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BRANCHING FRACTION IN THE
RADIOACTIVE DECAY OF ^{85m}Kr

by

A. H. Jaffey, E. P. Steinberg, J. E. Gindler,
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February 1980

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TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	7
I. INTRODUCTION	7
II. IRRADIATION	9
III. DISSOLUTION	9
IV. THE MASS-SPECTROMETER SPIKE SOLUTION	10
V. PREPARATION OF RUBIDIUM-85 FOR MASS-SPECTROMETRIC MEASUREMENTS.	13
A. Transfer of Dissolver Solution.	13
B. Contamination Problems.	13
C. Purification of Rubidium Sample for Mass-spectrometric Analysis	14
VI. MASS SPECTROMETRY	15
A. General	15
B. Analyzed Samples	15
C. Effect of Sample Purity on the Source	16
D. Equations Used in Mass-spectrometry Isotope-dilution Analysis	16
E. Bias Correction	17
VII. PREPARATION OF KRYPTON FOR COUNTING.	18
A. Purification	18
B. Dilution	18
VIII. KRYPTON-85 COUNTING	18
A. GM Counters	18
B. Absolute Counting	19

TABLE OF CONTENTS

	<u>Page</u>
IX. MASS-SPECTROMETRY RESULTS	22
A. Low-burnup Sample	22
B. High-burnup Sample	23
C. Evaluation of Rubidium Contamination in Original Capsules .	24
D. Net ^{85}Rb from Dissolver Solutions	25
X. RESULTS OF KRYPTON-85 ANALYSIS	25
A. General	25
B. Low-burnup Sample	26
C. High-burnup Sample	29
XI. BRANCHING-FRACTION RESULTS	32
XII. BURNUP EFFECTS IN HIGH-BURNUP EXPERIMENT	33
A. Burnup of ^{85}Rb and ^{85}Kr	33
D. Measurement of ^{235}U and Its Original Mass	34
XIII. OTHER MEASUREMENTS OF THE BRANCHING FRACTION ...	36
A. From Gamma-ray Intensity Measurements	36
B. From Mass-spectrometric Determination of ^{85}Kr and ^{85}Rb ..	37
C. Present Work: From Mass-spectrometric Determination of ^{85}Rb and Absolute Gas Counting of ^{85}Kr	37
ACKNOWLEDGMENTS	38
REFERENCES	38

FIGURE

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	End of Mass 85 Fission Chain	8

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Irradiation and Waiting-time History of the Samples.	9
II.	Low-burnup Isotope-dilution Results	22
III.	High-burnup Isotope-dilution Results	23
IV.	Concentration of ^{85}Rb in the Blank Solutions	24
V.	Net ^{85}Rb Yield	25
VI.	Low-burnup Sample: Dilution Factors and Counting Results for ^{85}Kr	26
VII.	Low-burnup Sample: ^{85}Kr Yield from Irradiated Sample	27
VIII.	High-burnup Sample: Dilution Factors and Counting Results for ^{85}Kr	29
IX.	High-burnup Sample: ^{85}Kr Yield from Irradiated Sample	30
X.	Calculation of Branching Fraction	33
XI.	Summary of Branching-fraction Measurements	38

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ABSTRACT

A measurement has been made of the branching fraction in the decay of the fission product ^{85m}Kr to the ground state ^{85}Kr (10.75 yr) relative to the total decay to both ^{85}Kr and ^{85}Rb . Samples of ^{235}U were highly irradiated in a high-flux reactor and dissolved. The ^{85}Rb was measured by isotope-dilution mass spectrometry, and the ^{85}Kr was counted in GM tubes whose counting efficiencies were calibrated with a standardized ^{85}Kr gas of known disintegration rate. The branching fraction measured with both a low-burnup sample and a high-burnup sample was 0.2160 ± 0.0019 , the largest error arising from the uncertainty in the calibration of the standardized gas.

I. INTRODUCTION

In most fission chains, the yield of a particular mass chain is ascertainable, after a sufficient waiting period, through measurement of the number of atoms of the stable isobar at the end of the chain produced from a known number of fissioned nuclei. In some cases, the precursor of this stable isobar is so long-lived that awaiting its complete decay would require an inordinate amount of time. In such a situation, measurement of the stable isobar may still be of value in ascertaining the chain yield if the irradiation history and waiting period are known; correction can then be made to allow for the amount of precursor remaining undecayed. Such a situation arises in the mass-85 chain, but is further complicated by the fact that a portion of the chain decay proceeds "rapidly" through the β decay of ^{85m}Kr to the stable isobar ^{85}Rb and a part passes through the long-lived ^{85}Kr , which beta decays (10.7-yr half-life) to ^{85}Rb (see Fig. 1). To make the holdup correction, we must know not only the irradiation history and the waiting period, but also the branching fraction between the direct beta decay of ^{85m}Kr to ^{85}Rb and the internal transition of ^{85m}Kr to the ground state ^{85}Kr .

Fig. 1

End of Mass 85 Fission Chain

This fraction has previously been measured by determination of the intensity ratio of the 305- and 151-keV γ -ray transitions shown in Fig. 1. The intensity of the first γ ray provides information on the amount of $^{85\text{m}}\text{Kr}$ decaying to ^{85}Kr , and from the intensity of the latter, one can determine the amount of $^{85\text{m}}\text{Kr}$ decaying directly to ^{85}Rb . The branching fraction may be calculated after internal-conversion corrections are made to the γ -ray transition intensities. This method is limited in the accuracy with which the branching fraction can be measured because of limitations on the measurement accuracy of absolute γ -ray intensities and of conversion coefficients.

An alternative procedure is to measure the relative amounts of ^{85}Rb and ^{85}Kr formed in the neutron irradiation of a ^{235}U sample. Mass measurement by mass-spectrometric isotope dilution allows an accuracy that could greatly improve the error of the branching-fraction value over that attainable by γ -ray measurement. However, to form enough material for the mass-spectrometric measurement, a lengthy irradiation of a uranium sample is required. The amount required is actually set by the need to form enough ^{85}Rb to overshadow the natural contamination that could occur either in the original material or in the subsequent handling.

We report here on such a measurement, in which the ^{85}Rb was measured by mass-spectrometric isotope dilution and the ^{85}Kr was measured by absolute counting of its beta radioactivity with calibrated GM tubes. Other measurements have been made at Allied Chemical Corporation, Idaho, using isotope-dilution measurements for both ^{85}Rb and ^{85}Kr . Results of the branching-fraction measurements from the various experiments are compared in Sec. XIII.

