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**BURNUP DETERMINATION OF NUCLEAR FUELS
PROJECT REPORT FOR THE
QUARTER JANUARY 1 - MARCH 31, 1967**

Edited by

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

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
IDAHO FALLS, IDAHO

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PROJECT REPORT
FOR THE QUARTER JANUARY 1 - MARCH 31, 1967

Edited by

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

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SUMMARY

In this continuing program to develop methods for the accurate determination of the burnup of nuclear fuels, the main emphasis this quarter was the measurement of the absolute, thermal fission yields of stable Rb-85, Rb-87, Sr-88, Ba-138 and 28y Sr-90 for U-233 and U-235 by isotope dilution mass spectrometry. These values, determined on each of four dissolved U-233 capsules and three U-235 capsules, are:

Fission Yields for U-233

<u>Isotope</u>	<u>Capsule</u>			
	<u>6-2-4</u>	<u>6-4-2</u>	<u>6-4-3</u>	<u>6-5-1</u>
Rb-85	2.25±0.02 ₈	2.20±0.03 ₈	2.12±0.04 ₀	2.34±0.05 ₀
Rb-87	4.09±0.04 ₉	4.04±0.07 ₁	3.90±0.07 ₄	4.28±0.09 ₂
Sr-88	5.55±0.06 ₃	5.62±0.08 ₉	5.39±0.10 ₀	5.89±0.12 ₈
Sr-90	6.93±0.08 ₅	6.99±0.11 ₇	6.71±0.12 ₆	7.36±0.16 ₀
Ba-138	6.01±0.07 ₁	6.03±0.09 ₈	5.79±0.10 ₇	6.21±0.13 ₃

Fission Yields for U-235

<u>Isotope</u>	<u>Capsule</u>		
	<u>6-2-2</u>	<u>6-4-1</u>	<u>6-6-2</u>
Rb-85	1.28±0.02 ₆	1.30±0.01 ₆	1.31±0.01 ₉
Rb-87	2.54±0.01 ₉	2.57±0.03 ₂	2.58±0.03 ₈
Sr-88	3.61±0.02 ₂	3.60±0.04 ₅	3.59±0.05 ₂
Sr-90	5.90±0.04 ₅	5.90±0.07 ₄	5.89±0.08 ₈
Ba-138	6.81±0.04 ₄	6.80±0.08 ₄	6.76±0.09 ₇

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

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

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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 analysis depends on three factors; the accuracy of the values used for the fission yields, the linearity between the number of fissions and the number of fission product atoms present at the end of the irradiation, and the reliability of the analysis methods.

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

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

ABSOLUTE U-233 AND U-235 THERMAL FISSION YIELDS
FOR THE STABLE AND LONG-LIVED FISSION PRODUCTS
OF RUBIDIUM, STRONTIUM, AND BARIUM

The major goal of the burnup project is the accurate determination of the thermal fission yields of long-lived and stable fission products. A series of capsules containing weighed amounts of the oxides of highly enriched U-233, U-235, Pu-239, and Pu-241 was prepared and irradiated in the MTR(I). During this quarter the absolute thermal fission yields of Rb-85, Rb-87, Sr-88, Sr-90, and Ba-138 for U-233 and U-235 were determined by isotope dilution mass spectrometry for four U-233 and three U-235 capsules.

IRRADIATION HISTORY OF CAPSULES

The U-233 and U-235 standard burnup capsules, which contained weighed amounts of U_3O_8 (highly enriched in the respective isotopes) blended with high purity aluminum powder and weld-sealed in aluminum tubes, were irradiated in the MTR at a position where the unperturbed flux was 6×10^{13} n/cm²-sec and the cadmium ratio for cobalt was 15. The irradiation histories of these capsules are given in Table I.

DISSOLUTION OF CAPSULES (R. E. Foster, Jr., S. F. Marsh)

Dissolution of the U-233 and U-235 capsules was carried out in a 4-in. lead-shielded facility. The dissolution apparatus has been previously described (1,2). The dissolver flask and reflux condenser were fabricated from high-purity quartz and an acid mixture of 6M HCl-1.6M HNO₃, prepared from quartz-distilled reagents, was the dissolution reagent(1).

The dissolutions proceeded smoothly and were usually complete in 3.5 hr. The fission gases, krypton and xenon, appeared approximately 1 hr after the start of dissolution and their evolution continued for about 2 hr. The final solutions (about 200 ml) were water clear and contained no residue.

After cooling, the dissolver solutions were quantitatively transferred either to a 250-ml class "A" Pyrex volumetric flask or to a calibrated 250-ml quartz volumetric flask with the aid of 6M HCl-1.6M HNO₃ rinses. The solutions were diluted to volume with quartz distilled water, a Teflon-coated bar magnet was introduced, and the contents were stirred for 1 hr.

DETERMINATION OF NUMBER OF FISSIONS (F. L. Lisman, 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 capsules. 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 numbers of fissions were calculated.

The U_3O_8 materials (obtained from ORNL) that were weighed into the capsules were analyzed for both weight percent uranium content and isotopic distribution. For the former, samples of U_3O_8 were weighed on a micro balance and dissolved in nitric acid, weight aliquots were mixed with weight aliquots of standard uranium spike solutions, and the mixtures were analyzed by isotope dilution mass spectrometry. Isotopic distributions were determined on unspiked samples.

The postirradiation atoms of each uranium isotope were determined in a similar manner with unspiked and spiked aliquots of the dissolver solutions.

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

$$F_3 = U^0 - U$$

in which:

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

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

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

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. In all cases, 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. The determined number of fissions and the associated standard deviation for each of the U-233 capsules, along with the appropriate references, are given in Table I.

Uranium-235 The number of U-235 fissions is calculated from the relationship:

$$F_5 = U^0 - U$$

in which:

F_5 = number of U-235 fissions

U^0 = total preirradiated atoms of uranium

U = total postirradiation atoms of uranium

The use of this relationship requires corrections for those atoms of U-236 and U-238 lost by neutron capture and subsequent decay to non-uranium isotopes. This was done with data obtained from a computer program which simulated the irradiation history of the various capsules. The determined number of fissions and the computed standard deviation for each of the U-235 capsules, along with the appropriate references, are given in Table I.

FISSION YIELDS OF Rb-85 AND Rb-87 (F. L. Lisman, F. W. Sprakles)

The absolute thermal fission yields of Rb-85 and Rb-87 were determined on each of the four U-233 and three U-235 standard burnup capsules. Four aliquots spiked with a known amount of Rb-85 and four without spike, from each dissolved capsule solution, were chemically separated and then analyzed by mass spectrometry.

The analysis results and calculated absolute thermal fission yields for U-233 are summarized in Table II and for U-235 in Table III. The method of calculation and the error propagation for the determined number of Rb-85 and Rb-87 atoms are given in Appendix I. To calculate the yield of the 85 mass chain, the number of determined Rb-85 atoms was added to the previously determined atoms of 10.7-yr Kr-85.

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.

The method of correcting for buildup and burnout of the fission products is based on a series of computer programs which simulate the irradiation history of the sample. These programs yield the number of individual fission product atoms formed during the irradiation and their manner of formation. In addition, the programs consider the decay of any radioactive nuclides for the time period between termination of the irradiation and sample analysis. Details of these programs have been previously reported (5).

In the case of Rb-85 and Rb-87, corrections for buildup and burnout by neutron capture and beta decay are small being about 0.2%. Figure I is a schematic diagram of the buildup and burnout program for rubidium.

Table I. COMPOSITION AND IRRADIATION HISTORY OF STANDARD BURNUP CAPSULES

Capsules	Fissionable Isotope	Enrichment wt %	mg U ₃ O ₈ in Capsules	MTR Cycles	% Burnup ^a	Determined Number of Fissions
6-2-4	U-233	99.908	58.8±0.2	8	32.40±0.37	(4.138±0.045)×10 ¹⁹ (Ref 4)
6-4-2	U-233	99.908	152.4±0.2	3	11.44±0.18	(3.786±0.059)×10 ¹⁹ (Ref 5)
6-4-3	U-233	99.908	126.0±0.2	3	12.39±0.23	(3.390±0.062)×10 ¹⁹ (Ref 6)
6-5-1	U-233	99.908	144.4±0.2	2	8.33±0.17	(2.614±0.055)×10 ¹⁹ (Ref 4)
6-4-1	U-235	99.1633	115.2±0.1	3	11.50±0.13	(2.839±0.032)×10 ¹⁹ (Ref 8)
6-6-2	U-235	99.1633	81.9±0.1	3	12.54±0.18	(2.201±0.030)×10 ¹⁹ (Ref 6)
6-2-2	U-235	99.1633	61.2±0.1	8	31.57±0.19	(4.142±0.022)×10 ¹⁹ (Ref 7)

