 ¹⁷	5.3973x10 ¹⁷
Std dev _{avg}	0.0292x10 ¹⁷	0.0026x10 ²⁰	0.0161x10 ¹⁷	0.0106x10 ¹⁷
Total atoms of uranium	(1.7702±0.0026)x10 ²⁰ (0.148%)			
<u>Postirradiation Data</u>				
mg U determined in				
dissolved sample (n=4)	58.407±0.061(0.104%)			
	<u>U-234</u>	<u>U-235</u>	<u>U-236</u>	<u>U-238</u>
Isotopic distribution, wt%	0.517	96.542	2.586	0.357
Std dev _{ind}	0.0026	0.0028	0.001	0.0014
Std dev _{avg} (n=4)	0.001	0.0014	0.001	0.001
Number of atoms	8.0439x10 ¹⁷	1.4965x10 ²⁰	3.9910x10 ¹⁸	5.4401x10 ¹⁷
Std dev _{avg}	0.0175x10 ¹⁷	0.0015x10 ²⁰	0.0042x10 ¹⁸	0.0121x10 ¹⁷
Total atoms of uranium	(1.54989±0.00155)x10 ²⁰ (0.104%)			
Number of atoms, corrected for capture loss of U-236 and U-238 and capture of U-234 to U-235				
	8.2736x10 ¹⁷	1.4963x10 ²⁰	4.0008x10 ¹⁸	5.4554x10 ¹⁷
Total atoms of uranium corrected for capture loss of U-236 and U-238	(1.55010±0.00155)x10 ²⁰ (0.104%)			

Capture-to-Fission Ratio for U-235, F. L. Lisman, W. J. Maeck, F. W. Sprakles

The effective capture-to-fission ratio, $\hat{\alpha}$, is calculated from the data in Table I. The number of U-235 atoms captured to form U-236 equals the postirradiation 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 previously, the number of U-236 atoms lost by capture was calculated from a computer program.

Calculation of α for U-235

(1) atoms U_6 present at end of irradiation	=	$(3.9910 \pm 0.0042) \times 10^{18}$
(2) atoms U_6 lost by capture	=	0.0098×10^{18}
(3) (1) + (2)	=	$(4.0008 \pm 0.0042) \times 10^{18}$
(4) atoms U_6^0 , initial atoms loaded into capsule	=	$(0.1063 \pm 0.0016) \times 10^{18}$
(5) atoms U_5^0 captured to U_6 , (3)-(4)	=	$(3.8945 \pm 0.0045) \times 10^{18}$
(6) atoms U_5^0 fissioned (see previous section)	=	$(22.010 \pm 0.305) \times 10^{18}$
(7) $\hat{\alpha}$, (atoms U_5^0 captured/atoms U_5^0 fissioned)	=	$\frac{(3.8945 \pm 0.0045) \times 10^{18}}{(22.010 \pm 0.305) \times 10^{18}}$
	=	0.1769 ± 0.0033

The propagated standard deviation value of 0.0033 corresponds to a relative error of 1.38%.

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 (3):

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

in which:

$$g = 1$$

$$T = 313^\circ K$$

$$s_{Co} = 1.795 \text{ (ref. 4)}$$

$$k = 2.2931 \text{ (for 0.040 in. of Cd) (ref. 4)}$$

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:

$$\begin{array}{ll} g_f = 0.0735 \text{ (ref.5)} & s_a = 0.1501 \text{ (ref. 5)} \\ s_f = -0.0316 \text{ (ref. 5)} & r = 0.031 \\ g_a = 0.9723 \text{ (ref. 5)} & \hat{\alpha} = 0.1769 \end{array}$$

Therefore:

$$\alpha_o \text{ for U-235} = 0.1715 \pm 0.0024 \text{ (1.38\%)}$$

The error (standard deviation) 0.0024 was computed assuming no uncertainty in the r, g, and s factors. This value of α_o agrees closely with our previously determined value of 0.1716 (2) and the value of 0.1718 recently published by Okazaki, Lounsbury, Durham, and Crocker(6).

Lanthanide Fission Yields of U-235, F. L. Lisman, S. F. Marsh, W. J. Maack, F. W. Sprakes

The absolute fission yields of neodymium, samarium, and cerium were determined. Four 1000- μ l aliquots of the irradiated U-235 dissolver solution were spiked with known amounts of Nd-150, Sm-144, and Ce-136. The preparation and standardization of the spike solutions has been described previously(1,2).

A three-column sequential procedure is used for the separation of the individual lanthanides. The first column separates the lanthanides from the actinides and most fission products. Cerium is separated from the other lanthanides on a second column. On the third column, the lanthanides are chromatographically separated with α -hydroxyisobutyric acid at controlled pH as the elutriant. Details of the procedure have been previously reported(2).

The analysis results and calculated U-235 thermal fission yields for neodymium, cerium, and samarium are summarized in Tables II, III, and IV respectively.

To accurately calculate the fission yields of the lanthanide 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. A detailed discussion of the determination of these correction factors is given in a later section (Corrections for the Burnout and Buildup of Stable Fission Products).

The yield of the mass 144 chain was calculated three different ways; from the neodymium analysis, from the cerium analysis, and from 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 5.46 ± 0.09 . 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 5.48 ± 0.09 . 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 5.48 ± 0.08 .

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 IV. 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 is 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 of U-235, M. E. Kussy, W. J. Maeck

The number of atoms of Cs-137 in the capsule dissolver solution was determined by gamma counting after radiochemistry 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 from each dilution then were analyzed by a modified ASTM cesium perchlorate procedure(1). Two different gamma counting techniques were used. One was based on using the 3x3 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.). The details of the analyses and computations for the two methods have been previously discussed(2).

The determined number of Cs-137 atoms by both methods and the U-235 thermal fission yield are given in Table V.

Krypton and Xenon Fission Yields for U-235, 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(1,2).