II. IRRADIATION

Samples were irradiated in the Engineering Test Reactor at the National Reactor Testing Station near Idaho Falls, Idaho, at a flux exceeding 10^{13} n/cm².s. Samples formed in this irradiation were examined by ourselves and by William Maeck and colleagues at Allied Chemical Corp.

The samples consisted of aluminum capsules containing 99.74% ²³⁵U as uranium oxide, mixed with fine aluminum metal powder. Similar capsules containing only the same amount of aluminum powder were used for measuring blank corrections. The composition and irradiation history of the two samples used in the present study are shown in Table I.

TABLE I. Irradiation and Waiting-time History of the Samples

	Low Burnup	High Burnup
Pin No.	8	37
Mass ²³⁵ U irradiated, mg	832	36.48 ^a
Approximate flux (nv), n/cm ² .s	$\sim 2 \times 10^{13}$	$\sim 5 \times 10^{13}$
Time in reactor, days	20	321
Time of exposure to neutrons, days	20	179
Approximate fluence (nvt)	$\sim 4 \times 10^{19}$	$\sim 8 \times 10^{20}$
Burnup	0.0017	0.418 ^a
Waiting time from end of irradiation to time of krypton measurement, days	1308	1800.7
Waiting time from midtime of irradiation to end of dissolution, days	1238	1593

^aSee Sec. XII.B.

III. DISSOLUTION

The irradiated capsule was dissolved in nitric acid in the presence of a measured volume of relatively inactive krypton gas, which served as the carrier gas. This gas was pumped through the system for several days, ensuring that the radioactive ⁸⁵Kr was intimately mixed with its carrier. The end of dissolution time was defined by the time at which the krypton was removed from the dissolver solution, so that ⁸⁵Rb could no longer be added to the solution from the ⁸⁵Kr decay.

After a careful dissolution of our first capsule (No. 13), attempts to measure the ⁸⁵Kr activity by counting demonstrated that most of the ⁸⁵Kr formed had leaked out before dissolution. To save the labor of a fruitless complete dissolution, preliminary tests for capsule leakage were included before another capsule was used. For the first test, the capsule was placed in an evacuated tube of diluted mineral oil and a visual examination was made for gas bubbles. This was followed by a more sensitive radiometric test.

A new dissolution system was filled with dead krypton, which was circulated for several days and then counted in GM tubes; this background activity was zero. The irradiated capsule was then placed in the system, which was evacuated and kept closed off for five days, after which dead krypton was admitted and circulated for several days. When the counted gas showed no activity, the capsule was accepted as intact.

The complete dissolution was carried out in a shielded cave. The capsule was placed in a quartz vessel, which was part of a 6-L dissolution system. The system was evacuated, flushed with helium, and then filled with dead krypton at 120 Torr (16 kPa) pressure. Concentrated HNO_3 was added several times in small amounts, along with some mercury metal to catalyze dissolution of the aluminum metal. A quartz vessel was used to avoid the possibility of introducing natural rubidium by leaching from the container walls. Prior to use, the vessel was extensively cleaned with purified nitric acid and protected from contamination. The gas was pumped continuously through the closed system, during and after dissolution at $\sim 1/2$ atm (50 kPa) pressure. It was circulated for about 3 days, although dissolution was practically complete after 5 h. At the end of dissolution, the solution was briefly heated to aid in dissolving a slight residue (identified in other experiments as ruthenium precipitated in the acid-depleted solution). If some of the residue still remained, it was dissolved with added HCl. The gas was then removed as described in Sec. VII.A. The rubidium in the dissolver solution and the krypton gas were purified separately (see Secs. V and VII), and the ^{85}Rb and ^{85}Kr components were analyzed. The krypton gas was purified with a gas chromatographic technique, and aliquots were further diluted for counting in efficiency-calibrated GM counters. The rubidium was chemically separated from the dissolver solution with a procedure that purified it from other fission products, aluminum, and the added reagents. The ^{85}Rb content of the purified material was determined by mass-spectrometric isotope-dilution analysis.

IV. THE MASS-SPECTROMETER SPIKE SOLUTION

For isotope-dilution analysis, it is necessary to use a rubidium sample of known isotopic composition and of known weight. Two standard solutions were prepared; comparison showed the calibrations to be consistent to high accuracy.

The primary standardizations of these spike solutions involved determination of the number of ^{85}Rb and ^{87}Rb atoms present in each milliliter (or gram) of solution. This was carried out by weighing a compound of known composition and then diluting it to known volume (or weight), and by mass-spectrometric analysis of the $^{85}\text{Rb}/^{87}\text{Rb}$ atom ratio. Since the relative fission yield of the mass-85 chain in ^{235}U fission is only about one-half that of the mass-87 chain, isotope-dilution analysis for the 85 chain is more sensitively carried out using an enriched ^{85}Rb sample.

Other rubidium standards were also prepared. The concentration of National Bureau of Standards natural rubidium was determined in the same manner by weighing and dilution. It was used as a spike to cross-check (by isotope dilution analysis) the concentration of the ^{85}Rb spike. Its isotopic composition had been determined by the NBS to have the atom ratio $^{85}\text{Rb}/^{87}\text{Rb} = 2.593 (\pm 0.1\%)$. This material was also used to calibrate the bias of the mass spectrometer, as discussed in Sec. VI.E.

A sample of spike material highly enriched in ^{87}Rb was also prepared for use in detecting contamination in the blanks. Since contamination arose from natural Rb ($^{87}\text{Rb}/^{85}\text{Rb} = 1/2.593$), the highest sensitivity is achieved by use of a ^{87}Rb spike.

Four spike materials were prepared:

1. $(\text{SP-1})_{85}$, of enriched ^{85}Rb , and used for analyzing the low-burnup sample. ($^{85}\text{Rb}/^{87}\text{Rb} = 216.5$)
2. $(\text{SP-2})_{85}$, of similar material, used for the high-burnup sample. ($^{85}\text{Rb}/^{87}\text{Rb} = 615.2$)
3. SP_{Nat} , of natural rubidium. ($^{85}\text{Rb}/^{87}\text{Rb} = 2.593$)
4. SP_{87} , of enriched ^{87}Rb , used for analyzing the blank samples. ($^{87}\text{Rb}/^{85}\text{Rb} = 103.2$)

Several years had elapsed from the time of the $(\text{SP-1})_{85}$ characterization to the dissolution time of the high-burnup sample. Because there was some concern that the original $(\text{SP-1})_{85}$ calibration might no longer be valid, $(\text{SP-2})_{85}$ was prepared. However, cross-checks showed that the original calibration had not changed sensibly, and indeed the comparison served to substantiate the original calibration value.