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

Table II. THERMAL FISSION YIELDS OF Rb-85 AND Rb-87 FOR U-233

	Capsule 6-2-4		Capsule 6-4-2		Capsule 6-4-3		Capsule 6-5-1	
	Rb-85	Rb-87	Rb-85	Rb-87	Rb-85	Rb-87	Rb-85	Rb-87
Determined number of atoms	7.3212×10^{17}	1.6907×10^{18}	6.5481×10^{17}	1.5299×10^{18}	5.6571×10^{17}	1.3218×10^{18}	4.8625×10^{17}	1.1181×10^{18}
Std Dev _{avg} (n = 4)	0.0492×10^{17}	0.0082×10^{18}	0.0492×10^{17}	0.0113×10^{18}	0.0339×10^{17}	0.0079×10^{18}	0.0258×10^{17}	0.0056×10^{18}
Atoms after correction for neutron capture and decay	7.3292×10^{17}	1.6912×10^{18}	6.5499×10^{17}	1.5301×10^{18}	5.6590×10^{17}	1.3221×10^{18}	4.8636×10^{17}	1.1183×10^{18}
Std Dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.57	0.48	0.75	0.74	0.60	0.60	0.53	0.50
Number of fissions	$(4.138 \pm 0.045) \times 10^{19}$ (Ref 4)		$(3.786 \pm 0.059) \times 10^{19}$ (Ref 5)		$(3.390 \pm 0.062) \times 10^{19}$ (Ref 6)		$(2.614 \pm 0.055) \times 10^{19}$ (Ref 4)	
Previously reported number of 10.7-yr Kr-85 atoms	$(1.9900 \pm 0.0282) \times 10^{17}$ (Ref 4)		$(1.781 \pm 0.027) \times 10^{17}$ (Ref 5)		$(1.5322 \pm 0.0245) \times 10^{17}$ (Ref 6)		$(1.2654 \pm 0.0018) \times 10^{17}$ (Ref 4)	
Total mass 85 atoms	$(9.3192 \pm 0.056) \times 10^{17}$		$(8.3309 \pm 0.0561) \times 10^{17}$		$(7.1912 \pm 0.0418) \times 10^{17}$		$(6.1290 \pm 0.0259) \times 10^{17}$	
Percent fission yield	2.25	4.09	2.20	4.04	2.12	3.90	2.34	4.28
Std Dev	0.028	0.019	0.038	0.071	0.040	0.074	0.050	0.092
Std Dev, %	1.25	1.15	1.71	1.75	1.90	1.90	2.14	2.16

Table III. THERMAL FISSION YIELDS OF Rb-85 AND Rb-87 FOR U-235

	Capsule 6-2-2		Capsule 6-4-1		Capsule 6-6-2	
	Rb-85	Rb-87	Rb-85	Rb-87	Rb-85	Rb-87
Determined number of atoms	4.3608×10^{17}	1.0507×10^{18}	2.9696×10^{17}	7.2912×10^{17}	2.3276×10^{17}	5.6886×10^{17}
Std Dev _{avg} (n = 4)	0.0229×10^{17}	0.0053×10^{18}	0.0154×10^{17}	0.0154×10^{17}	0.0140×10^{17}	0.0306×10^{17}
Atoms after correction for neutron capture and decay	$5.3221 \times 10^{17} (a)$	1.0510×10^{18}	2.9704×10^{17}	7.3028×10^{17}	2.3282×10^{17}	5.6898×10^{17}
Std Dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	2.0	0.51	0.52	0.52	0.60	0.54
Number of fissions	$(4.142 \pm 0.022) \times 10^{19}$ (Ref 7)		$(2.839 \pm 0.032) \times 10^{19}$ (Ref 8)		$(2.201 \pm 0.030) \times 10^{19}$ (Ref 6)	
Previously reported number of 10.7-yr Kr-85 atoms	(a)		$(0.7144 \pm 0.0143) \times 10^{17}$ (Ref 8)		$(0.5533 \pm 0.0085) \times 10^{17}$ (Ref 6)	
Total mass 85 atoms	$(5.3221 \pm 0.1064) \times 10^{17} (a)$		$(3.6848 \pm 0.0405) \times 10^{17}$		$(2.8815 \pm 0.0164) \times 10^{17}$	
Percent fission yield	1.28	2.54	1.30	2.57	1.31	2.58
Std Dev	0.02 ₆	0.01 ₉	0.01 ₆	0.03 ₂	0.01 ₉	0.03 ₈
Std Dev, %	2.07	0.73	1.22	1.24	1.46	1.48

(a) The krypton was lost inadvertently during dissolution. The corrected number of Rb-85 atoms is based on a calculated estimate of the 10.7-yr Kr-85 contribution.

Figure 1. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT RUBIDIUM

CHEMICAL SEPARATION OF RUBIDIUM (R. E. Foster, Jr., M. E. Kussy)

The procedure developed for the carrier-free separation of rubidium prior to the mass spectrometric analysis is rapid and simple. It essentially is a one-step procedure in which ammonium hydroxide is added to precipitate aluminum hydroxide which carries hydrolyzable fission products. In aged fission product solutions no fission products which interfere with the mass spectrometric determination of rubidium remain in the supernatant solution.

Because natural rubidium is only diisotopic (Rb-85 and Rb-87), contamination with natural rubidium must be prevented as both these isotopes are fission products and Rb-85 is the isotope dilution "spike". The three reagents, ammonium hydroxide, nitric acid, and hydrochloric acid, were tested for natural rubidium contamination by isotope dilution mass spectrometry using Rb-85 as the "spike". Only quartz or polypropylene ware was used in the separation procedure.

The separation procedure and the preparation and standardization of the Rb-85 spike are detailed in Appendix I.

MASS SPECTROMETRIC ANALYSIS OF RUBIDIUM (R. M. Abernathey, L. W. Buttars, R. A. Nielsen, R. E. McAtee, G. D. Workman)

The mass spectrometer used for this work was a Nuclide Corporation, TDD-1, tandem, 12-in., 90° instrument. For the rubidium analysis, a Faraday Cage, electrometer, strip chart detection system was used. Details of this instrument and the detection system have been reported previously (2). An aqueous chloride solution of the separated rubidium was mounted on a single rhenium filament.

The procedure for the mass spectrometric analysis is detailed in Appendix I. Typical data showing the reproducibility of the isotope dilution analysis, obtained for four aliquots of a dissolved, irradiated U-235 sample, are summarized in Table IV. Table V compares the isotopic distribution of natural rubidium obtained with this system to data reported in the Nuclear Data Sheets (3).

FISSION YIELDS OF Sr-88 AND Sr-90 (F. L. Lisman, F. W. Sprakles)

The absolute thermal fission yields of Sr-88 and Sr-90 were determined on each of the four U-233 and three U-235 standard burnup capsules. Four aliquots spiked with a known amount of Sr-87 and four without spike, from each dissolver solution, were chemically separated and then analyzed by mass spectrometry.

The analysis results and calculated absolute thermal fission yields for U-233 are summarized in Table VI and for U-235 in Table VII. The method of calculation and the error propagation for the determined number of Sr-88 and Sr-90 atoms are given in Appendix II.

Table IV. REPRODUCIBILITY OF MASS SPECTROMETRIC PROCEDURE

<u>Aliquot</u>	<u>Rubidium Isotope (atoms/ml x 10⁻¹⁵)</u>	
	<u>85</u>	<u>87</u>
1	1.580	3.807
2	1.576	3.798
3	1.578	3.803
4	1.576	3.804
avg	1.577	3.803
Std Dev (a)	0.002	0.004
% Std Dev	0.13	0.10

(a) Standard deviation for an individual result

Table V. COMPARISON OF ISOTOPIC DISTRIBUTION OF NATURAL RUBIDIUM

<u>Isotope</u>	<u>Value, Atom Percent</u>	
	<u>This Work</u> (a)	<u>Nuclear Data Sheets</u>
85	72.101±0.026	72.15
87	27.899±0.025	27.85

(a) Error is expressed as the standard deviation of the average of four determinations.

Table VI. THERMAL FISSION YIELDS OF Sr-88 AND Sr-90 FOR U-233

	Capsule 6-2-4		Capsule 6-4-2		Capsule 6-4-3		Capsule 6-5-1	
	Sr-88	Sr-90	Sr-88	Sr-90	Sr-88	Sr-90	Sr-88	Sr-90
Determined number of atoms	2.2984×10^{18}	2.6601×10^{18}	2.1296×10^{18}	2.4529×10^{18}	1.8281×10^{18}	2.1076×10^{18}	1.5391×10^{18}	1.7896×10^{18}
Std Dev _{avg} (n = 4)	0.0067×10^{18}	0.0069×10^{18}	0.0061×10^{18}	0.0065×10^{18}	0.0053×10^{18}	0.0055×10^{18}	0.0031×10^{18}	0.0047×10^{18}
Atoms after correction for neutron capture and decay	2.2978×10^{18}	2.8681×10^{18}	2.1293×10^{18}	2.6479×10^{18}	1.8278×10^{18}	2.2752×10^{18}	1.5388×10^{18}	1.9236×10^{18}
Std Dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.29	0.56	0.29	0.56	0.29	0.56	0.53	0.56
Number of fissions	$(4.138 \pm 0.045) \times 10^{19}$ (Ref 4)		$(3.786 \pm 0.059) \times 10^{19}$ (Ref 5)		$(3.390 \pm 0.062) \times 10^{19}$ (Ref 6)		$(2.614 \pm 0.055) \times 10^{19}$ (Ref 4)	
Percent fission yield	5.55	6.93	5.62	6.99	5.39	6.71	5.89	7.36
Std Dev	0.06 ₃	0.08 ₅	0.08 ₉	0.11 ₇	0.10 ₀	0.12 ₆	0.12 ₈	0.16 ₀
Std Dev, %	1.13	1.23	1.59	1.67	1.85	1.88	2.17	2.17

Table VII. THERMAL FISSION YIELDS OF Sr-88 AND Sr-90 FOR U-235

	Capsule 6-2-2		Capsule 6-4-1		Capsule 6-6-2	
	Sr-88	Sr-90	Sr-88	Sr-90	Sr-88	Sr-90
Determined number of atoms	1.4957×10^{18}	2.2690×10^{18}	1.0217×10^{18}	1.5543×10^{18}	7.8950×10^{17}	1.2089×10^{18}
Std Dev _{avg} (n = 4)	0.0042×10^{18}	0.0059×10^{18}	0.0054×10^{18}	0.0034×10^{18}	0.0320×10^{17}	0.0032×10^{18}
Atoms after correction for neutron capture and decay	1.4954×10^{18}	2.4446×10^{18}	1.0216×10^{18}	1.6741×10^{18}	7.8938×10^{17}	1.2965×10^{18}
Std Dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.28	0.56	0.53	0.55	0.40	0.56
Number of fissions	$(4.142 \pm 0.022) \times 10^{19}$ (Ref 7)		$(2.839 \pm 0.032) \times 10^{19}$ (Ref 8)		$(2.201 \pm 0.030) \times 10^{19}$ (Ref 6)	
Percent fission yield	3.61	5.90	3.60	5.90	3.59	5.89
Std Dev	0.02 ₂	0.04 ₅	0.04 ₅	0.07 ₄	0.05 ₂	0.08 ₈
Std Dev, %	0.60	0.77	1.25	1.25	1.44	1.49

As in the case of the thermal fission yields of rubidium, corrections for the buildup and burnout of the strontium isotopes is based on a computer program which simulates the irradiation history of the capsules. A schematic diagram of the program for strontium is given in Figure 2.