The computed fission yields of the stable krypton and xenon isotopes and of 10.76-yr Kr-85 are given in Table VI. The error associated with each value is the standard deviation.

Table II. DETERMINED ATOMS OF EACH NEODYMIUM ISOTOPE AND
CALCULATED NEODYMIUM THERMAL FISSION YIELDS FOR U-235

$$\text{Number of Fissions} = (2.2010 \pm 0.0305) \times 10^{19} \quad (1.38\%)$$

	Nd-143	Nd-144	Nd-145	Nd-146	Nd-148	Nd-150
Determined number of atoms	1.2702×10^{18}	7.4388×10^{17}	8.5125×10^{17}	6.5694×10^{17}	3.7319×10^{17}	1.4225×10^{17}
Std dev _{avg} (n=4)	0.0086×10^{18}	0.0401×10^{17}	0.0410×10^{17}	0.0279×10^{17}	0.0201×10^{17}	0.0075×10^{17}
Atoms after correction for neutron capture	1.3009×10^{18}	1.2015×10^{18}	8.5626×10^{17}	6.5202×10^{17}	3.7224×10^{17}	1.4227×10^{17}
Std dev, % (includes estimated uncertainties in correction factors)	0.67	0.75	0.50	0.43	0.54	0.53
Percent fission yield	5.91	5.46	3.89	2.96	1.69	0.646
Std dev	0.09 ₀	0.08 ₆	0.05 ₈	0.04 ₃	0.02 ₅	0.009 ₆
Std dev, %	1.53	1.58	1.49	1.45	1.48	1.48

Table III. DETERMINED ATOMS OF EACH CERIUM ISOTOPE AND
CALCULATED CERIUM THERMAL FISSION YIELDS FOR U-235

Number of Fissions = $(2.2010 \pm 0.0305) \times 10^{19}$ (1.38%)

	Ce-140	Ce-142	Ce-144
Determined number of atoms	1.4125×10^{18}	1.3063×10^{18}	4.5563×10^{17}
Std dev _{avg} (n=1)	0.0020	0.0026	0.0063
Atoms after correction for neutron capture	1.4110×10^{18}	1.3034×10^{18}	1.2051×10^{18} ^a
Std dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.36	0.36	1.00
Percent fission yield	6.41	5.92	5.48
Std dev	0.09 ₁	0.08 ₄	0.09 ₄
Std dev, %	1.42	1.42	1.71

^a Includes in-pile and out-pile decay corrections.

Table IV. DETERMINED ATOMS OF EACH SAMARIUM ISOTOPE AND
CALCULATED SAMARIUM THERMAL FISSION YIELDS FOR U-235

Number of Fissions = $(2.2010 \pm 0.0305) \times 10^{19}$ (1.38%)

	Sm-147	Sm-148	Sm-149	Sm-150	Sm-151	Sm-152	Sm-154
Determined number of atoms	1.1106×10^{17}	4.9125×10^{15}	2.4556×10^{16}	1.9881×10^{17}	2.9812×10^{16}	1.0737×10^{17}	1.2356×10^{16}
Std dev (n=4) avg	0.0012×10^{17}	0.0125×10^{15}	0.0093×10^{16}	0.0062×10^{17}	0.0119×10^{16}	0.0046×10^{17}	0.0076×10^{16}
Atoms after correction for neutron capture and radioactive decay	4.7636×10^{17}	None	2.2337×10^{17}	None	9.6034×10^{16}	5.0272×10^{16}	1.2360×10^{16}
Std dev % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	3.00		0.30		5.01	3.03	0.61
Percent fission yield	2.16		1.01		0.436	0.228	0.056
Std dev	0.07_1		0.01_4		0.023	0.007_6	0.001
Std dev, %	3.30		1.41		5.20	3.33	1.51

Table V. DETERMINED ATOMS OF Cs-137 AND
CALCULATED U-235 FISSION YIELD

Atoms Cs-137 using crystal as standard

$$= (1.3569 \pm 0.0341) \times 10^{18} \quad (2.5\%)$$

Atoms Cs-137 by comparison method

$$= (1.3772 \pm 0.0432) \times 10^{18} \quad (3.1\%)$$

Weighted value, atoms Cs-137^a

$$= (1.3648 \pm 0.0266) \times 10^{18} \quad (1.9\%)$$

Number of fissions

$$= (2.2010 \pm 0.0304) \times 10^{19} \quad (1.4\%)$$

Fission Yield Cs-137

$$= 6.20 \pm 0.15 \quad (2.4\%)$$

^a

Values were averaged by weighting the reciprocals of the variances.

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

<u>Isotope</u>	<u>Percent Fission Yield</u>
Kr-83	0.499 $\pm$ 0.010
Kr-84	0.936 $\pm$ 0.019
Kr-85 (10.76 yr)	0.266 $\pm$ 0.005
Kr-86	1.82 $\pm$ 0.04
Xe-131	2.87 $\pm$ 0.06
Xe-132	4.28 $\pm$ 0.09
Xe-134	7.73 $\pm$ 0.20

ABSOLUTE U-233 THERMAL FISSION YIELDS OF LONG-LIVED AND STABLE FISSION PRODUCTS

In addition to the dissolution and analysis of the second U-235 standard burnup capsule discussed in the previous section, a second U-233 capsule was dissolved. The absolute fission yields were measured for many lanthanides, krypton, xenon, and Cs-137. Also measured was α_o^3 , the thermal capture-to-fission ratio for U-233.

Irradiation History of U-233 Capsule

Standard burnup capsule CPP-6-4-3, which contained 126.0 mg of U-233 enriched U_3O_8 , was irradiated for three 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 June 1, 1965.

Dissolution of U-233 Capsule, R. E. Foster, S. F. Marsh

The dissolution of the capsule and treatment of the dissolver solution was the same as described in the previous section for the U-235 capsule.