The rubidium content of the spike solution was determined by quantitative chemical analysis. For $(\text{SP-1})_{85}$, an aliquot was evaporated in H_2SO_4 , forming Rb_2SO_4 , which was weighed. This was transformed to RbCl by separating the SO_4^{2-} ion through ion exchange. The weight of rubidium agreed in both. Another aliquot was prepared as the sulfate, with the results:

$$\begin{array}{rcl} \text{Rb}_2\text{SO}_4, \text{RbCl} & 0.5183 & \text{mg/mL} \\ \text{Rb}_2\text{SO}_4 & 0.5190 & \\ \hline \text{Av.} & 0.5186_5 & \text{mg/mL} \end{array}$$

For SP_{Nat} , a similar preparation of two aliquots as Rb_2SO_4 and RbCl gave the results:

$$\begin{array}{rcl} & 10.397 & \text{mg/g soln.} \\ & 10.396 & \text{mg/g soln.} \end{array}$$

For (SP-2)₈₅, a double analysis was then not considered necessary and only the sulfate was used.

Isotope-dilution analysis was used to cross-check the primary chemical-analysis calibration of the spike solutions. Thus, SP_{Nat} was used to check the concentration of the (SP-2)₈₅ solution. Known volumes (or weights) of SP_{Nat} and (SP-2)₈₅ were mixed and mass-spectrometer-analyzed. Since both solutions had known isotopic ratios and had been subjected to primary standardization, the ⁸⁵Rb/⁸⁷Rb ratio could be calculated from the composition. It could also be determined from the bias-corrected mass-spectrometer measurement. Three mixtures gave the ratios

$$\frac{(^{85}\text{Rb}/^{87}\text{Rb})_{\text{Calc}}}{(^{85}\text{Rb}/^{87}\text{Rb})_{\text{Mass Spec}}} \left\{ \begin{array}{l} \frac{5.0825}{5.0844} = 0.9996 \\ \frac{7.0550}{7.0548} = 1.0000_3 \\ \frac{3.6240}{3.6266} = 0.9993 \end{array} \right.$$

The SP_{Nat} solution was also used as a spike to analyze the concentration of diluted fractions of (SP-1)₈₅, with the resulting concentration values:

From original primary standardization, 2.7491×10^{15}
Rb atoms/mL soln.

From isotope dilution, 2.7524×10^{15} ⁸⁵Rb atoms/mL soln,
with $2.7491/2.7524 = 0.9988$.

These cross-checks indicate that both (SP-1)₈₅ and (SP-2)₈₅ were satisfactorily calibrated.

Since SP₈₇ was used only for spiking the blank samples, where only moderate accuracy was needed, it was not subjected to primary chemical standardization, but was calibrated by isotope dilution.

V. PREPARATION OF RUBIDIUM-85 FOR MASS-SPECTROMETRIC MEASUREMENTS

A. Transfer of Dissolver Solution

The quartz dissolver vessel was transferred to a glovebox, in which all subsequent handling was carried out. Although the high-burnup sample had a radiation level of 100 R at the vessel surface, it was mostly low energy and readily shielded. The solution (about 100 mL) was transferred to a cleaned quartz beaker, with several washes using 4 M HNO_3 . The resulting solution was 3-4 M HNO_3 with ~5 g of mercury. The efficiency of all transfers was monitored by following the ^{137}Cs gamma ray emitted from the solution with a Ge(Li) detector; the transfer efficiency was never less than 99.98%. The sample was evaporated, picked up with HCl and transferred to a precleaned quartz volumetric flask. For the low-burnup sample, the total solution was diluted to the calibrated volume (~100 mL) of this flask; all aliquoting was done with volume-calibrated quartz- and glassware. For the high-burnup sample, the sample was also diluted to the ~100-mL calibrated volume of the flask. However, aliquoting was carried out by weighing the solutions, although it was checked by volume measurement as well. After transfer to the preweighed and volume-calibrated flask, the total weight (140.2564 g) of the solution was measured.

B. Contamination Problems

The ratio of the 87- to 85-chain fission yields is ~1.9. Although this is not the ratio in the dissolver solution, because of the ^{85}Kr holdup, it may be seriously changed if natural rubidium ($^{87}\text{Rb}/^{85}\text{Rb} = 0.3860$) enters the solution. At the very low levels of rubidium formed in the irradiation, contamination by the ubiquitous rubidium-containing dust or other sources was highly likely without strong precautions. Rubidium contamination could be present from the original material in the capsule (uranium and aluminum) or could be introduced by the dissolution or analytical processes.

Contamination in the irradiated capsules could only be evaluated by measuring the rubidium content of unirradiated aluminum and uranium samples taken from the starting material. Such "blank" measurements are described in Sec. IX.C below. Contamination in the processing was minimized by careful cleaning and the use of superpure chemicals. Unavoidable contamination with natural rubidium was checked by "blank" measurements involving only the reagents, columns, glassware, and other containers used. It was further important to remove solids that could be present in the final solution and would interfere with reliable evaporation of minute amounts of rubidium from the mass-spectrometer source filament.

The following procedures were adopted:

1. All operations were carried out in a clean glovebox.
2. Commercial stored-in-quartz triply distilled reagents (acids, etc.) were used (Baker Co. superpure ULTREX).
3. Water of the highest practical purity was used, equivalent in conductivity to, but purer than, triply distilled water (MILLI-Q2 purified water, generated by the MILLIPORE Corporation filter system). This was used for all washings and solution preparation.
4. Only quartzware containers were used for storing the dissolver solutions, and such ware was boiled in concentrated HCl and concentrated HNO₃ and rinsed with the Millipore-filtered water. The Pyrex pipets were rinsed with the same acids and filtered water. Pyrex glassware was used for operations involving short-time contact with the solutions, and these were similarly cleaned.
5. All Dowex ion-exchange resins were washed with Ultrex acids and Millipore-filtered water.
6. The oxalic acid used in the ion-exchange process was purified by crystallization and was further cleaned by treatment with several ion-exchange columns.
7. The Bio-Rex 40 resin was cleaned with water washes and was size-graded.
8. Blank samples were run in the dissolution vessel before and after sample dissolution.
9. Blank solutions were run at every operation; everytime a rubidium sample was run, 10 times as much reagent solution was used for the blank.