CHEMICAL SEPARATION OF STRONTIUM (R. E. Foster, Jr., M. E. Kussy)

The procedure developed for the carrier-free separation of strontium prior to the mass spectrometric analysis is a rapid two-step procedure. Strontium is carried on lead nitrate which is precipitated from fuming nitric acid. After separation, the lead nitrate precipitate is dissolved in quartz-distilled water and diluted with 1.5M HCl. The solution is passed through an anion exchange column, in the chloride form, which retains the lead. The strontium effluents, which contains no fission products that interfere with the subsequent mass spectrometric determination of strontium, is evaporated to dryness and the residue is dissolved in a few drops of water.

To minimize contamination by natural strontium, all reagents are quartz distilled and only polypropylene ware is used in the separation procedure.

The separation procedure and the preparation and standardization of the Sr-87 spike are detailed in Appendix II.

MASS SPECTROMETRIC ANALYSIS OF STRONTIUM (R. M. Abernathey, L. W. Buttars, R. E. McAtee, R. A. Nielsen, G. D. Workman)

The mass spectrometer and associated detection system was the same as described in the previous section. The aqueous chloride solution of the separated strontium was mounted on a single rhenium filament.

The procedure for the mass spectrometric analysis is detailed in Appendix II. Typical data showing the reproducibility of the isotope dilution analysis, obtained for four aliquots of a dissolved, irradiated U-235 sample, are summarized in Table VIII. Table IX compares the isotopic distribution of natural strontium obtained with this system to data reported in the Nuclear Data sheets (3).

FISSION YIELD OF Ba-138 (F. L. Lisman, F. W. Sprakles)

The absolute thermal fission yield of Ba-138 was determined on each of the four U-233 and the three U-235 standard burnup capsules. Four aliquots, spiked with a known amount of Ba-135 and four without spike, from each dissolver solution, were chemically separated and then analyzed by mass spectrometry.

The analysis results and calculated absolute thermal fission yields for U-233 are summarized in Table X, and for U-235 in Table XI. The method of calculation and the error propagation for the determined number of Ba-138 atoms are given in Appendix III.

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

Table VIII. REPRODUCIBILITY OF MASS SPECTROMETRIC PROCEDURE

<u>Aliquot</u>	<u>Strontium Isotope (atoms/ml x 10⁻¹⁵)</u>	
	<u>88</u>	<u>90</u>
1	3.1551	4.8296
2	3.1599	4.8456
3	3.1503	4.8342
4	3.1716	4.8412
Avg	3.1592	4.8376
Std Dev (a)	0.0091	0.0071
% Std Dev	0.29	0.15

(a) Standard deviation for an individual result.

Table IX. COMPARISON OF ISOTOPIC DISTRIBUTION OF NATURAL RUBIDIUM

<u>Isotope</u>	<u>Value, Atom Percent</u>	
	<u>This Work (a)</u>	<u>Nuclear Data Sheets</u>
84	0.5413±0.0007	0.56
86	9.7423±0.0062	9.86
87	6.9683±0.0014	7.02
88	82.7483±0.0071	82.56

(a) Error is expressed as the standard deviation of the average for triplicate determinations.

Table X. THERMAL FISSION YIELD OF Ba-138 FOR U-233

	Capsule 6-2-4	Capsule 6-4-2	Capsule 6-4-3	Capsule 6-5-1
Determined number of atoms	2.4868×10^{18}	2.2837×10^{18}	1.9628×10^{18}	1.6223×10^{18}
Std Dev _{avg} (n = 4)	0.0116×10^{18}	0.0102×10^{18}	0.0082×10^{18}	0.0063×10^{18}
Atoms after correction for neutron capture and decay	2.4872×10^{18}	2.2839×10^{18}	1.9629×10^{18}	1.6223×10^{18}
Std Dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.47	0.45	0.42	0.39
Number of fissions	$(4.139 \pm 0.045) \times 10^{19}$ (Ref 4)	$(3.786 \pm 0.059) \times 10^{19}$ (Ref 5)	$(3.390 \pm 0.062) \times 10^{19}$ (Ref 6)	$(2.614 \pm 0.055) \times 10^{19}$ (Ref 4)
Percent fission yield	6.01	6.03	5.79	6.21
Std Dev	0.07 ₁	0.09 ₈	0.10 ₇	0.13 ₃
Std Dev, %	1.18	1.53	1.85	2.14

Figure 3. COMPUTER PROGRAM SCHEMATIC FOR BUILDUP AND BURNOUT OF FISSION PRODUCT BARIUM

As in the case of the thermal yields of rubidium and strontium, corrections for the buildup and burnout of Ba-138 were based on a computer program which simulated the irradiation history of the capsules. A schematic diagram of the program for Ba-138 is given in Figure 3.

CHEMICAL SEPARATION OF BARIUM (R. E. Foster, Jr., M. E. Kussy)

The procedure used for the carrier-free separation of barium prior to the mass spectrometric analysis is essentially identical to the strontium procedure. Barium is carried on lead nitrate and the lead is subsequently separated from the barium on an anion exchange column. In this procedure, cesium, a potential interference in the mass spectrometric analysis of barium, follows the barium. However, the cesium interference is eliminated in the mass spectrometric analysis procedure for barium described below.

To minimize contamination by natural barium, all reagents were quartz-distilled and only polypropylene ware was used in the separation procedure.

The separation procedure and the preparation and standardization of the Ba-135 spike are detailed in Appendix III.

MASS SPECTROMETRIC ANALYSIS OF BARIUM (R. M. Abernathey, L. W. Buttars, R. A. Nielsen)

The mass spectrometer used was a Phillips Petroleum Co. Model Ma-31, single stage, 12 in., 60° instrument. For the barium analysis a Faraday Cage, electrometer, strip chart recording system was used. The aqueous solution of the separated barium was mounted on a single tantalum filament. The interference due to the small amounts of cesium which carry through the barium separation is minimized by "baking" the cesium from the filament at relatively low filament temperatures. The thermal ionization temperature of cesium is less than that of barium.

The procedure for the mass spectrometric analysis is detailed in Appendix III. Typical data showing the reproducibility of the isotope dilution analysis obtained for four aliquots of a dissolved, irradiated U-235 sample, are summarized in Table XII. Table XIII compares the isotopic distribution of natural strontium obtained with this system to data reported in the Nuclear Data sheets (3).

Table XI. THERMAL FISSION YIELD OF Ba-138 FOR U-235

	Capsule 6-2-2	Capsule 6-4-1	Capsule 6-6-2
Determined number of atoms	2.8220×10^{18}	1.9295×10^{18}	1.4888×10^{18}
Std Dev _{avg} (n = 4)	0.0103×10^{18}	0.0096×10^{18}	0.0059×10^{18}
Atoms after correction for neutron capture and decay	2.8257×10^{18}	1.9296×10^{18}	1.4889×10^{18}
Std Dev, % (includes estimated uncertainties in correction factors for neutron capture and radioactive decay)	0.36	0.50	0.40
Number of fissions	$(4.142 \pm 0.022) \times 10^{19}$ (Ref 7)	$(2.839 \pm 0.032) \times 10^{19}$ (Ref 8)	$(2.201 \pm 0.030) \times 10^{19}$ (Ref 6)
Percent fission yield	6.81	6.80	6.76
Std Dev	0.04 ₄	0.08 ₄	0.09 ₇
Std Dev, %	0.64	1.23	1.44

Table XII. REPRODUCIBILITY OF MASS SPECTROMETRIC PROCEDURE

<u>Aliquot</u>	<u>Ba-138 (atom/ml x 10⁻¹⁵)</u>
1	1.9624
2	1.9646
3	1.9540
4	1.9704
Avg	1.9628
Std Dev	0.0068
% Std Dev	0.35

(a)

Standard deviation for an individual result.

Table XIII. COMPARISON OF ISOTOPIC DISTRIBUTION
OF NATURAL BARIUM

<u>Isotope</u>	<u>Value, Atom Percent</u>	
	<u>This Work</u> ^(a)	<u>Nuclear Data Sheets</u> ^(b)
134	2.4343±0.0124	2.43
135	6.5097±0.0154	6.60
136	7.8607±0.0032	7.83
137	11.2330±0.0110	11.34
138	71.8830±0.0339	71.80

(a)

Error is expressed as the standard deviation of the average for triplicate determinations.

(b)

The N.D.S. data have been normalized to 100% for the isotopes 134 through 138.

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APPENDIX I

PROCEDURE FOR THE SEPARATION, MASS SPECTROMETRIC ANALYSIS, CALCULATION, AND ERROR PROPAGATION FOR THE DETERMINATION OF FISSION PRODUCT RUBIDIUM

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F. L. Lisman, R. A. Nielsen, R. E. McAtee,
G. D. Workman, S. S. Yamamura

PREPARATION OF Rb-85 SPIKE

A 1.4-mg sample of isotopically enriched Rb-85, obtained from ORNL as the chloride, was dissolved in quartz-distilled 1M HCl and diluted to 1 liter with 1M HCl. Weighed 3-ml aliquots were transferred to glass ampoules which were flame sealed.