Determination of Number of U-233 Fissions, R. M. Abernathy, L. W. Buttars, M. E. Kussy, F. L. Lisman, W. J. Maeck, R. A. Nielsen, F. W. Sprakes

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 and dissolved in nitric acid, weight aliquots were mixed with weight aliquots of a standard U-235 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 126.0 ± 0.2 mg U_3O_8) are summarized in Table VII. Of interest was the finding that the uranium content of the U_3O_8 corresponded to 99.525% of theoretical.

The postirradiation atoms of each uranium isotope were determined in a similar manner with unspiked and U-235 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 VII.

To accurately compute the number of U-233 fissions by the relationship:

$$F_3 = U^0 - U$$

in which:

$$F_3 = \text{number of U-233 fissions}$$

U^0 = total preirradiated atoms of uranium

U = total postirradiated atoms of uranium

one must account for those atoms of U-235 and U-238 lost by fission and capture. 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 loss of U-235 and U-238 during the irradiation amounted to less than 0.002% of the total uranium measured at the end of the irradiation.

From the results:

$$U^0 = (2.73920 \pm 0.00600) \times 10^{20}$$

$$U = (2.40016 \pm 0.00140) \times 10^{20}$$

$$\text{The number of fissions} = F_3 = (3.3904 \pm 0.0616) \times 10^{19}$$

The propagated standard deviation error of 0.0616×10^{19} for the computed number of fissions corresponds to a relative error of 1.82%. The atom percent U-233 fissioned (defined as atoms of U-233 fissioned per 100 initial atoms of U-233) is:

$$\begin{aligned} \% F_3 &= 100 \frac{(3.3904 \pm 0.0616) \times 10^{19}}{(2.7367 \pm 0.0060) \times 10^{20}} \\ &= (12.39 \pm 0.23) \end{aligned}$$

in which the value of 0.23 is the propagated standard deviation error and corresponds to a relative error of 1.83%.

Table VII. ANALYSIS RESULTS OF IRRADIATED U-233
CAPSULE AND CALCULATED NUMBERS OF ATOMS

<u>Preirradiation Data</u>				
Initial Loading	126.0±0.2 mg U ₃ O ₈			
mg U-233/mg U ₃ O ₈	0.84046±0.00079			
mg U-233 in capsule	105.898±0.232(0.22%).			
	<u>U-233</u>	<u>U-234</u>	<u>U-235</u>	<u>U-238</u>
Isotopic distribution, wt%	99.908	0.026	0.0032	0.062
Std dev _{ind}	0.004	0.002	0.0014	0.003
Std dev _{avg} (n=4)	0.002	0.001	0.0007	0.0015
Number of atoms	2.7367x10 ²⁰	7.0945x10 ¹⁶	8.7654x10 ¹⁵	1.6627x10 ¹⁷
Std dev _{avg}	0.0060x10 ²⁰	0.2738x10 ¹⁶	1.9108x10 ¹⁵	0.0411x10 ¹⁷
Total atoms of uranium	(2.7392±0.0060)x10 ²⁰ (0.22%).			
<u>Postirradiation Data</u>				
mg U determined in dissolved sample (n=4)				
91.533±0.055(0.060%)				
	<u>U-233</u>	<u>U-234</u>	<u>U-235</u>	<u>U-236</u> <u>U-238</u>
Isotopic distribution, wt%	98.551	1.346	0.036	0.0009 0.067
Std dev _{ind}	0.002 ₈	0.002	0.002	0.0004 0.001
Std dev _{avg} (n=4)	0.001 ₄	0.001	0.001	0.0002 0.001
Number of atoms	2.3655x10 ²⁰	3.2161x10 ¹⁸	8.6400x10 ¹⁶	2.0641x10 ¹⁵ 1.5840x10 ¹⁷
Std dev _{avg}	0.0014x10 ²⁰	0.0029x10 ¹⁸	0.2418x10 ¹⁶	0.5036x10 ¹⁵ 0.0237x10 ¹⁷
Total atoms of uranium	(2.40012±0.00140)x10 ²⁰ (0.06%)			
Total atoms of uranium corrected for loss by processes other than U-233 fission	(2.40016±0.00140)x10 ²⁰ (0.06%)			

Capture-to-Fission Ratio for U-233, F. L. Lisman, W. J. Maeck, F. W. Spraktes

The effective capture-to-fission ratio $\hat{\alpha}_3$ is calculated from the data in Table VII by the method of Okazaki, Lounsbury, and Durham(8). In the relationship

$$\frac{\alpha}{1+\alpha} = \frac{\text{Number of captures}}{\text{Number of absorptions}},$$

the number of absorptions is equal to the loss of U-233 during the irradiation and the number of captures is equal to

$$(N_4 + N_5 + N_6 + \beta + f_5^0) - (N_4^0 + N_5^0) \quad \text{where}$$

N = number of atoms in postirradiated sample

N^0 = preirradiated number of atoms (the subscripts 4, 5, 6 refer to U-234, U-235, and U-236 respectively)

β = number of U-235 fissions resulting from the U-235 formed by capture on U-233 and U-234 (equal to $N_6/\hat{\alpha}_5$ where $\hat{\alpha}_5 = 0.1770$)