C. Purification of Rubidium Sample for Mass-spectrometric Analysis*

The procedure removed the solids and fission products by anion exchange, and then separated rubidium from alkaline earths and other alkalis with a Bio-Rex cation-exchange resin. Under the conditions used, the rubidium and impurities were either tightly bound or completely washed out of the Dowex columns. For separating from the chemically similar alkaline earths and from other alkalis, it was necessary to use the differential elution rates

*A detailed description will be given in a forthcoming paper. (Ref. 1.) Dowex is an ion-exchange material supplied by Dow Chemical Company. Dowex 1 is an anion-type resin; Dowex 50 is a cation-type resin. The X8 designation describes the amount of cross-linkage. Bio-Rex 40 is a cation resin supplied by Bio-Rad, Inc.

from the Bio-Rex column. The calibration of an identical column with radioactive rubidium tracer provided information on the volume of eluting agent required to remove rubidium with a minimum of foreign contamination.

An aliquot of the dissolver solution was evaporated twice with 8 M HCl, taken up in 8 M HCl, placed on a Dowex 1-X8 anion-exchange column, and the rubidium eluted from the column with 8 M HCl. The eluate was evaporated and the residue dissolved in 0.2 M oxalic acid (0.01% H_2O_2), and the rubidium was placed on a Dowex 1-X8 anion-exchange column. The rubidium was eluted with 0.2 M oxalic acid, and was then passed through a Dowex 50-X8 cation-exchange resin. The column was washed with water, and the rubidium was eluted with 4 M HCl. The solution was evaporated; the residue was dissolved in water, placed on the calibrated Bio-Rex 40-X8 exchange column, and washed with 0.2 M HCl; and the rubidium was eluted with 0.85 M HCl, separating the rubidium from alkaline earths and other alkali metals. Despite the small amount of rubidium in the eluted fraction ($\sim 2 \times 10^{15}$ atoms), this procedure gave a product so free of extraneous material that the rubidium volatilized reliably from the filament of the mass-spectrometer source. A sample even more free of extraneous solid material was sometimes made by evaporating the final rubidium solution to dryness and then volatilizing the rubidium with induction heating.

VI. MASS SPECTROMETRY

A. General

Mass-spectrometric analyses were made with two different instruments. Earlier analyses employed a 12-in. (30-cm) 60° sector magnet with electron-multiplier detection, a single-filament thermal-ionization source, and magnetic stepping. Later analyses used a spectrometer with a 90° magnet, a deep Faraday-cup detector (with efficient secondary repression), a triple-filament thermal-ionization source, and magnetic stepping. The filaments were of predegassed zone-refined rhenium.

B. Analyzed Samples

Samples used in the mass-spectrometer isotope-dilution analysis were of four types:

1. Unspiked sample. Aliquot from the dissolver solution, purified as described.
2. Spiked sample. (The "spike" solution contains a known concentration of rubidium with known isotopic ratio.) This sample contains a dissolver solution aliquot, mixed with a known amount of $(\text{SP-1})_{85}$ or $(\text{SP-2})_{85}$ spike solution, purified as described.

3. Chemistry blank. A known amount of the same spike solution as used in type 2, purified as described.

4. Contamination blank. A known amount of SP_{87} added to the dissolved unirradiated aluminum or uranium capsules to check for rubidium in the original materials, purified as described.

C. Effect of Sample Purity on the Source

The reproducible and steady volatilization of rubidium samples in the mass-spectrometer source was found to be more difficult than for many other elements. Crucial to the success of this experiment was elimination of extraneous solid material by the method described in Sec. V.C. above. Even quite small amounts of solid interfered with volatilization from the source. Also of importance was the discovery that use of a higher temperature while "loading" the filament gave much better results.

D. Equations Used in Mass-spectrometry Isotope-dilution Analysis

Two samples were analyzed:

1. Measured amount of spike solution added to measured amount of "unknown" solution (M ; mixture).
2. Unknown solution (U).

The atom ratios in the two samples are

$$R_M = \frac{{}^{85}\text{Rb}}{{}^{87}\text{Rb}} \text{ in sample 1 and } R_U = \frac{{}^{85}\text{Rb}}{{}^{87}\text{Rb}} \text{ in sample 2.}$$

Because of mass-spectrometer bias, the measured values are R_M and R_U . The bias effect is calibrated by measuring the factor B_f , such that

$$R_M = B_f R_M; R_U = B_f R_U. \quad (1)$$

(The definition and measurement of B_f are discussed in Sec. VI.E.)

Let

S_5 = amount of ${}^{85}\text{Rb}$ added in spike to sample 1,

S_7 = amount of ${}^{87}\text{Rb}$ added in spike to sample 1,

U_5 = amount of ${}^{85}\text{Rb}$ added from unknown to sample 1,

and

U_7 = amount of ${}^{87}\text{Rb}$ added from unknown to sample 1.

Since the ratio is the same for the unknown material in both samples 1 and 2,

$$U_5 = U_7 R'_U. \quad (2)$$

In sample 1,

$$R'_M = \frac{S_5 + U_5}{S_7 + U_7}. \quad (3)$$

From Eqs. 2 and 3.

$$U_7 = \frac{S_5 - R'_M S_7}{R'_M - R'_U} \quad (4a)$$

and

$$U_5 = U_7 R'_U. \quad (4b)$$

Changing to measured ratios with Eq. 1,

$$U_7 = \frac{S_5 - S_7 R_M B_f}{(R_M - R_U) B_f} \quad (5a)$$

and

$$U_5 = U_7 R_U B_f. \quad (5b)$$

Equations 5a and 5b give the results in terms of the measured quantities S_5 , S_7 , R_M , R_U , and B_f .

E. Bias Correction

The intensity ratio at the two mass numbers, as measured with the mass spectrometer, must be corrected for a mass-dependent bias effect inherent in the technique. Since the bias depends primarily on the two masses and (within limits) does not depend on the intensity ratios, the bias factor can be calibrated if a mixture of known composition is measured in the spectrometer. The bias factor is defined here as

$$B_f = \frac{(^{85}\text{Rb}/^{87}\text{Rb})_{\text{true}}}{(^{85}\text{Rb}/^{87}\text{Rb})_{\text{Mass Spec}}} \quad (6)$$

The light mass tends to be accentuated in intensity, so $B_f < 1$.