CALIBRATION OF Rb-85 SPIKE

The calibration technique was isotope dilution mass spectrometry with natural rubidium as the primary standard. Approximately 1 g of "Specpure" rubidium chloride was dissolved in 1M HCl and diluted to 1 liter with quartz-distilled 1M HCl. Weighed 100-ml aliquots were gravimetrically analyzed by fuming to the sulfate with sulfuric acid. The rubidium sulfate samples were then dried overnight at 700°C in a muffle furnace prior to weighing. The determined concentration was 1.0771 mg Rb/g of solution with a relative standard deviation for the average of 0.32%.

Weight aliquots containing approximately 3 mg of rubidium were transferred to glass ampoules which were flame sealed. When used, the entire contents of an ampoule were diluted 1000 fold (by weight) and weight aliquots of the dilution were individually combined with four weight aliquots of the Rb-85 spike solution. The four mixtures then were analyzed by isotope dilution mass spectrometry. The determined concentration of the Rb-85 in the "spike" was 7.6280×10^{15} atoms/g solution with a relative standard deviation for the average of 0.42%.

CHEMICAL SEPARATION

1. Quantitatively transfer approximately 3 μ g of Rb-85 spike to a clean 10-ml polypropylene tube.
2. Accurately pipet a 1-ml aliquot of the dissolver solution into the 10-ml polypropylene tube. Use a calibrated pipet.
3. Add one drop of methyl red indicator solution.
4. Stir the solution. A Vortex mixer is satisfactory.
5. Add quartz-distilled NH_4OH dropwise until the solution turns yellow.
6. Centrifuge.

7. Transfer the supernatant solution to a clean 15-ml polypropylene tube.	Discard any precipitate which usually is a metal hydroxide(s) forming from fuel components.
8. Add 1 ml of conc quartz-distilled HNO_3 .	
9. Evaporate to dryness using a filter air flow and heat lamp.	During the evaporation, NH_4Cl may precipitate. If so, add an additional drop each of conc HNO_3 and HCl and continue the evaporation.

MASS SPECTROMETRIC ANALYSIS

1. Prepare a canoe-shaped filament from high purity, 1-x 30-mil rhenium ribbon, and vacuum bake it for 1 hr at 4 A at a pressure of 1×10^{-6} torr. Assemble the sample filament in the center (ionizing) position.	Most of the trace rubidium impurities are volatilized.
2. Using a new, freshly cleaned, disposable glass pipet, add a few drops of quartz-distilled water to the residue from step 9 (previous section). Dissolve the residue and transfer the solution to the filament. Evaporate the solution by passing 1.2 A through the filament.	
3. Mount the filament holder on the ion source carriage and insert the carriage into the vacuum lock.	Allow about 1-min pumping time at each lock stage.
4. Adjust the filament current to 0.6 to 0.8 A and locate the major isotope beam by reference to the mass-vs DVM (accelerating voltage) curve for the magnetic field in use. Adjust the source voltage divider controls for the maximum beam. Make a rapid voltage scan in both directions to verify the mass identification.	Use the electron multiplier. Keep the J2 setting low, the J10 setting maximum, and the J4-J5 setting near center. The source may need to be rotated to get J4-J5 centered with the maximum beam.
5. Insert the Faraday Cage collector and increase the filament current to obtain an ion current of 3×10^{-11} A. Scan the spectrum	

magnetically in both directions, recording the sensitivity settings used to give near full-scale peak heights. Obtain three charts of 4-1/2 double scans.

6. Graphically interpolate by drawing lines on the chart which connect the tops of the successive peaks of each isotope. Draw vertical lines on the chart at each turnaround in the scans, and take peak height readings at the intersections of these lines with those connecting the peak tops. Sum the readings for each isotope.

Use the same number of readings in each sum. If the atom percent of an individual isotope is desired, it is given by:

$$\text{Atom \% Rb}^A = 100 \frac{\sum \text{peak heights}^A}{\sum (\sum \text{peak heights})}$$

CALCULATIONS

Let R_{sf} be the atom ratio of Rb-85 to Rb-87 in the spiked fission product sample. Then:

$$R_{sf} = \frac{N_s^5 + N_f^5}{N_s^7 + N_f^7} \quad (1)$$

where:

N_s^5 = number of spike Rb-85 atoms in the sample

N_s^7 = number of spike Rb-87 atoms in the sample

N_f^5 = number of fission product Rb-85 atoms in the sample

N_f^7 = number of fission product Rb-87 atoms in the sample

To eliminate the N_f^5 term from equation (1), let R_f be the atom ratio of fission product Rb-85 to Rb-87 in an unspiked product sample. Then:

$$R_f = \frac{N_f^5}{N_f^7}$$

and $N_f^5 = R_f N_f^7$

Substituting for N_f^5 in equation (1) yields

$$R_{sf} = \frac{N_s^5 + R_f N_f^7}{N_s^7 + N_f^7}$$

or $R_{sf} = \frac{N_s^5 + R_f N_f^7}{R_s N_s^5 + N_f^7} \quad (2)$

where:

R_s = atom ratio of Rb-87 to Rb-85 in the Rb-85 spike.

Solving equation (2) for N_f^7 yields:

$$N_f^7 = \frac{N_s^5 (1 - R_{sf} R_s) V_d}{(R_{sf} - R_f) V_x} \quad (3)$$

where:

V_d = volume of the dissolver solution

V_x = volume of sample aliquot

Normally R_f is obtained on four unspiked aliquots and the average $\bar{R}_f$ is used to calculate N_f^7 .

Thus:

$$N_f^7 = \frac{N_s^5 (1 - R_{sf} R_s) V_d}{(R_{sf} - \bar{R}_f) V_x} \quad (4)$$

The number of fission product Rb-85 atoms is:

$$N_f^5 = N_f^7 \bar{R}_f \quad (5)$$

ERROR PROPAGATION FOR Rb-87

The number of Rb-87 atoms, by equation (4) of the previous section is:

$$N_f^7 = \frac{N_s^5 (1 - R_{sf} R_s) V_d}{(R_{sf} - \bar{R}_f) V_x}$$

Reported values are the average of several determinations:

$$\bar{N}_f^7 = \sum_{i=1}^n N_{f_i}^7$$

where:

$\bar{N}_f^7$ = average number of Rb-87 atoms

$N_{f_i}^7$ = number of Rb-87 atoms in the i^{th} sample

n = number of determinations

or:

$$\bar{N}_f^7 = \sum_{i=1}^n \frac{N_{s_i}^5 (1 - R_{sf_i} R_s) V_{d_i}}{(R_{sf_i} - \bar{R}_f) V_{x_i}}$$

V_d and usually V_x are the same for all samples. Also $\bar{R}_f$ and R_s are the same for all determinations on a given capsule.

Thus:

$$\bar{N}_f^7 = \frac{V_d}{nV_x} \sum_{i=1}^n \frac{N_{s_i}^5 (1 - R_{sf_i} R_s)}{(R_{sf_i} - \bar{R}_f)}$$

The variance of $\bar{N}_f^7$ is then given approximately by:

$$\sigma^2 \bar{N}_f^7 = \left(\frac{\partial \bar{N}_f^7}{\partial V_d} \right)^2 \sigma^2 V_d + \left(\frac{\partial \bar{N}_f^7}{\partial V_x} \right)^2 \sigma^2 V_x + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^7}{\partial N_{s_i}^5} \right)^2 \sigma^2 N_{s_i}^5$$

$$\begin{aligned}
& + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^7}{\partial R_{sf_i}} \right)^2 \sigma^2 R_{sf_i} + \left(\frac{\partial \bar{N}_f^7}{\partial R_s} \right)^2 \sigma^2 R_s + \left(\frac{\partial \bar{N}_f^7}{\partial \bar{R}_f} \right)^2 \sigma^2 \bar{R}_f \\
& + 2 \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{\partial \bar{N}_f^7}{\partial N_{s_i}^5} \right) \left(\frac{\partial \bar{N}_f^7}{\partial N_{s_j}^5} \right) \sigma^2 N_{s_{ij}}^5 \\
& + (\text{additional covariance terms})
\end{aligned}$$

The following assumptions are made:

(a) The additional covariance terms are negligible

$$\text{(b) } \frac{\sigma^2 N_{s_{ij}}^5}{\sigma N_{s_i}^5 \sigma N_{s_j}^5} = 1$$

and

$$\begin{aligned}
\sigma^2 \bar{N}_f^7 = & \frac{1}{n^2 V_x^2} \left\{ V_d^2 \left[\frac{1}{V_x^2} \left(\sum_{i=1}^n \frac{N_{s_i}^5 [1 - R_{sf_i} R_s]}{[R_{sf_i} - \bar{R}_f]} \right)^2 \sigma^2 V_x \right. \right. \\
& + \left(\sum_{i=1}^n \frac{N_{s_i}^5 R_{sf_i}}{[R_{sf_i} - \bar{R}_f]} \right)^2 \sigma^2 R_s + \sum_{i=1}^n \left(\frac{[1 - R_{sf_i} R_s]}{[R_{sf_i} - \bar{R}_f]} \right)^2 \sigma^2 N_{s_i}^5 \\
& + \left(\sum_{i=1}^n \frac{N_{s_i}^5 [1 - R_{sf_i} R_s]}{[R_{sf_i} - \bar{R}_f]^2} \right)^2 \sigma^2 \bar{R}_f + \sum_{i=1}^n \left(\frac{N_{s_i}^5 [\bar{R}_f R_s - 1]}{[R_{sf_i} - \bar{R}_f]^2} \right)^2 \sigma^2 R_{sf_i} \Big] \\
& + \left. \left(\sum_{i=1}^n \frac{N_{s_i}^5 [1 - R_{sf_i} R_s]}{[R_{sf_i} - \bar{R}_f]} \right)^2 \sigma^2 V_d \right\} \\
& + 2 \frac{V_d^2}{n^2 V_x^2} \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{[1 - R_{sf_i} R_s]}{[R_{sf_i} - \bar{R}_f]} \right) \left(\frac{[1 - R_{sf_j} R_s]}{[R_{sf_j} - \bar{R}_f]} \right) \sigma N_{s_i}^5 \sigma N_{s_j}^5
\end{aligned}$$

The variance $\sigma^2 \bar{R}_f$ was obtained from multiple determinations on unspiked samples. The variance $\sigma^2 R_{sf_i}$ was obtained from multiple scans on the

individual spiked samples. The variances $\sigma^2 R_s$, $\sigma^2 N_{s_i}^5$, $\sigma^2 V_x$, and $\sigma^2 V_d$ were obtained from independent measurements.