f_5^0 = U-235 fissions from U-235 initially present

CALCULATION OF $\hat{\alpha}$

(1) Atoms U_3^O	=	$(2.7367 \pm 0.0060) \times 10^{20}$	
(2) Atoms U_3	=	$(2.3655 \pm 0.0014) \times 10^{20}$	
(3) $1 + \hat{\alpha}$ (Total absorptions), (1)-(2)	=	$(0.3712 \pm 0.0062) \times 10^{20}$	(1.67%)
(4) Atoms N_4	=	$(3.2161 \pm 0.0029) \times 10^{18}$	
(5) Atoms N_5	=	$(0.0864 \pm 0.0024) \times 10^{18}$	
(6) Atoms N_6 less N_6 formed from N_5^O	=	$(0.0019 \pm 0.0005) \times 10^{18}$	
(7) $\beta = (6)/0.1770$	=	$(0.0107 \pm 0.0029) \times 10^{18}$	
(8) r_5^O	=	$(0.0011) \times 10^{18}$	
(9) Sum (4) thru (8)	=	$(3.3164 \pm 0.0048) \times 10^{18}$	(0.14%)
(10) N_4^O	=	$(0.0709 \pm 0.0027) \times 10^{18}$	
(11) N_5^O	=	$(0.0088 \pm 0.0019) \times 10^{18}$	
(12) Sum (10) and (11)	=	$(0.0797 \pm 0.0033) \times 10^{18}$	(4.14%)
(13) Number of captures, (9)-(12)	=	$(3.2367 \pm 0.0058) \times 10^{18}$	(0.18%)
(14) $\frac{\hat{\alpha}}{1 + \hat{\alpha}} = \frac{(13)}{(3)}$	=	0.0872 ± 0.0015	(1.68%)
(15) $\hat{\alpha}$	=	0.0955 ± 0.0016	(1.68%)

The propagated standard deviation value of 0.0016 corresponds to a relative error of 1.68%.

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.

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

$$\begin{array}{ll} g_f &= 1.0005 \text{ (ref 3)} & s_a &= 1.287 \text{ (ref 8)} \\ s_f &= 1.165 \text{ (ref 8)} & r &= 0.031 \\ g_a &= 0.9979 \text{ (ref 3)} & \hat{\alpha} &= 0.0955 \end{array}$$

The computed value of α_0 is 0.0942 ± 0.0016 . The error (standard deviation) of 0.0015 was computed assuming no uncertainty in the r, g, and s factors.

Lanthanide Fission Yields of U-233, F. L. Lisman, S. F. Marsh, W. J. Maeck, F. W. Sprakles

The absolute fission yields of neodymium, samarium, and cerium were determined in the same manner as described in the previous section for the lanthanide fission yields of U-235.

A three-column sequential procedure is used for the separation of the individual lanthanides. The first column separates the lanthanides from the actinides and most fission products. Cerium is separated from the other lanthanides on a second column. On the third column, the lanthanides are chromatographically separated with α -hydroxyisobutyric acid at controlled pH as the elutriant. Details of the procedure have been previously reported(2).

The analysis results and computed absolute U-233 fission yields for neodymium, cerium, and samarium are summarized in Tables VIII, IX, and X respectively.

Again the yield of the mass 144 chain was calculated three different ways; from the neodymium analysis, from the cerium analysis, and from 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 4.48 ± 0.09 . 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 4.46 ± 0.13 . 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 4.48 ± 0.09 .

The yield of Sm-149 was determined by summing the determined atoms of Sm-149 and Sm-150. Small corrections were made for the loss of Sm-150 by neutron capture.

Cs-137 Fission Yield of U-233, M. E. Kussy, W. J. Maeck

The number of atoms of Cs-137 in the capsule dissolver solution was determined by gamma counting after radiochemistry separation. Two different gamma counting techniques were used. One was based on using the 3x3 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.). The computation of the fission yield is given in Table XI.

Table VIII. U-233 THERMAL FISSION YIELDS OF THE STABLE NEODYMIUM ISOTOPES

$$\text{Number of Fissions} = (3.3904 \pm 0.0620) \times 10^{19} \quad (1.82\%)$$

	Nd-143	Nd-144	Nd-145	Nd-146	Nd-148	Nd-150
Determined number of atoms	1.842×10^{18}	0.936×10^{18}	1.087×10^{18}	3.253×10^{17}	4.238×10^{17}	1.599×10^{17}
Std dev _{avg} (n=4)	0.011×10^{18}	0.0049×10^{18}	0.005×10^{18}	0.012×10^{17}	0.006×10^{17}	0.007×10^{17}
Atoms after correction for neutron capture	1.889×10^{18}	1.520×10^{18} ^a	1.094×10^{18}	8.187×10^{17}	4.225×10^{17}	1.599×10^{17}
Std dev, % (includes estimated uncertainties in correction factors.)	0.67	1.1	0.49	0.20	0.20	0.44
Percent fission yield	5.57	4.48	3.22	2.41	1.25	0.471
Std dev	0.12	0.09	0.06	0.04	0.02	0.009
Std dev, %	2.15	2.01	1.86	1.66	1.60	1.91

^a Includes correction for holdup by Ce-144.

Table IX. U-233 THERMAL FISSION YIELDS OF LONG-LIVED AND STABLE CERIUM ISOTOPES

$$\text{Number of Fissions} = (3.3904 \pm 0.0620) \times 10^{19} \quad (1.82\%)$$

	Ce-140	Ce-142	Ce-144
Determined number of atoms	2.139×10^{18}	2.159×10^{18}	5.401×10^{17}
Std dev _{avg} (n=4)	0.020×10^{18}	0.028×10^{18}	0.117×10^{17}
Atoms after correction for neutron capture and decay	2.137×10^{18}	2.155×10^{18}	1.511×10^{18a}
Std dev % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.92	1.28	2.40
Percent fission yield	6.30	6.35	4.46
Std dev	0.13	0.14	0.13
Std dev, %	2.08	2.23	3.02

a

Includes in-pile and out-pile decay corrections.