Calibration was carried out through the use of natural rubidium (SP_{Nat} in Sec. IV), for which the unbiased ratio 2.593 has been evaluated by the NBS with an uncertainty of $<0.1\%$. Typical values for B_f were 0.9850 ± 0.0013 in the low-burnup experiment (60° sector machine) and 0.9980 ± 0.0005 in the high-burnup experiment (90° machine), where the error represents data scatter only and does not include the error in the NBS evaluation.

VII. PREPARATION OF KRYPTON FOR COUNTING

A. Purification

Since a measured amount of nonradioactive (dead) krypton gas had been added to the dissolver volume, a quantitative recovery of the krypton was unnecessary. Even with the dilution provided by the initial charge of dead krypton, the gas was much too active for counting in GM tubes and required further dilution with dead krypton. Before an accurate dilution could be made, the active krypton had to be separated from other gases, which included primarily hydrogen, nitrogen oxides, and some nitrogen and oxygen. One-half of the gas volume in the dissolver system was set aside to ensure against catastrophic loss in handling. For the remainder, the gases were extracted from the dissolver volume by adsorption on a large liquid-nitrogen-cooled activated-charcoal trap. Purification was carried out with a gas chromatograph, in which the elution time of krypton had been calibrated under defined conditions. The selected (krypton) peak was passed through the chromatograph several times until no impurity peaks were observed. Examination with a cycloidal mass spectrometer showed impurities to be $<0.1\%$. The same separation and purification procedures were applied to the set-aside krypton gas, and the two portions were combined.

B. Dilution

The purified gas was then further diluted with dead krypton. A double-bulb dilution apparatus was used; it was calibrated by multiple weighing of contained water and had a volume ratio of $\sim 4 \text{ mL}/1000 \text{ mL}$. Two stages of dilution were needed to bring the activity level down to that usable in the counters.

VIII. KRYPTON-85 COUNTING

A. GM Counters

Some of the diluted krypton, in measured amount, was introduced, along with an ethyl ether quench component, into a small ($\sim 45 \text{ mL}$) GM counter made of very pure copper. Each sample was counted for 900-2000 min at a counting rate of 500-600 counts/min (c/m) within a heavily shielded steel

"tomb." A plastic scintillator in anticoincidence removed cosmic-ray background (~ 24 c/m), leaving a net background of ~ 0.6 c/m.

A number of counters were filled and counted. The counting results and dilution factors are given in Sec. X.

B. Absolute Counting

The small GM tubes used had a counting efficiency of about 87-90%, depending upon the gas pressure. It was necessary to calibrate the efficiency before absolute counting of the ^{85}Kr could be carried out. A sample of ^{85}Kr (standardized gas), which had been calibrated some years ago by the double-tube method,² was used for the efficiency calibration.

Six stainless steel GM tubes were used: three of "full" size (~ 2600 mL) and three of "half" size (~ 1300 mL). These were constructed so they were as identical as possible in every important respect, except that the lengths differed. In particular, the tube diameter, the wire size, and the construction of the endpieces were made as alike as possible in all the counters. The counting efficiency of a GM counter is measured by the fraction of cases in which a disintegration occurring within the gas gives rise to a countable pulse. Ideally, every disintegration would yield a pulse, and the counting efficiency would then be 100%. In an actual GM tube, there are two causes for the efficiency being $<100\%$. The most important is the "end effect." This arises because some of the electrons released in the gas near the end of the counter do not result in the usual GM multiplication process, primarily because the electric-field distribution at the end (where the wire is brought in through an insulator) is quite different from the field in the center of the counter. When measured amounts of the gas being standardized are counted in the two sizes of tubes, a comparison of the counting results allows cancelation of the end effect, yielding a measurement of the counting rate per unit length of the midcounter.

A secondary cause of inefficiency is the "wall effect," which arises because some β particles emitted near to and headed toward the cylindrical wall do not cause ionization within the gas and hence do not give rise to countable pulses. If a β -particle track in the gas is short, there is a finite probability that no ionization will occur before the β particle collides with the wall. Since this probability decreases with increasing gas pressure, extrapolation of measurements made at several pressures allows evaluation of the wall effect. With these two corrections made, it is possible to calculate the absolute disintegration rate of the gas being standardized.

The measurement provided a value for the absolute specific activity $[(d/m)/\text{mL krypton}]$ for the standardized gas at standard pressure and temperature. The counter efficiencies of the GM tubes used in the ^{85}Kr measurements were evaluated by counting measured amounts of standardized gas.

The 1- σ error in the standardized gas activity is estimated to be ~1%, and this sets the major limit on the accuracy of the present experiment.

Examination of the relations used in the calibration process shows that our measurement of the ^{85}Kr activity essentially evaluates the ratio of the specific activity of the diluted krypton to that of the standardized gas.

With the notation

N_{ST} = number of moles of standardized gas used in
GM tube K,

N_U = number of moles of diluted gas used in tube K,

S_{ST} = d/m per mole of standardized gas,

S_U = d/m per mole of diluted gas,

$A_{ST} = S_{ST}N_{ST}$ = d/m from standardized gas in tube K,

and

M_{ST} = measured counting rate in tube K,

(7a)

the counting efficiency is defined as

$$\epsilon(N_{ST}) = \frac{M_{ST}}{A_{ST}}. \quad (7b)$$

The quantity $\epsilon(N_{ST})$ is a function of N_{ST} , typically varying from 0.87 to 0.90 over the widest useful pressure range for the GM tube. The quantity $\epsilon(N_{ST})$ is measured for a number of N_{ST} values. The fitted calibration curve (ϵ versus N_{ST}) is generally a straight line, but may sometimes show a slight curvature.

For the unknown, N_U is measured and $\epsilon(N_U)$ is derived from the calibration curve. Hence, with M_U = measured counting rate,

$$A_U = \frac{M_U}{\epsilon(N_U)} \text{ and } S_U = \frac{A_U}{N_U}. \quad (8)$$

Then

$$\frac{S_U}{S_{ST}} = \frac{M_U}{M_{ST}} \frac{N_{ST}}{N_U} \frac{\epsilon(N_{ST})}{\epsilon(N_U)}. \quad (9)$$

(The M values are the measured rates corrected for dead-time losses and background, and N values are calculated from the pressure, volume, and temperature measurements made on each gas, with $PV = NRT$.)