ERROR PROPAGATION FOR Rb-85

The number of fission product Rb-85 atoms is given by:

$$N_{f_i}^5 = N_{f_i}^7 \bar{R}_f$$

Reported values are the average of several determinations and:

$$\bar{N}_f^5 = \sum_{i=1}^n \frac{N_{f_i}^7 \bar{R}_f}{n}$$

or:

$$\bar{N}_f^5 = \frac{V_d}{nV_x} \sum_{i=1}^n \frac{N_{s_i}^5 (1 - R_{sf_i} R_s) \bar{R}_f}{(R_{sf_i} - \bar{R}_f)}$$

The variance of $\bar{N}_f^5$ is given approximately by:

$$\begin{aligned} \sigma^2 \bar{N}_f^5 &= \left(\frac{\partial \bar{N}_f^5}{\partial V_d} \right)^2 \sigma^2 V_d + \left(\frac{\partial \bar{N}_f^5}{\partial V_x} \right)^2 \sigma^2 V_x + \left(\frac{\partial \bar{N}_f^5}{\partial R_s} \right)^2 \sigma^2 R_s \\ &+ \left(\frac{\partial \bar{N}_f^5}{\partial \bar{R}_f} \right)^2 \sigma^2 \bar{R}_f + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^5}{\partial N_{s_i}^5} \right)^2 \sigma^2 N_{s_i}^5 + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^5}{\partial R_{sf_i}} \right)^2 \sigma^2 R_{sf_i} \\ &+ 2 \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{\partial \bar{N}_f^5}{\partial N_{s_i}^5} \right) \left(\frac{\partial \bar{N}_f^5}{\partial N_{s_j}^5} \right) \sigma^2 N_{s_{ij}}^5 + (\text{additional covariance terms}) \end{aligned}$$

The following assumptions are made:

(a) The additional covariance terms are negligible

$$(b) \quad \frac{\sigma^2 N_{s_{ij}}^5}{\sigma N_{s_i}^5 \sigma N_{s_j}^5} = 1$$

and

$$\begin{aligned}
\sigma^2 \bar{N}_f^5 &= \frac{1}{n^2 V_x^2} \left\{ V_d^2 \left[\frac{1}{V_x^2} \left(\sum_{i=1}^n \frac{N_{s_i}^5 (1 - R_{sf_i} R_s) \bar{R}_f}{(R_{sf_i} - \bar{R}_f)} \right)^2 \sigma^2 V_x \right. \right. \\
&+ \left(\sum_{i=1}^n \frac{N_{s_i}^5 \bar{R}_f R_{sf_i}}{(R_{sf_i} - \bar{R}_f)} \right)^2 \sigma^2 R_s + \left(\sum_{i=1}^n \frac{R_{sf_i} N_{s_i}^5 (1 - R_{sf_i} R_s)}{(R_{sf_i} - \bar{R}_f)^2} \right)^2 \sigma^2 \bar{R}_f \\
&+ \left. \sum_{i=1}^n \left(\frac{(1 - R_{sf_i} R_s) \bar{R}_f}{(R_{sf_i} - \bar{R}_f)} \right)^2 \sigma^2 N_{s_i}^5 + \sum_{i=1}^n \left(\frac{N_{s_i}^5 \bar{R}_f (\bar{R}_f R_s - 1)}{(R_{sf_i} - \bar{R}_f)^2} \right)^2 \sigma^2 R_{sf_i} \right] \\
&+ \left. \left(\sum_{i=1}^n \frac{N_{s_i}^5 (1 - R_{sf_i} R_s) \bar{R}_f}{(R_{sf_i} - \bar{R}_f)} \right)^2 \sigma^2 V_d \right\} \\
&+ 2 \frac{V_d^2}{n^2 V_x^2} \bar{R}_f^2 \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{(1 - R_{sf_i} R_s)}{(R_{sf_i} - \bar{R}_f)} \right) \left(\frac{(1 - R_{sf_j} R_s)}{(R_{sf_j} - \bar{R}_f)} \right) \sigma N_{s_i}^5 \sigma N_{s_j}^5
\end{aligned}$$

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APPENDIX II

PROCEDURE FOR THE SEPARATION, MASS SPECTROMETRIC ANALYSIS, CALCULATION, AND ERROR PROPAGATION FOR THE DETERMINATION OF FISSION PRODUCT STRONTIUM

R. M. Abernathey, L. W. Buttars, R. E. Foster, Jr., M. E. Kussy,
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PREPARATION OF Sr-87 SPIKE

A 3.3-mg sample of isotopically enriched Sr-87, obtained from ORNL as the nitrate, was dissolved in quartz-distilled 1M HCl and diluted to 1 liter with 1M HCl. Weighed 3-ml aliquots were transferred to glass ampoules which were flame sealed.

CALIBRATION OF Sr-87 SPIKE

The calibration technique was isotope dilution mass spectrometry with natural strontium as the primary standard. The natural strontium was standardized by EDTA titration of "Specpure" strontium nitrate in a 50% ethanolic medium using metalphthalein as the indicator. The determined concentration was 1.007 mg Sr/g solution with a relative standard deviation of 0.10%. A weight aliquot of this standard solution was diluted 1000 fold, by weight, with 1M HCl and weight aliquots containing 3 mg of strontium were transferred to glass ampoules which were flame sealed.

To standardize the Sr-87 spike, the contents of individual ampoules of the natural strontium standard were mixed with the contents of individual ampoules of Sr-87 "spike". These mixtures were then analyzed by isotope dilution mass spectrometry. The determined concentration of the Sr-87 "spike" was 1.07×10^{16} atoms/g solution with a relative standard deviation for the average of four determinations of 0.23%.

CHEMICAL SEPARATION

1. Accurately pipet a 1-ml aliquot of the dissolver solution into an ampoule of Sr-87 "spike".
The solution is mixed by repeated fillings and discharging of the transfer pipet.
2. Transfer 2 ml of the above solution to a 50-ml polypropylene tube.
3. Add 100 μ l of $\text{Pb}(\text{NO}_3)_2$ (30 mg/ml) solution and 30 ml of fuming HNO_3 .
4. Cool in an ice bath for approximately 30 min and centrifuge. Discard the supernatant solution.
Caution, the supernatant solution is fuming HNO_3 .
5. Dissolve the precipitate in two drops of quartz-distilled water.
6. Add 1 ml of 1.5M HCl and pass the solution through a small Dowex 1x8 (100 to 200 mesh) column.
Fill a 2-ml polyethylene medicine dropper with approximately 1 in. of Dowex 1x8 resin. Pretreat the column with 1 ml of 1.5M HCl.

7. Wash the column with 15 drops of 1.5M HCl.
8. Collect the sample and washings in a 3-ml polypropylene centrifuge tube.
9. Evaporate to dryness using a filtered air flow and heat lamp.

MASS SPECTROMETRIC ANALYSIS

1. Prepare a canoe-shaped filament from high purity, 1-x 30-mil rhenium ribbon, and vacuum bake it for 1 hr at 4A at a pressure of 1×10^{-6} torr. Assemble the sample filament in the center (ionizing) position. Most of the trace rubidium and strontium impurities are volatilized.
2. Using a new, freshly cleaned, disposable glass pipet, add a few drops of water to the residue from step 9 (previous section). Dissolve the precipitate and transfer the solution to the filament. Evaporate the solution by passing 1.2A through the filament.
3. Mount the filament holder on the ion source carriage and insert the carriage into the vacuum lock. Allow about 1-min pumping time at each lock stage.
4. Adjust the filament current to 2.0 to 2.4A and locate the major isotope beam by reference to the mass-vs-DVM (accelerating voltage) curve for the magnetic field in use. Adjust the source voltage divider controls for the maximum beam. Make a rapid voltage scan in both directions to verify the mass identification. Use the electron multiplier. Keep the J2 setting low, the J10 setting maximum, and the J4-J5 setting near center. The source may need to be rotated to get J4-J5 centered with the maximum beam.
5. Insert the Faraday Cage collector and increase the filament current to obtain an ion current of 3×10^{-11} A. Scan the spectrum magnetically in both directions, recording the sensitivity settings used to give near full-

scale peak heights. Obtain three charts of 4-1/2 double scans.

6. Graphically interpolate by drawing lines on the chart which connect the tops of the successive peaks of each isotope. Draw vertical lines on the chart at each turnaround in the scans, and take peak height readings at the intersections of these lines with those connecting the peak tops. Sum the readings for each isotope.

Use the same number of readings in each sum. If the atom percent of an individual isotope is desired, it is given by:

$$\text{Atom \% Sr}^A = 100 \frac{\sum \text{peak heights}^A}{\sum (\sum \text{peak heights})}$$

CALCULATIONS

Using the spiked fission product samples, define:

$R_{sf}^{0/6}$ = atom ratio of Sr-90 to Sr-86 in the spiked fission product sample

$R_{sf}^{0/7}$ = atom ratio of Sr-90 to Sr-87 in the spiked fission product sample

$R_s^{6/7}$ = atom ratio of Sr-86 to Sr-87 in the "spike"

$R_n^{6/7}$ = atom ratio of Sr-86 to Sr-87 in natural strontium

N_f^0 = number of atoms of fission product Sr-90 in the sample

N_s^7 = number of atoms of "spike" Sr-87 added to the sample

N_n^7 = number of atoms of natural Sr-87 in the sample

V_d = volume of the dissolver solution

V_x = volume of sample aliquot taken for analysis

and

$$R_{sf}^{0/6} = \frac{N_f^0}{R_s^{6/7} N_s^7 + R_n^{6/7} N_n^7} \quad (1)$$

It is assumed here that the numbers of fission product Sr-86 and Sr-87 atoms are negligible. For the samples investigated, the number of fission product Sr-86 atoms is estimated to be less than 0.05% of the total Sr-86 in the samples.