Table X. U-233 THERMAL FISSION YIELDS OF STABLE SAMARIUM ISOTOPES

Number of Fissions = $(3.3904 \pm 0.0620) \times 10^{19}$ (1.82%)

	Sm-147	Sm-148	Sm-149	Sm-150	Sm-151	Sm-152	Sm-154
Determined number of atoms	1.389×10^{17}	0.066×10^{17}	0.255×10^{17}	2.157×10^{17}	0.368×10^{17}	1.289×10^{17}	0.812×10^{16}
Std dev _{avg} (n=4)	0.033×10^{17}	0.001×10^{17}	0.0027×10^{17}	0.004×10^{17}	0.002×10^{17}	0.005×10^{17}	0.008×10^{18}
Atoms after correction for neutron capture and radioactive decay	6.044×10^{17}	None	2.406×10^{17}	None	1.231×10^{17}	0.564×10^{17}	0.812×10^{16}
Std dev % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	3.82		0.20		5.03	3.04	1.00
Percent fission yield	1.78		0.710		0.363	0.166	0.024
Std dev	0.07		0.013		0.019	0.006	0.0005
Std dev, %	4.2		1.8		5.3	3.5	2.1

Table XI. U-233 THERMAL FISSION YIELD OF Cs-137

Atoms Cs-137 using crystal as standard

$$= (2.229 \pm 0.056) \times 10^{18} \quad (2.5\%)$$

Atoms Cs-137 by comparison method

$$= (2.255 \pm 0.070) \times 10^{18} \quad (3.1\%)$$

Weighted value, atoms Cs-137^a

$$= (2.239 \pm 0.044) \times 10^{18} \quad (1.9\%)$$

Number of fissions

$$= (3.3904 \pm 0.0062) \times 10^{19} \quad (1.8\%)$$

Fission yield Cs-137

$$= 6.60 \pm 0.17 \quad (2.6\%)$$

^a

Values were averaged by weighting the reciprocals of the variances.

Krypton and Xenon Fission Yields for U-233, R. E. Foster, W. J. Maeck,
F. W. Spraktes

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(1,2).

The computed fission yields of the stable krypton and xenon isotopes and of 10.76-yr Kr-85 are given in Table XII. The error associated with each value is the standard deviation.

Table XII. U-233 THERMAL FISSION YIELDS OF
STABLE KRYPTON AND XENON ISOTOPES

<u>Isotope</u>	<u>Percent Fission Yield</u>
Kr-83	0.985 ± 0.024
Kr-84	1.63 ± 0.04
Kr-85 (10.76 yr)	0.484 ± 0.012
Kr-86	2.74 ± 0.07
Xe-131	3.44 ± 0.09
Xe-132	4.71 ± 0.12
Xe-134	5.93 ± 0.15

SUMMARY OF DETERMINED U-233 AND U-235 THERMAL FISSION YIELDS
AND CAPTURE-TO-FISSION RATIO

Summarized in Table XIII are the determined U-235 absolute thermal fission yields from the most recently dissolved standard burnup capsule. Also reported in this table are the determined yields from the first capsule. From these two sets of data a weighted fission yield value has been calculated. This weighted value was obtained by weighting the reciprocals of the individual variances. In all cases except two (Ce-140 and Nd-144), the individual values could be pooled based on a statistical "t" test at the 95% confidence level.

The U-233 absolute thermal fission yields determined on the second U-233 standard burnup capsule are summarized in Table XIV. Also reported in this table are the determined yields from the first U-233 capsule. A statistical "t" test at the 95% confidence level of the individual values showed that many of the values were significantly different and hence a weighted mean could not be calculated. In all cases the fission yields determined on the second capsule, 6-4-3, were significantly lower than those of the first capsule, 6-4-2. Also the computed U-233 capture-to-fission ratio for capsule 6-4-3 is 0.0942 ± 0.0016 which is significantly lower than 0.0986 ± 0.0015 , the value for capsule 6-4-2. Interestingly, both the value of the U-233 capture-to-fission ratio and many of the determined U-233 fission yields for capsule 6-4-3 are 4 to 6% smaller than the values determined on capsule 6-4-2. Thus we believe that the source of error resulting in these differences is probably in the determined number of fissions. The main uncertainty is the initial loading of the capsule. At a U-233 burnup of $\sim 12\%$, a 0.5% error in the initial loading results in $\sim 4\%$ error in the determined number of fissions.

From the available data on these two capsules it is not possible to determine which set of values is most nearly correct. However; in considering a recently determined value for the U-233 thermal capture-to-fission ratio of 0.0939 ± 0.0012 by Okazaki et al(8) and a recommended weighted value by Westcott et al(9) of 0.0921 ± 0.0029 , we believe that the values determined on capsule 6-4-3 are more nearly correct. At the 95% confidence level, both of these literature values for α_0 are statistically consistent with our determined value of 0.0942 ± 0.0016 for capsule 6-4-3 while the value of 0.0986 ± 0.0015 for capsule 6-4-2 is not.

Admittedly, this type of evaluation is not entirely valid. However, this discrepancy should be resolved in this program as additional capsules are analyzed.

Summarized in Table XV are the determined effective and thermal capture-to-fission ratios for U-233 and U-235. Discrepancies in the U-233 values have been discussed above. Excellent agreement for the U-235 capture-to-fission ratio was obtained between the two analyzed capsules.

Table XIII. SUMMARY OF U-235 FISSION YIELDS

<u>Mass No.</u>	<u>Element</u>	<u>Capsule 6-4-1</u>	<u>Capsule 6-6-2</u>	<u>Weighted Value</u>
83	Kr	0.491±0.020	0.499±0.010	0.497±0.009
84	Kr	0.912±0.027	0.936±0.019	0.920±0.016
85(10.76y)	Kr	0.262±0.007	0.266±0.005	0.265±0.004
86	Kr	1.79±0.06	1.82±0.04	1.81±0.03
131	Xe	2.78±0.09	2.87±0.06	2.84±0.05
132	Xe	4.15±0.14	4.28±0.09	4.24±0.08
134	Xe	7.53±0.25	7.73±0.20	7.62±0.15
137(30.3y)	Cs	6.22±0.14	6.20±0.15	6.21±0.10
140	Ce	6.24±0.07	6.41±0.09	6.30±0.06 ^a
142	Ce	5.84±0.07	5.92±0.08	5.87±0.05
143	Nd	5.88±0.07	5.91±0.09	5.89±0.06
144	Ce	5.36±0.06	5.48±0.09	5.40±0.05
144	Nd	5.31±0.06	5.46±0.09	5.36±0.05 ^a
144	Ce+Nd	5.36±0.06	5.48±0.08	5.40±0.05
145	Nd	3.87±0.05	3.89±0.06	3.88±0.04
146	Nd	2.94±0.03	2.96±0.04	2.95±0.02
147	Sm	2.09±0.07	2.16±0.07	2.13±0.05
148	Nd	1.68±0.02	1.69±0.03	1.68±0.02
149	Sm	1.00±0.01	1.01±0.01	1.00±0.01
150	Nd	0.634±0.007	0.646±0.010	0.630±0.006
151	Sm	0.426±0.022	0.436±0.023	0.431±0.016
152	Sm	0.225±0.007	0.228±0.008	0.226±0.005
154	Sm	0.057±0.003	0.056±0.001	0.056±0.001