The ratio $\epsilon(N_{ST})/\epsilon(N_U)$ is derived from the calibration curve and is generally close to unity ($= 1$ if $N_{ST} = N_U$). The other ratios are measured. Thus, the fractional error in the calibration of the standardized gas is transmitted directly into the S_U error.

IX. MASS-SPECTROMETRY RESULTS

A. Low-burnup Sample

The isotope dilution results for three measurements are shown in Table II. Note from Eqs. 5a and 5b that the error in the bias measurement has relatively little impact on the error in the ^{85}Rb content. This is due primarily to the fact that the spike is predominantly ^{85}Rb .

TABLE II. Low-burnup Isotope-dilution Results^a

	Sample No.		
	1	2	3
$S_5 \times 10^{-15}{}^b$	10.999 ± 0.017	10.992 ± 0.017	10.992 ± 0.017
$S_7 \times 10^{-15}{}^b$	0.0508 ± 0.0002	0.0508 ± 0.0002	0.0508 ± 0.0002
$R_M{}^c$	1.4457 ± 0.0027	1.7775 ± 0.0048	1.7802 ± 0.0006
$R_U{}^c$	0.4315 ± 0.0002	0.4313 ± 0.0007	0.4313 ± 0.0007
$B_f{}^c$	0.9849 ± 0.0013	0.9850 ± 0.0013	0.9850 ± 0.0013
$U_5(x 10^{-15}) \text{ atoms}^d$ (^{85}Rb in mixture solution)	4.648 ± 0.015	3.493 ± 0.016	3.486 ± 0.009
$U_7(x 10^{-15}) \text{ atoms}^d$ (Rubidium in-mixture solution)	10.930 ± 0.036	8.222 ± 0.034	8.206 ± 0.017
Volume Ratio ^e	50.012 ± 0.012	66.584 ± 0.019	66.627 ± 0.021
$^{85}\text{Rb}(x 10^{-15}) \text{ atoms}^d$ in dissolver	232.5 ± 0.7	232.6 ± 1.0	232.2 ± 0.6
$^{87}\text{Rb}(x 10^{-15}) \text{ atoms}^d$ in dissolver	547.1 ± 1.7	547.5 ± 1.9	546.7 ± 0.8

^aMeaning of terms is described in Sec. VI.D.

^bError is based on error in calibrating the spike solution and the aliquoting error.

^cError represents the measured scatter of the mass-spectrometer ratios.

^dError is propagated from Eqs. 5a and 5b using the errors of the measured quantities.

^eVolume ratio = $V_{\text{diss}}/V_{\text{aliquot}}$ where V_{diss} = volume of dissolver solution, and V_{aliquot} = volume of the solution aliquot put into the mixture.

The average value for the content of the entire dissolver solution is

$$(232.4 \pm 0.5) \times 10^{15} \text{ atoms } ^{85}\text{Rb}.$$

The errors were derived by propagating the errors in Table II rather than from the observed scatter, which yields a considerably smaller error value. The error in the calibration value of the spike solution, which enters into all three measurements, has been appropriately treated.

This result has not been corrected for contamination, which is treated in Sec. IX.D below.

B. High-burnup Sample

The isotope-dilution results for three measurements are shown in Table III. The average value for the content of the entire dissolver solution is

$$(366.00 \pm 0.34) \times 10^{15} \text{ atoms } ^{85}\text{Rb}.$$

The error was taken by propagating the errors in Table III, although it was only a little larger than that calculated from the data scatter. The fact that the spike-solution calibration error and the error in R_U were common to all three measurements was taken into consideration.

TABLE III. High-burnup Isotope-dilution Results^a

	Sample No.		
	1	2	3
$S_5 \times 10^{-5}{}^b$	22.690 ± 0.013	12.273 ± 0.007	11.618 ± 0.006
$S_7 \times 10^{-15}{}^b$	0.03636 ± 0.00002	0.01967 ± 0.00001	0.01862 ± 0.00001
$R_M{}^c$	2.0541 ± 0.0012	1.1793 ± 0.0007	1.2128 ± 0.0007
$R_U{}^{c,d}$	0.43257 ± 0.00017	0.43257 ± 0.00017	0.43257 ± 0.00017
$B_f{}^{c,d}$	0.9970 ± 0.0007	0.9970 ± 0.0007	0.9970 ± 0.0007
$U_5(x \ 10^{-15}) \text{ atoms}^e$ ^{85}Rb in mixture solution	6.0330 ± 0.0064	7.0962 ± 0.0083	6.4285 ± 0.0077
$U_7(x \ 10^{-15}) \text{ atoms}^e$ ^{85}Rb in mixture solution	13.989 ± 0.016	16.454 ± 0.022	14.906 ± 0.019
Mass Dilution Ratio ^f	60.623	51.619	56.919
$^{85}\text{Rb}(x \ 10^{-15}) \text{ atoms}^e$ in dissolver	365.74 ± 0.39	366.30 ± 0.43	365.91 ± 0.44
$^{87}\text{Rb}(x \ 10^{-15}) \text{ atoms}^e$ in dissolver	848.06 ± 0.99	849.34 ± 1.12	848.46 ± 1.07

^aMeaning of terms described in Sec. VI.D.

^bError is based on error in calibrating the spike solution and the weight aliquoting error.

^cThe error represents the measured scatter of the mass spectrometric ratios.

^dThe same measured ratio was used for all three measurements.

^eError is propagated from Eqs. 5a and 5b, using the errors of the measured quantities.

^fMass ratio = $M_{\text{diss}}/M_{\text{aliq}}$, where M_{diss} = weight of dissolver solution and M_{aliq} = weight of the solution aliquot put into the mixture.

C. Evaluation of Rubidium Contamination in Original Capsules

The values given above constitute the total recovery of ^{85}Rb from the dissolver solution. Contamination from the chemical treatment, although constantly checked for, proved to be negligible. However, it was necessary to estimate the amount of rubidium (most likely natural) that was incorporated in the original capsules.

The irradiated uranium samples consisted of uranium oxide, mixed with aluminum powder (for heat conductivity) and encapsulated in an aluminum metal container, with welded closure. Several capsules with the same kind of encapsulation were analyzed to evaluate the rubidium content of the original material. One was an irradiated capsule containing only aluminum powder, and two were unirradiated samples containing the uranium oxide-aluminum powder mixture.