Also:

$$R_{sf}^{0/7} = \frac{N_f^0}{N_s^7 + N_n^7}$$

or

$$N_n^7 = \frac{N_f^0 - R_{sf}^{0/7} N_s^7}{R_{sf}^{0/7}} \quad (2)$$

Substituting for N_n^7 in equation (1) and solving for N_f^0 yields:

$$N_f^0 = \frac{N_s^7 R_{sf}^{0/6} [R_s^{6/7} - R_n^{6/7}]}{\left[1 - \frac{R_{sf}^{0/6} R_n^{6/7}}{R_{sf}^{0/7}} \right]} \quad (3)$$

and for a sample aliquot from a dissolver solution.

$$N_f^0 = \frac{N_s^7 R_{sf}^{0/6} [R_s^{6/7} - R_n^{6/7}] V_d}{\left[1 - \frac{R_{sf}^{0/6} R_n^{6/7}}{R_{sf}^{0/7}} \right] V_x} \quad (4)$$

To calculate the atoms of Sr-88 in the spiked fission product samples define:

$R_{sf}^{8/7}$ = atom ratio of Sr-88 to Sr-87 in the "spiked" fission product sample

$R_n^{8/7}$ = atom ratio of Sr-88 to Sr-87 in natural strontium

$R_s^{8/7}$ = atom ratio of Sr-88 to Sr-87 in the "spike"

N_f^8 = atoms of fission product Sr-88 in the sample

and

$$R_{sf}^{8/7} = \frac{N_f^8 + R_n^{8/7} N_n^7 + R_s^{8/7} N_s^7}{N_s^7 + N_n^7} \quad (5)$$

Substituting equation (2) for N_n^7 and solving for N_f^8 yields:

$$N_f^8 = N_s^7 [R_n^{8/7} - R_s^{8/7}] + \frac{N_f^0}{R_{sf}^{0/7}} [R_{sf}^{8/7} - R_n^{8/7}] \quad (6)$$

and for a sample aliquot from a dissolver solution.

$$N_f^8 = \frac{V_d}{V_x} \left\{ N_s^7 [R_n^{8/7} - R_s^{8/7}] + \frac{N_f^0}{R_{sf}^{0/7}} [R_{sf}^{8/7} - R_n^{8/7}] \right\} \quad (7)$$

ERROR PROPAGATION FOR Sr-88 AND Sr-90

The number of fission product Sr-90 atoms, by equation (4) of the previous section is:

$$N_f^0 = \frac{N_s^7 R_{sf}^{0/6} [R_s^{6/7} - R_n^{6/7}] V_d}{\left[1 - \frac{R_{sf}^{0/6} R_n^{6/7}}{R_{sf}^{0/7}} \right] V_x}$$

Reported values are the average of several determinations:

$$\bar{N}_f^0 = \frac{\sum_{i=1}^n N_{fi}^0}{n}$$

where

$\bar{N}_f^0$ = average number of Sr-90 atoms

N_{fi}^0 = atoms of Sr-90 in the i^{th} sample.

n = number of determinations

V_d and usually V_x are the same for all samples and

$$\bar{N}_f^0 = \frac{V_d}{nV_x} \sum_{i=1}^n \left(\frac{N_{si}^7 R_{sfi}^{0/6} [R_s^{6/7} - R_n^{6/7}]}{\left[1 - \frac{R_{sfi}^{0/6} R_n^{6/7}}{R_{sfi}^{0/7}} \right]} \right)$$

The variance of $\bar{N}_f^0$ is then given approximately by:

$$\begin{aligned}
\sigma^2 \bar{N}_f^0 &= \left(\frac{\partial \bar{N}_f^0}{\partial V_d} \right)^2 \sigma^2 V_d + \left(\frac{\partial \bar{N}_f^0}{\partial V_x} \right)^2 \sigma^2 V_x + \left(\frac{\partial \bar{N}_f^0}{\partial R_s^{6/7}} \right)^2 \sigma^2 R_s^{6/7} \\
&+ \left(\frac{\partial \bar{N}_f^0}{\partial R_n^{6/7}} \right)^2 \sigma^2 R_n^{6/7} + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^0}{\partial R_{sf_i}^{0/6}} \right)^2 \sigma^2 R_{sf_i}^{0/6} + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^0}{\partial R_{sf_i}^{0/7}} \right)^2 \sigma^2 R_{sf_i}^{0/7} \\
&+ \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^0}{\partial N_{s_i}^7} \right)^2 \sigma^2 N_{s_i}^7 + 2 \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{\partial \bar{N}_f^0}{\partial N_{s_i}^7} \right) \left(\frac{\partial \bar{N}_f^0}{\partial N_{s_j}^7} \right) \sigma^2 N_{s_i s_j}^7 \\
&+ (\text{additional covariance terms})
\end{aligned}$$

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

$$\begin{aligned}
\frac{\sigma^2 N_{s_{ij}}^7}{\sigma N_{s_i}^7 \sigma N_{s_j}^7} &= 1 \\
\text{and} \\
\sigma^2 \bar{N}_f^0 &= \frac{1}{n^2 V_x^2} \left\{ V_d^2 \left[\frac{1}{V_x^2} \left(\sum_{i=1}^n \left(\frac{R_{sf_i}^{0/6} N_{s_i}^7 [R_s^{6/7} - R_n^{6/7}]}{1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}}} \right)^2 \right) \sigma^2 V_x \right. \right. \\
&\quad \left. \left. + \left(\sum_{i=1}^n \left(\frac{R_{sf_i}^{0/6} N_{s_i}^7}{1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}}} \right)^2 \right) \sigma^2 R_s^{6/7} \right] \right\}
\end{aligned}$$

$$\begin{aligned}
& + \left(\sum_{i=1}^n \frac{R_{sf_i}^{0/6} N_{s_i}^7 \left[\frac{R_s^{6/7} R_{sf_i}^{0/6}}{R_{sf_i}^{0/7}} - 1 \right]}{\left[1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}} \right]^2} \right)^2 \sigma^2 R_n^{6/7} \\
& + \sum_{i=1}^n \left(\frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}]}{\left[1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}} \right]^2} \right)^2 \sigma^2 N_{s_i}^7 \\
& + \sum_{i=1}^n \left(\frac{N_{s_i}^7 [R_s^{6/7} - R_n^{6/7}]}{\left[1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}} \right]^2} \right)^2 \sigma^2 R_{sf_i}^{0/6} \\
& + \sum_{i=1}^n \left(\frac{- \left(\frac{R_{sf_i}^{0/6}}{R_{sf_i}^{0/7}} \right)^2 N_{s_i}^7 R_n^{6/7} \left[- R_n^{6/7} + R_s^{6/7} \right]}{\left[1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}} \right]^2} \right)^2 \sigma^2 R_{sf_i}^{0/7} \\
& + \left(\sum_{i=1}^n \left(\frac{R_{sf_i}^{0/6} N_{s_i}^7 [R_s^{6/7} - R_n^{6/7}]}{\left[1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}} \right]^2} \right)^2 \sigma^2 V_d \right)
\end{aligned}$$

$$+ 2 \frac{V_d^2}{n^2 V_x^2} \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}]}{1 - \frac{R_{sf_i}^{0/6} R_n^{6/7}}{R_{sf_i}^{0/7}}} \right) \left(\frac{R_{sf_j}^{0/6} [R_s^{6/7} - R_n^{6/7}]}{1 - \frac{R_{sf_j}^{0/6} R_n^{6/7}}{R_{sf_j}^{0/7}}} \right) \sigma N_{s_i}^7 \sigma N_{s_j}^7$$

The number of Sr-88 atoms, by equation (7) of the previous section is:

$$N_f^8 = \frac{V_d}{V_x} \left\{ N_s^7 [R_n^{8/7} - R_s^{8/7}] + \frac{N_f^0}{R_{sf}^{0/7}} [R_{sf}^{8/7} - R_n^{8/7}] \right\}$$

Substituting for N_f^0 from equation (3) of the previous section yields:

$$N_f^8 = \frac{V_d}{V_x} \left(N_s^7 [R_n^{8/7} - R_s^{8/7}] + \frac{R_{sf}^{0/6} N_s^7 [R_s^{6/7} - R_n^{6/7}] [R_{sf}^{8/7} - R_n^{8/7}]}{[R_{sf}^{0/7} - R_{sf}^{0/6} R_n^{6/7}]} \right)$$

Reported values are the average of several determinations:

$$\bar{N}_f^8 = \frac{V_d}{n V_x} \sum_{i=1}^n \left[N_{s_i}^7 \left([R_n^{8/7} - R_s^{8/7}] + \frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}] [R_{sf_i}^{8/7} - R_n^{8/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]} \right) \right]$$