^a Based on a "t" test at the 95% confidence level, the two individual values were significantly different and hence the weighted value is not statistically valid at this confidence level.

Table XIV. SUMMARY OF U-233 FISSION YIELDS

<u>Mass No.</u>	<u>Element</u>	<u>Capsule 6-4-2</u>	<u>Capsule 6-4-3</u>
83	Kr	1.02 ± 0.02	0.985 ± 0.024
84	Kr	1.68 ± 0.04	1.63 ± 0.04
85 (10.76y)	Kr	0.497 ± 0.011	0.484 ± 0.012
86	Kr	2.84 ± 0.06	2.74 ± 0.07
131	Xe	3.56 ± 0.07	3.44 ± 0.09
132	Xe	4.86 ± 0.10	4.71 ± 0.12
134	Xe	6.10 ± 0.13	5.93 ± 0.15
137 (30.3y)	Cs	6.69 ± 0.17	6.60 ± 0.17
140	Ce	6.54 ± 0.10	6.30 ± 0.13
142	Ce	6.66 ± 0.11	6.35 ± 0.14
143	Nd	5.87 ± 0.10	5.57 ± 0.12
144	Ce	4.63 ± 0.08	4.46 ± 0.13
144	Nd	4.74 ± 0.09	4.48 ± 0.09
144	Nd+Ce	4.69 ± 0.08	4.48 ± 0.09
145	Nd	3.40 ± 0.06	3.22 ± 0.06
146	Nd	2.54 ± 0.04	2.41 ± 0.04
147	Sm	1.79 ± 0.06	1.78 ± 0.07
148	Nd	1.31 ± 0.02	1.25 ± 0.02
149	Sm	0.746 ± 0.013	0.710 ± 0.013
150	Nd	0.498 ± 0.008	0.471 ± 0.009
151	Sm	0.366 ± 0.019	0.363 ± 0.019
152	Sm	0.177 ± 0.006	0.166 ± 0.006
154	Sm	0.023 ± 0.007	0.024 ± 0.0005

Table XV. CAPTURE-TO-FISSION RATIO
FOR U-233 AND U-235

<u>U-233</u>		
<u>Capsule</u>	<u>$\alpha_{\text{eff.}}$</u>	<u>α_0</u>
6-4-2	0.0999 $\pm$ 0.0015	0.0986 $\pm$ 0.0015
6-4-3	0.0955 $\pm$ 0.0016	0.0942 $\pm$ 0.0016
<u>U-235</u>		
<u>Capsule</u>	<u>$\alpha_{\text{eff.}}$</u>	<u>α_0</u>
6-4-1	0.1770 $\pm$ 0.0019	0.1716 $\pm$ 0.0019
6-6-2	0.1769 $\pm$ 0.0033	0.1715 $\pm$ 0.0024

CORRECTIONS FOR THE BUILDUP AND BURNOUT OF STABLE FISSION PRODUCTS

F. L. Lisman, W. J. Maeck, F. W. Spraktes

The accurate determination of absolute fission yields, for samples which have undergone long term irradiations, requires corrections for the buildup and burnout of the various fission products due to neutron capture. Also required are corrections for the decay of radioactive nuclides during reactor shutdown periods.

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 samples. These programs yield the number of individual fission product atoms formed during the irradiation and their manner of formation. In addition the programs consider the decay of any radioactive nuclides for the time period between termination of the irradiation and sample analysis.

From these computer data the total number of atoms of a particular mass number (A) formed directly from the fission process is obtained. This number excludes any nuclides of mass A formed by neutron capture on A-1 or lower A nuclides but includes any nuclides of mass A which have been transformed to A+1 or higher nuclides by neutron capture. Also obtained is the number of atoms of the particular nuclide of mass A, the nuclide for which analysis is made, present at the time of analysis. The ratio of these two numbers (formed/present) is taken as a measure of the buildup and burnout of that particular mass chain.

The number of atoms of a particular fission product found experimentally to be present at the time of sample analysis is then multiplied by the above ratio to give the true number of atoms of mass A formed directly in the fission process. The absolute fission yield for the mass chain is then calculated using this corrected number.

$$(FY)_A = \frac{K_A N_a}{F^0}$$

where:

$(FY)_A$ = fission yield of chain A.

K_A = number of atoms of mass A formed in fission/
number of atoms of "a" present at analysis.
(both numbers being obtained from the computer
program).

N_a = number of "a" atoms experimentally found to
be present.

F^0 = number of fissions.

Input data for the computer programs include the irradiation history of the sample, total number of fissions which occurred, relative fission yields, decay properties of significant radionuclides, and the neutron capture cross sections for the various fission products.

The irradiation history of the sample is obtained from reactor operations, with the total integrated flux and number of fissions being obtained from the depletion of the original fissile nuclide. The relative fission yields used were those reported by Farrar(10,11) for U-235 and by Ferguson(12) for U-233. It is thought that in using this ratio technique any error involved in normalization of these relative fission yields will be minimized. Neutron capture cross sections present a slightly more complex problem. For the majority of fission products the capture cross sections are small ($< 20b$) and their buildup and burnout is not appreciable. However, a few (notably the samarium isotopes) possess very large capture cross sections ($> 100b$) and in these instances buildup and burnout is appreciable. For these cases it is necessary to know the respective cross sections to a fairly high degree of accuracy ($\sim 5\%$). Current literature values do not indicate that this is the case. Pending the acquisition of more precise cross section data, a technique is being employed which it is hoped will circumvent these uncertainties in the cross section data.