Each was dissolved and diluted to 100.0 mL, using the same precautions as were involved in dissolving the irradiated uranium samples. One of the uranium-containing samples (No. 25) contained 0.06 g of uranium and was dissolved on the bench top in a cleaned, quartz beaker. The other (No. 67) contained 0.76 g of uranium and was dissolved in a new dissolver apparatus like that used for the irradiated uranium capsules.

The results are presented in Table IV. Chemistry blanks run at the same time showed considerably less contamination, so it may be inferred that the results really correspond to contamination in the original material. All the results are consistent with a constant contamination value, and from the fact that the uranium content (from 0 to 0.76 g) made little or no difference, it may be concluded that the aluminum contained most of the contamination.

TABLE IV. Concentration of ^{85}Rb in the Blank Solutions^a

From	Concentration
Aluminum capsule (irradiated)	1.193 1.116 1.165
Uranium capsule No. 25 (0.06 g of uranium) (unirradiated)	1.195
Uranium capsule No. 67 (0.76 g of uranium) (unirradiated)	1.205 1.292
Average = 1.194 ± 0.024	

^aNumber of atoms ($\times 10^{-13}$) of ^{85}Rb per milliliter of solution.

Since each sample was dissolved in 100 mL of solution, the blank correction is

$$1.194 \times 10^{13} \times 100.0 = 1.19(\pm 0.02) \times 10^{15} \text{ atoms } ^{85}\text{Rb}.$$

D. Net ^{85}Rb from Dissolver Solutions

The amount of ^{85}Rb ascribed to the original rubidium contamination was subtracted from the measured ^{85}Rb yield; the net values are shown in Table V.

TABLE V. Net ^{85}Rb Yield

Sample	Measured ^{85}Rb Yield, atoms $\times 10^{-15}$	Contamination, atoms $\times 10^{-15}$	Net ^{85}Rb Yield, atoms $\times 10^{-15}$
Low burnup	232.4 ± 0.5	1.19 ± 0.02	231.2 ± 0.5
High burnup	366.0 ± 0.34	1.19 ± 0.02	364.8 ± 0.34

X. RESULTS OF KRYPTON-85 ANALYSIS

A. General

The ^{85}Kr analysis must include a correction for decay from the time of formation to the time of counting.

At present, the most reliable half-life value is taken as

$$T_{1/2}(^{85}\text{Kr}) = 10.75 \pm 0.02. \quad (10)$$

This is a weighted average of the value 10.76 yr from Ref. 3 and 10.72 yr from Ref. 4, as recalculated in Ref. 5. In Ref. 3, six samples were followed for 0.8 to 0.95 of a half-life, and the half-life values from the six samples were averaged. The error quoted was the standard error of the mean of the six numbers. Each sample was contained in a thin-walled glass vial and β particles were counted in an internal bell-shaped proportional counter; uranium samples were used as constant reference standards. The result in Ref. 4 involved the measurement of bremsstrahlung from a single sealed ^{85}Kr sample measured in a total ionization chamber with a reentrant thimble. Measurements were followed for 1.5 half-lives. The air-containing ionization chamber was open to the atmosphere. The error quoted is that derived from the least-squares formalism applied to the data from the single sample. The results of both experiments have been combined here to give the value 10.75 yr, greater weight being given to Ref. 3, because of the redundancy of samples and the greater reliability of counting β particles in an internal counter as compared to measuring ionization from the relatively soft bremsstrahlung with an external chamber. The errors calculated from each experiment (~ 0.007 yr) were much smaller than the difference in half-life values (~ 0.04 yr); the estimated error has therefore been enlarged to 0.02 yr.

The error in the ^{85}Kr analysis then contains the error in the decay correction, as well as those errors arising from the filling and counting process, namely: (1) dilution errors, (2) counting statistics and (3) fluctuations in GM-tube calibration. These three errors, which are more or less random, will tend to partially average out. Another error component, that in the calibration of the standardized gas, constitutes a systematic effect and will be discussed separately, as will also be done for the error in the half-life.

B. Low-burnup Sample

Three sequences of dilutions were made from the original gas, and each was separately entered into a number of GM tubes and counted. Table VI contains the pertinent data. The counting rates have been corrected for decay, so that all rates correspond to those that would have been measured were they measured simultaneously. The counting periods were ~ 1000 min, and in a few cases, samples were counted twice. With few exceptions, the samples were counted in different GM tubes, so the calibration values were mostly taken from independently measured calibration curves. A small correction ($<0.02\%$) has been applied to allow for the slight activity (above background) of the dilution gas. The dilution factors were measured with an error of $\sim 0.1\%$. It is evident from Table VI that the largest error source arises from the combined effect of random errors in tube filling and GM-tube calibration. The scatter of the tabulated results indicates that the observed error per sample is sizably larger than the counting error ($\sim 0.1\%$). The error of the average given in the table is an estimate of the standard error of the mean, $\sigma(\bar{x}) = \sigma(x)/\sqrt{n}$.

TABLE VI. Low-burnup Sample: Dilution Factors and Counting Results for ^{85}Kr

Dilution Factor	Sample No.	Count Time, min	Specific Activity, (d/m)/mL ^a	Average ^b
(A) 1.6683×10^7	1	940	352.6	352.8 ± 0.8
	2	940	349.1	
	3	940	354.4	
	4	940	353.2	
	5	940	352.9	
	6	940	354.8	
(B) 1.4640×10^7	7	984	402.6	401.5 ± 0.9
	8	984	400.5	
	9	984	400.3	
	10	984	400.0	
	11	984	405.4	
	12	984	400.1	
(C) 2.2769×10^7	13	1986	258.6	259.4 ± 0.3
	14	1986	260.6	
	15	1986	259.0	
	16	1986	259.3	
	17	1986	259.7	

^ad/m based on the efficiency calibration. Amount of gas in tube was reduced to volume at 0.101 MPa (760 Torr), 25°C. All results were decay-corrected to the midpoint of the counting period of the A dilution.

^bThe error is the estimated standard error of the mean, $s/\sqrt{n}$.

The dilution factors and averaged counting rates were combined as in Table VII to give

$$A_I = (5.890 \pm 0.008) \times 10^9 \text{ d/m } ^{85}\text{Kr},$$

the activity of the krypton from the irradiated capsule as measured at the midpoint of the first counting period. To this result must be applied two corrections for ^{85}Kr decay.