The variance of $\bar{N}_f^8$ is then given approximately by:

$$\begin{aligned} \sigma^2 \bar{N}_f^8 &= \left(\frac{\partial \bar{N}_f^8}{\partial R_n^{8/7}} \right)^2 \sigma^2 R_n^{8/7} + \left(\frac{\partial \bar{N}_f^8}{\partial R_s^{8/7}} \right)^2 \sigma^2 R_s^{8/7} + \left(\frac{\partial \bar{N}_f^8}{\partial R_s^{6/7}} \right)^2 \sigma^2 R_s^{6/7} \\ &+ \left(\frac{\partial \bar{N}_f^8}{\partial R_n^{6/7}} \right)^2 \sigma^2 R_n^{6/7} + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^8}{\partial N_{s_i}^7} \right)^2 \sigma^2 N_{s_i}^7 + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^8}{\partial R_{sf_i}^{0/6}} \right)^2 \sigma^2 R_{sf_i}^{0/6} \\ &+ \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^8}{\partial R_{sf_i}^{8/7}} \right)^2 \sigma^2 R_{sf_i}^{8/7} + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^8}{\partial R_{sf_i}^{0/7}} \right)^2 \sigma^2 R_{sf_i}^{0/7} + \left(\frac{\partial \bar{N}_f^8}{\partial V_x} \right)^2 \sigma^2 V_x \\ &+ \left(\frac{\partial \bar{N}_f^8}{\partial V_d} \right)^2 \sigma^2 V_d + 2 \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{\partial \bar{N}_f^8}{\partial N_{s_i}^7} \right) \left(\frac{\partial \bar{N}_f^8}{\partial N_{s_j}^7} \right) \sigma^2 N_{s_{ij}}^7 \end{aligned}$$

+ (additional covariance terms)

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

$$\frac{\sigma^2 N_{s_{ij}}^7}{\sigma N_{s_i}^7 \sigma N_{s_j}^7} = 1$$

and

$$\begin{aligned} \sigma^2 \bar{N}_f^8 = & \frac{1}{n^2 V_x^2} \left\{ V_d^2 \left[\frac{1}{V_x^2} \left(\sum_{i=1}^n \left[N_{s_i}^7 \left(R_n^{8/7} - R_s^{8/7} \right) \right. \right. \right. \right. \\ & + \left. \left. \left. \frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}] [R_{sf_i}^{8/7} - R_n^{8/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]} \right] \right) \right]^2 \sigma^2 V_x \\ & + \left(\sum_{i=1}^n \left[N_{s_i}^7 \left(1 - \frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]} \right) \right]^2 \sigma^2 R_n^{8/7} + \left(\sum_{i=1}^n N_{s_i}^7 \right)^2 \sigma^2 R_s^{8/7} \right. \\ & + \left(\sum_{i=1}^n N_{s_i}^7 \left(\frac{R_{sf_i}^{0/6} [R_{sf_i}^{8/7} - R_n^{8/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]} \right) \right]^2 \sigma^2 R_s^{6/7} \\ & + \left(\sum_{i=1}^n N_{s_i}^7 \left(\frac{R_{sf_i}^{0/6} [R_{sf_i}^{0/6} R_s^{6/7} - R_{sf_i}^{0/7}] [R_{sf_i}^{8/7} - R_n^{8/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]^2} \right) \right]^2 \sigma^2 R_n^{6/7} \\ & + \sum_{i=1}^n \left| \left[R_n^{8/7} - R_s^{8/7} \right] + \frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}] [R_{sf_i}^{8/7} - R_n^{8/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]} \right|^2 \sigma^2 N_{s_i}^7 \\ & + \sum_{i=1}^n \left| N_{s_i}^7 \left(\frac{[R_{sf_i}^{8/7} - R_n^{8/7}] R_{sf_i}^{0/7} [R_s^{6/7} - R_n^{6/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]^2} \right) \right|^2 \sigma^2 R_{sf_i}^{0/6} \\ & + \sum_{i=1}^n \left| N_{s_i}^7 \left(\frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]} \right) \right|^2 \sigma^2 R_{sf_i}^{8/7} \end{aligned}$$

$$\begin{aligned}
& + \sum_{i=1}^n \left| N_{s_i}^7 \left(\frac{-R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}] [R_{sf_i}^{8/7} - R_n^{8/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]^2} \right) \right|^2 \sigma^2 R_{sf_i}^{0/7} \\
& + \left\{ \sum_{i=1}^n N_{s_i}^7 \left| \left[R_n^{8/7} - R_s^{8/7} \right] + \frac{R_{sf_i}^{0/6} [R_s^{6/7} - R_n^{6/7}] [R_{sf_i}^{8/7} - R_n^{8/7}]}{[R_{sf_i}^{0/7} - R_{sf_i}^{0/6} R_n^{6/7}]} \right|^2 \sigma^2 V_d \right\} \\
& + 2 \frac{V_d^2}{n^2 V_x^2} \sum_{i=1}^{n-1} \sum_{j>i}^n Q_i Q_j \alpha N_{s_i}^7 \sigma N_{s_j}^7
\end{aligned}$$

where:

$$Q = \frac{[R_n^{8/7} - R_s^{8/7}] + R_{sf}^{0/6} [R_s^{6/7} - R_n^{6/7}] [R_{sf}^{8/7} - R_n^{8/7}]}{[R_{sf}^{0/7} - R_{sf}^{0/6} R_n^{6/7}]}$$

The variances $\sigma^2 R_{sf_i}^{0/6}$ and $\sigma^2 R_{sf_i}^{0/7}$ were obtained from multiple scans on individual spiked samples. The variances $\sigma^2 V_d$, $\sigma^2 V_x$, $\sigma^2 R_s^{6/7}$, $\sigma^2 R_n$ and $\sigma^2 N_{s_i}^7$ were obtained from independent measurements.

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APPENDIX III

PROCEDURE FOR THE SEPARATION AND MASS SPECTROMETRIC ANALYSIS OF FISSION PRODUCT BARIUM

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7. Wash the column with 15 drops of 1.5M HCl.
8. Collect the sample and washings in a 3-ml polypropylene centrifuge tube.
9. Evaporate to dryness using a filtered air flow and heat lamp.

MASS SPECTROMETRIC ANALYSIS

1. Prepare a canoe-shaped filament from high purity, 1-x30-mil tantalum ribbon and assemble in the normal configuration.
2. Dissolve the residue from step 9 (previous section) in quartz-distilled 1M HNO₃ and using a new, freshly cleaned, disposable pipette, transfer the solution to the filament. Evaporate the solution by passing 1.5 A through the filament.
3. Mount the filament holder on the ion source carriage and insert the carriage into the vacuum lock. Allow about 5-min pumping time at the intermediate stage.
4. Adjust the filament current to 1.4 A and locate the Cs-133 beam by reference to the accelerating voltage, magnet current, and magnet shunt position. Adjust the focus voltage for the maximum beam. Increase the filament current slowly to 1.9 A and "bake" until the Cs-133 peak is down to an insignificant level. The cesium as first observed is fission product as indicated by the 133, 135 and 137 peaks. In some cases the ion current was greater than 2×10^{-11} A (the maximum detectable on the instrument). From 10 min to several hours may be required for "bake out" of the cesium.
5. Increase the filament current to 2.2 A to obtain an ion current of 5×10^{-12} A. Scan the spectrum magnetically in both directions, recording the sensitivity settings used to give near full-scale peak heights. Obtain three charts of 3-1/2 double scans. After the barium beam increased, it was not evident whether the cesium present was entirely fission product or a combination of fission product and natural cesium. The Cs-133 was "baked" out until its intensity was 1% or less of the Ba-135 peak.

PREPARATION OF Ba-135 SPIKE

A 1.9-mg sample of isotopically enriched Ba-135, obtained from ORNL as the nitrate, was dissolved in quartz-distilled 1M HCl and diluted to 1 liter with quartz distilled 1M HCl. Weighed 3-ml aliquots were transferred to quartz ampoules which were flame sealed.

CALIBRATION OF Ba-135 SPIKE

The calibration technique was isotope dilution mass spectrometry with natural barium as the primary standard. Approximately 0.4 g of "Specpure" barium carbonate was ignited at 800°C for 1 hr, dissolved in quartz-distilled 1M HCl and evaporated to dryness. The resulting barium chloride sample was dried at approximately 275°C for 1 hr and weighed. The sample was dissolved in 1M HCl and diluted with 1M HCl to approximately 220 ml. Weight aliquots were taken and the barium concentration was determined by EDTA titrimetry. The determined concentration was 1.030 mg Ba/g solution with a relative standard deviation of 0.10%. A weight aliquot of this standard solution was diluted 1000 fold, by weight, with 1M HCl and weight aliquots of this solution containing approximately 3 µg of Ba were mixed with weight aliquots of the Ba-135 spike. The mixtures were analyzed by isotope dilution mass spectrometry. The determined Ba-135 concentration was 5.9074×10^{15} atoms/g solution with a relative standard deviation for the average of 0.34%.

CHEMICAL SEPARATION

1. Accurately pipet a 1-ml aliquot of the dissolver solution into an ampoule of Ba-135 "spike".
The solution is mixed by repeated fillings and dischargings of the transfer pipet.
2. Transfer 2 ml of the above solution to a 50-ml polypropylene centrifuge tube.
3. Add 100 µl of $\text{Pb}(\text{NO}_3)_2$ (20 mg/ml) solution and 30 ml of fuming HNO_3 .
4. Cool in an ice bath for approximately 30 min and centrifuge. Discard the supernatant.
Caution: The supernatant solution is essentially fuming HNO_3 .
5. Dissolve the precipitate in two drops of quartz-distilled water.
6. Add 1 ml of 1.5M HCl and pass the solution through a small Dowex 1 x 8 (100 to 200 mesh) column.
Fill a 2 ml polyethylene medicine dropper with approximately 1 in. of Dowex 1 x 8 resin. Pretreat the column with 1 ml of 1.5M HCl.