This technique requires that the various isotopic ratios, for a series of isotopes, as predicted by the computer programs be in strict agreement with those found experimentally. If there is disagreement, the error is assumed to be in the various neutron capture cross sections used in the programs, and these values are adjusted to bring the calculated and experimental isotopic ratios into agreement. In this manner it is hoped that working values for neutron capture cross sections will be obtained which will then enable the determination of absolute fission fields for Pu-239 and Pu-241 samples. In the case of these later two fissile nuclides, reliable relative fission yield data are not available and a treatment similar to that being used for U-235 and U-233 is not possible.

An error has been assigned to each of the correction factors applied to the U-235 and U-233 data. These errors are estimates of the reliability of these factors based upon considerations of the magnitude of the correction factor, and the estimated accuracy of the various neutron capture cross sections and relative fission yields used in the computer programs.

The various correction factors used for U-235 and U-233 capsules are given in Table XVI. The computer programs are shown schematically in the Appendix along with the neutron capture cross sections and half lives used in each program. A specimen calculation for fission product Nd-143 is given in the Appendix for the U-235 capsule, 6-6-2.

Table XVI. CORRECTION FACTORS AND ESTIMATED ERRORS FOR THE BUILDUP AND BURNOUT OF FISSION PRODUCTS FOR U-235 AND U-233 CAPSULES

Mass No.	Element	Capsule 6-4-3 (U-233)		Capsule 6-6-2 (U-235)	
		Correction Factor	% Estimated Error	Correction Factor	% Estimated Error
83	Kr	1.0200	0.3	1.0216	0.3
84	Kr	0.9868	0.3	0.9891	0.3
85(10.76y)	Kr	1.0054	0.3	1.0054	0.3
86	Kr	0.9998	nil	0.9998	nil
131	Xe	1.0090	0.3	1.0086	0.3
132	Xe	0.9937	0.3	0.9944	0.3
134	Xe	0.9955	0.3	0.9961	0.3
140	Ce	0.9989	0.3	0.9989	0.3
142	Ce	0.9978	0.3	0.9978	0.3
143	Nd	1.0256	0.3	1.0241	0.3
144	Nd	1.5404	1.0	1.6152	1.0
144	Ce	2.7987	1.0	2.6449	1.0
144	Ce+Nd	0.9521 ^a	0.5	0.9642 ^a	0.5
145	Nd	1.0062	0.3	1.0059	0.3
146	Nd	0.9920	0.3	0.9925	0.3
147	Sm	4.3510	3.0	4.2892	3.0
148	Nd	0.9970	0.3	0.9974	0.3
149	Sm	0.9973	0.3	0.9991	nil
150	Nd	1.0000	nil	1.0001	nil
151	Sm	3.3439	5.0	3.2213	5.0
152	Sm	0.4376	3.0	0.4682	3.0
154	Sm	1.0001	nil	1.0004	nil

^a Applies only to the Nd-144 fraction of the total 144 atoms.

REFERENCES

1. Maeck, W. J., Rein, J. E., Editors, Burnup Determination of Nuclear Fuels: 1964 Annual Report, IDO-14656 (1965).
2. Maeck, W. J., Rein, J. E., Editors, Burnup Determination of Nuclear Fuels: Project Report for the Quarter January 1-March 31, 1965, IDO-14660 (1965).
3. Westcott, C. H., Effective Cross Section Values for Well Moderated Thermal Reactor Spectra, CRRP-960 (AECL-1101) (1960).
4. Westcott, C. H., Walker, W. H., Alexander, T. K., Effective Cross Sections and Cadmium Ratios for the Neutron Spectra of Thermal Reactors, A/Conf. 15/P/202 (1958).
5. Critoph, E., Effective Cross Sections for U-235 and Pu-239, CRRP-1191 (CRRP-960 Addendum) (1964).
6. Okazaki, A., Lounsbury, M., Durham, R. W., Crocker, I. H., A Determination of the Ratio of Capture to Fission Cross Sections of U-235, AECL-1965 (1964).
7. Heath, R. L., Scintillation Spectrometry Gamma Ray Spectrum Catalogue, 2nd Ed., Vol. 1 of 2, IDO-16880 (1964).
8. Okazaki, A., Lounsbury, M., Durham, R. W., A Determination of the Ratio of Capture-to-Fission Cross Section of U-233, AECL-2148 (1964).
9. Westcott, C. H., Ekbeng, K., Hanna, G. C., Pattenden, N. J., Sanatani, S., Attree, P. M., "A Survey of Values of the 2200 m/s Constants for Four Fissile Nuclides," Atomic Energy Review, Vol. 3, No. 2, 3 (1965).
10. Farrar, H., Tomlinson, R. H., "Cumulative Yields of the Heavy Fragments in U-235 Thermal Neutron Fission," Nucl. Phys. 34, 367 (1962).
11. Farrar, H., Fickel, H. R., Tomlinson, R. H., "Cumulative Yields of Light Fragments in U-235 Thermal Neutron Fission," Can. J. Phys., 33, 1017 (1962).
12. Ferguson, R. L., O'Kelly, G. D., A Survey and Evaluation of U-233, Fission Yield Data, ORNL-3305 (1962).

APPENDIX

COMPUTER PROGRAM SCHEMATICS FOR THE BUILDUP AND BURNOUT OF
FISSION PRODUCTS AND SAMPLE CALCULATION OF THE CORRECTION
FACTOR FOR ND-143

F. L. Lisman, W. J. Maeck, F. W. Spraktes

APPENDIX

Computer Programs for the Buildup and Burnout of Fission Products

Schematics of the computer programs used to determine the buildup and burnout of fission product Sm, Nd, Ce, Xe, Kr, are shown in Figures A-1 thru A-4 respectively. These programs were written such that they simulate the actual irradiation history of the various capsules and print out data for the beginning and end of each shutdown period, which occurred in the MTR, during the course of the irradiation.

The number of fissions is obtained from the depletion of the original U-235 or U-233 in the various capsules(2). The fission yield values employed for the U-235 capsules are shown above the lines which connect the fissile nuclide with the first significant nuclide for each mass chain. Fission yield values employed for the U-233 capsules are listed below these lines. The neutron capture cross sections used are shown between the respective A and (A+1) nuclides and are listed in barns (10^{-24} cm²). These values are effective cross sections for $\phi_{res} = \frac{1}{28.6} \phi_{th}$ or a Westcott epithermal index "r" of 0.0311. Where possible these values were obtained using the standard Westcott convention.

$$\hat{\sigma} = \sigma_0 (g+rs)$$

The necessary half lives for radioactive nuclides are shown between the respective Z and Z+1 nuclides. The majority of these half lives were obtained from the GE Chart of the Nuclides (1964 Ed.). The various paths for which calculations were carried out are indicated in the schematics by connecting arrows.

Calculation of the Correction Factor for the Buildup and Burnout of Fission Product Nd-143

This specimen calculation is given for the Nd-143 which resulted from the neutron irradiation of the U-235 containing capsule labeled 6-6-2. This particular capsule was irradiated in the MTR for three fuel cycles with an accumulation of approximately 2.3×10^{20} n/cm². The total number of fissions which occurred in the capsule was determined, from the depletion of the original U-235 loading, to be 2.201×10^{19} . The fission yield for Nd-143 used in the computer program along with the various neutron capture cross sections and half lives are given in the computer program schematic for the neodymium isotopes in the appendix.

The correction factor for buildup and burnout is taken as the predicted ratio of the number of atoms with A of 143 which were formed initially from the fission process to the number of Nd-143 atoms present at the time the neodymium samples were analyzed.

Figure A-1. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT SAMARIUM

Figure A-2. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT CERIUM AND NEODYMIUM

Figure A-3. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT XENON

Figure A-4. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP
AND BURNOUT OF FISSION PRODUCT KRYPTON

The predicted number of Nd-143 atoms present at the time of analysis is simply the number of atoms predicted to be present at the time the capsule was discharged from the reactor plus any Ce-143 and Pr-143 which was present at discharge and decayed to Nd-143 during the time which lapsed between discharge and separation of the neodymium fraction from the gross sample. In the case of capsule 6-6-2, a period of 342 days elapsed between capsule discharge and the neodymium separation. Thus essentially all of the Ce-143 and Pr-143 would have decayed to Nd-143.

The number of atoms with A of 143 actually formed from fission is obtained by summing all nuclides of the 143 mass chain which have fissioning U-235 as their origin as calculated by the computer program. This summation must include any of the 143 chain which is lost by neutron capture to the 144 and higher mass chains. However it must not include any mass 143 nuclides which were formed by neutron capture on nuclides of $A \leq 142$. The detailed calculation is given below.

CALCULATION OF THE BUILDUP AND BURNOUT OF FISSION PRODUCT Nd-143 FOR THE U-235 CAPSULE 6-6-2

A. Nd-143 present at the time of sample analysis: $t = 342$ days

$$\begin{aligned}
 \text{Nd-143 at discharge} &= 8.07214 \times 10^{17} \text{ atoms} \\
 \text{Pr-143} \rightarrow \text{Nd-143} &= 4.24514 \\
 \text{Ce-143} \rightarrow \text{Pr-143} \rightarrow \text{Nd-143} &= \underline{0.51506} \\
 &= 12.83234 \times 10^{17}
 \end{aligned}$$

B. Nuclides with A = 143 formed from the fission process:

$$\begin{aligned}
 \text{U-235} \rightarrow \text{Ce-143} &= 2.17403 \times 10^{17} \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Ce-144} &= 0.00063 \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Pr-143} &= 8.15459 \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Pr-143} \rightarrow \text{Pr-144} &= 0.00006 \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Pr-143} \rightarrow \text{Pr-144} \rightarrow \text{Nd-144} &= 0.02382 \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Pr-143} \rightarrow \text{Pr-144} \rightarrow \text{Nd-144} \rightarrow \text{Nd-145} &= \text{nil} \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Pr-143} \rightarrow \text{Nd-143} &= 2.76764 \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Pr-143} \rightarrow \text{Nd-143} \rightarrow \text{Nd-144} &= 0.02146 \\
 \text{U-235} \rightarrow \text{Ce-143} \rightarrow \text{Pr-143} \rightarrow \text{Nd-143} \rightarrow \text{Nd-144} \rightarrow \text{Nd-145} &= \underline{\text{nil}} \\
 &= 13.14223 \times 10^{17}
 \end{aligned}$$

Correction factor (K) = formed/present = B/A = $13.14223 \times 10^{17} / 12.83234 \times 10^{17}$

$$K = 1.02414$$

estimated error = 0.3%

$$K = 1.02414 \pm 0.00307$$

Fission Yield of Nd-143:

Determined No. of fissions = $(2.2010 \pm 0.0305) \times 10^{19}$

Determined No. of Nd-143 atoms = $(1.2702 \pm 0.0086) \times 10^{18}$

$$(FY) = \frac{K_A N_a}{F^0}$$

$$(FY) = \frac{(1.0241 \pm 0.0031)(1.2702 \pm 0.0086) \times 10^{18}}{(2.2010 \pm 0.0305) \times 10^{19}}$$

$$(FY) = 5.91 \pm 0.09$$

**PHILLIPS
PETROLEUM
COMPANY**

ATOMIC ENERGY DIVISION