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

Use the same number of readings in each sum. If the atom percent of an individual isotope is desired, it is given by:

$$\text{Atom\% Ba}^A = 100 \frac{\sum \text{peak heights A}}{\sum (\sum \text{peak heights})}$$

CALCULATIONS

Using the spiked fission product samples define:

$R_{sf}^{8/5}$ = atom ratio of Ba-138 to Ba-135 in the spiked fission product sample

$R_{sf}^{8/6}$ = atom ratio of Ba-138 to Ba-136 in the spiked fission product sample

$R_s^{8/5}$ = atom ratio of Ba-138 to Ba-135 in the "spike"

$R_s^{6/5}$ = atom ratio of Ba-136 to Ba-135 in the "spike"

$R_n^{5/8}$ = atom ratio of Ba-135 to Ba-138 in natural barium

$R_n^{6/8}$ = atom ratio of Ba-136 to Ba-138 in natural barium

N_s^5 = number of atoms of "spike" Ba-135 in the sample

N_f^8 = number of atoms of fission product Ba-138 in the sample

N_n^8 = number of atoms of natural Ba-138 in the sample

N_s^8 = number of atoms of "spike" Ba-138 in the sample

N_n^5 = number of atoms of natural Ba-135 in the sample

V_d = volume of dissolver solution

V_x = volume of sample aliquot taken for analysis

and

$$R_{sf}^{8/5} = \frac{N_f^8 + N_n^8 + R_s^{8/5} N_s^5}{N_s^5 + R_n^{5/8} N_n^8} \quad (1)$$

and

$$R_{sf}^{8/6} = \frac{N_f^8 + N_n^8 + R_s^{8/5} N_s^5}{R_n^{6/8} N_n^8 + R_s^{6/5} N_s^5} \quad (2)$$

solving equation (2) for N_n^8 yields:

$$N_n^8 = \frac{N_f^8 + N_s^5 \left[R_s^{8/5} - R_{sf}^{8/6} R_s^{6/5} \right]}{\left[R_{sf}^{8/6} R_n^{6/8} - 1 \right]} \quad (3)$$

substituting for N_n^8 in equation (1) and solving for N_f^8 in a sample aliquot from a dissolver solution yields:

$$N_f^8 = \frac{N_s^{5V_d} \left(\frac{\left[R_{sf}^{8/5} R_n^{5/8} - 1 \right] \left[R_s^{8/5} - R_{sf}^{8/6} R_s^{6/5} \right]}{\left[R_{sf}^{8/6} R_n^{6/8} - 1 \right]} + R_{sf}^{8/5} - R_s^{8/5} \right)}{V_x \left(1 - \frac{\left[R_{sf}^{8/5} R_n^{5/8} - 1 \right]}{\left[R_{sf}^{8/6} R_n^{6/8} - 1 \right]} \right)} \quad (4)$$

ERROR PROPAGATION FOR Ba-138

Reported values are the average of several determinations:

$$N_f^8 = \frac{\sum_{i=1}^n N_{fi}^8}{n}$$

where:

$\bar{N}_f^8$ = average number of Ba 138 atoms

N_{fi}^8 = atoms of Ba-138 in the i^{th} sample

n = number of determinations

The number of Ba-138 (N_{fi}^8) atoms in an individual sample is given by equation (4) of the previous section. Since V_d and usually V_x are the same for all samples, $\bar{N}_f$ is:

$$\bar{N}_f^8 = \frac{V_d}{nV_x} \sum_{i=1}^n \frac{N_{s_i}^5 \left(\frac{[R_{sf_i n}^{8/5} R_n^{5/8} - 1][R_s^{8/5} - R_{sf_i s}^{8/6} R_s^{6/5}]}{[R_{sf_i n}^{8/6} R_n^{6/8} - 1]} + R_{sf_i}^{8/5} - R_s^{8/5} \right)}{\left(1 - \frac{[R_{sf_i n}^{8/5} R_n^{5/8} - 1]}{[R_{sf_i n}^{8/6} R_n^{6/8} - 1]} \right)}$$

The variance of $\bar{N}_f^8$ is given approximately by:

$$\begin{aligned} \sigma^2 \bar{N}_f^8 &= \left(\frac{\partial \bar{N}_f^8}{\partial V_d} \right)^2 \sigma^2 V_d + \left(\frac{\partial \bar{N}_f^8}{\partial V_x} \right)^2 \sigma^2 V_x + \left(\frac{\partial \bar{N}_f^8}{\partial R_s^{8/5}} \right)^2 \sigma^2 R_s^{8/5} \\ &+ \left(\frac{\partial \bar{N}_f^8}{\partial R_n^{5/8}} \right)^2 \sigma^2 R_n^{5/8} + \left(\frac{\partial \bar{N}_f^8}{\partial R_s^{6/5}} \right)^2 \sigma^2 R_s^{6/5} + \left(\frac{\partial \bar{N}_f^8}{\partial R_n^{6/8}} \right)^2 \sigma^2 R_n^{6/8} \\ &+ \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^8}{\partial N_{s_i}^5} \right)^2 \sigma^2 N_{s_i}^5 + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^8}{\partial R_{sf_i}^{8/5}} \right)^2 \sigma^2 R_{sf_i}^{8/5} + \sum_{i=1}^n \left(\frac{\partial \bar{N}_f^8}{\partial R_{sf_i}^{8/6}} \right)^2 \sigma^2 R_{sf_i}^{8/6} \\ &+ 2 \sum_{i=1}^{n-1} \sum_{j>i}^n \left(\frac{\partial \bar{N}_f^8}{\partial N_{s_i}^5} \right) \left(\frac{\partial \bar{N}_f^8}{\partial N_{s_j}^5} \right) \sigma^2 N_{s_{ij}}^5 + (\text{additional covariance terms}) \end{aligned}$$

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

$$\frac{\sigma^2 N_{s_{ij}}^5}{\sigma N_{s_i}^5 \sigma N_{s_j}^5} = 1$$

Let

$$R_{sf_i n}^{8/5} R_n^{5/8} - 1 = Y_i$$

$$R_{sf_i n}^{8/6} R_n^{6/8} - 1 = X_i$$

$$\left[\begin{array}{c} 1 - \frac{[R_{sf_i n}^{8/5} R_n^{5/8} - 1]}{[R_{sf_i n}^{8/6} R_n^{6/8} - 1]} \end{array} \right] = Z_i$$

and

$$\sigma^2 \bar{N}_f^8 = \frac{1}{n^2 V_x^2} \left\{ V_d^2 \right.$$

$$\frac{1}{V_x^2} \left(- \sum_{i=1}^n N_{s_i}^5 \frac{\left(\frac{Y_i [R_s^{8/5} - R_{sf_i}^{8/6} R_s^{6/5}]}{X_i} + R_{sf_i}^{8/5} - R_s^{8/5} \right)}{Z_i} \right)^2 \sigma^2 V_x$$

$$+ \left(\sum_{i=1}^n - N_{s_i}^5 \right)^2 \sigma^2 R_s^{8/5}$$

$$+ \left(\sum_{i=1}^n - \left(\frac{N_{s_i}^5 R_{sf_i}^{8/5} [R_{sf_i}^{8/6} R_s^{6/5} - R_{sf_i}^{8/5}]}{X_i Z_i^2} \right) \right)^2 \sigma^2 R_n^{5/8}$$

$$+ \left(\sum_{i=1}^n \frac{- N_{s_i}^5 R_{sf_i}^{8/6} Y_i}{X_i Z_i} \right)^2 \sigma^2 R_s^{6/5}$$

$$\begin{aligned}
& + \left(\sum_{i=1}^n \frac{\frac{R_{sf_i}^{8/6} N_{s_i}^5 Y_i [R_{sf_i}^{8/6} R_s^{6/5} - R_{sf_i}^{8/5}]}{X_i^2}}{Z_i^2} \right)^2 \sigma_{R_n}^{2R^{6/8}} \\
& + \sum_{i=1}^n \left(\frac{\frac{Y_i [R_s^{8/5} - R_{sf_i}^{8/6} R_s^{6/5}]}{X_i} + R_{sf_i}^{8/5} - R_s^{8/5}}{Z_i} \right)^2 \sigma_{N_{s_i}}^{2N_{s_i}^5} \\
& + \sum_{i=1}^n \left(\frac{\frac{N_{s_i}^5 R_{sf_i}^{8/6} [R_n^{6/8} - R_s^{6/5} R_n^{5/8}]}{X_i}}{Z_i^2} \right)^2 \sigma_{R_{sf_i}}^{2R_{sf_i}^{8/5}} \\
& + \sum_{i=1}^n \left(\frac{\frac{N_{s_i}^5 Y_i}{X_i^2 Z_i^2} \left[R_{sf_i}^{8/5} (R_s^{6/5} R_n^{5/8} - R_n^{6/8}) \right]}{Z_i} \right)^2 \sigma_{R_{sf_i}}^{2R_{sf_i}^{8/6}} \\
& + \left(\sum_{i=1}^n \frac{N_{s_i}^5 \left[\frac{Y_i [R_s^{8/5} - R_{sf_i}^{8/6} R_s^{6/5}]}{X_i} + R_{sf_i}^{8/5} - R_s^{8/5} \right]}{Z_i} \right)^2 \sigma_{V_d}^{2V_d} \Bigg\} \\
& + 2 \frac{V_d^2}{n^2 V_x^2} \sum_{i=1}^{n-1} \sum_{j>i}^n Q_i Q_j \sigma_{N_{s_i}}^5 \sigma_{N_{s_j}}^5
\end{aligned}$$

where

$$Q = \left(\frac{\frac{Y [R_s^{8/5} - R_{sf}^{8/6} R_s^{6/5}]}{X} + R_{sf}^{8/5} - R_s^{8/5}}{Z} \right)$$

The variances $\sigma^2_{sf_i} R^{8/5}$ and $\sigma^2_{sf_i} R^{8/6}$ were obtained from multiple scans on individual samples. The variances $\sigma^2 V_d$, $\sigma^2 V_x$, $\sigma^2 R_s^{8/5}$, $\sigma^2 R_n^{5/8}$, $\sigma^2 R_n^{6/5}$, $\sigma^2 R_n^{6/8}$ and $\sigma^2 N_{s_i}^5$ were obtained from independent measurements.