TABLE VII. Low-burnup Sample: ^{85}Kr Yield from Irradiated Sample

Dilution Sequence	Observed Specific Activity in Final Dilution (d/m)/mL ^a	Dilution Factor ^b ($\times 10^{-7}$)	Total d/m of ^{85}Kr ^c ($\times 10^{-9}$)
A	352.8 ± 0.8	1.6683	5.886 ± 0.013
B	401.5 ± 0.9	1.4640	5.878 ± 0.013
C	259.4 ± 0.3	1.2769	5.906 ± 0.007
Average			5.890 ± 0.008^d

^aVolume reported at the standardized conditions, 25°C, 0.101 MPa (760 Torr).

^bEssentially number of milliliters of dead krypton added in this dilution sequence. Includes the original volume added in the dissolution process.

^cFrom the dissolver solution. As of the midpoint of the first counting period.

^dThe propagated error of the average is 0.0065, whereas the estimated standard error of the mean, $s/\sqrt{3}$, is 0.008. The latter, being larger, is used.

If T_I = time from the end of the irradiation to the midpoint of the first count, then the activity at the end of the irradiation is

$$B_I = A_I \exp(\lambda T_I), \quad (11)$$

where λ is the decay constant of ^{85}Kr corresponding to the half-life in Eq. 10.

There is also a decay correction for the decay of ^{85}Kr during the period of formation. The number of ^{85}Kr atoms present at any time t during the irradiation is given by the difference between the formation rate (q) and the decay rate (λK), i.e.,

$$\frac{dK}{dt} = q(t) - \lambda K, \quad (12)$$

where K = number of ^{85}Kr atoms present at time t and λ = decay constant. The formation rate is

$$q(t) = (\sigma_f F) N_{25}(t) \phi(t), \quad (13)$$

where σ_f = ^{235}U fission cross section, F = factor containing the fission yield of the 85 chain and the branching fraction leading to the formation of ^{85}Kr , N_{25} = number of ^{235}U atoms present and $\phi(t)$ = neutron flux at the sample.

The general solution for Eq. 12 is

$$K = e^{-\lambda t} \int e^{\lambda t} q(t) dt + \rho e^{-\lambda t}, \quad (14)$$

where ρ is derived from the condition that $K = 0$ at $t = 0$, i.e.,

$$\rho = - \left[\int_0^\infty e^{\lambda t} q(t) dt \right]_{t=0}. \quad (15)$$

In general, Eqs. 14 and 15 require numerical integration, because N_{25} decreases with irradiation time and ϕ varies with time. For the low-burnup sample, the total irradiation time τ was 20 days and the reactor power level remained fairly constant during this time. Since the total burnup was ~1%, N_{25} changed little during this time. Hence, $q(t)$ may be treated as constant, whence $\rho = -q/\lambda$ and, at $t = \tau$,

$$K(\tau) = \frac{q}{\lambda} (1 - e^{-\lambda \tau}) \quad (16)$$

is the amount at the end of the irradiation. If ^{85}Kr did not decay during the irradiation, the amount formed would have been $K_\infty = q\tau$. Hence, since $\lambda\tau$ is small (~0.0035),

$$\frac{K}{K_\infty} \approx \frac{1}{\lambda\tau} \left\{ 1 - \left[1 - \lambda\tau + \frac{(\lambda\tau)^2}{2} \right] \right\} \approx \left(1 - \frac{\lambda\tau}{2} \right) \approx e^{-\lambda\tau/2}. \quad (17)$$

Thus, for the low-burnup sample, the number of ^{85}Kr atoms formed may be determined by correcting the number of atoms present at the end of irradiation for decay from the midtime of the irradiation.

Then, from Eq. 11, the activity of the ^{85}Kr atoms formed was

$$C_I = B_I \exp(\lambda T_I) \exp(\lambda\tau/2). \quad (18)$$

With $T_I = 1308$ days and $\tau = 20$ days,

$$\frac{dN_{85}}{dt} = C_I = (5.890 \times 10^9) \times 1.2597 \times 1.0018 = (7.433 \pm 0.010) \times 10^9 \text{ d/m},$$

and from Eq. 10, the number of ^{85}Kr atoms formed was

$$N_{85} = \frac{dN_{85}/dt}{\lambda} = (6.063 \pm 0.008) \times 10^{16}.$$

C. High-burnup Sample

Here, too, three dilution sequences were made, and the gas was counted in a number of GM tubes. Four of the tubes were used twice, so there is more overlap than in Sec. X.B of the calibration errors; however, the bulk of the counts were taken in separately calibrated tubes. In all other respects, the operations and corrections were like those used for the low-burnup sample. Data are given in Table VIII.

TABLE VIII. High-burnup Sample: Dilution Factors and Counting Results for ^{85}Kr

Dilution Factor	Sample No.	Count Time, min	Specific Activity, (d/m)/mL ^a	Average ^b
(E) 8.8180×10^7	1	2000	92.56	92.48 ± 0.10
	2	2000	92.26	
	3	2000	92.67	
	4	2000	92.72	
	5	2000	92.20	
	6	2000	92.48	
	7	2000	92.32	
	8	2000	92.59	
(F) 9.2965×10^7	9	2000	87.51	87.42 ± 0.11
	10	2000	87.34	
	11	2000	87.80	
	12	2000	87.12	
	13	2000	87.52	
	14	2000	87.65	
	15	2000	87.29	
	16	2000	87.65	
	17	2000	87.31	
	18	2000	86.99	
(G) 10.4680×10^7	19	2000	78.09	77.64 ± 0.14
	20	2000	77.65	
	21	2000	77.53	
	22	2000	77.39	
	23	2000	77.70	
	24	2000	78.21	
	25	2000	77.65	
	26	2000	77.89	

^ad/m based on the efficiency calibration. Amount of gas in tube was reduced to volume at 0.101 MPa (760 Torr), 25°C. All results were decay-corrected to the midpoint of the first counting period of the E dilution.

^bThe error is the estimated standard error of the mean $s/\sqrt{n}$, where n is taken as one-half the number of samples.

The dilution factors and averaged counting rates were combined in Table IX to give

$$A_{II} = (8.136 \pm 0.009) \times 10^9 \text{ d/m } ^{85}\text{Kr}$$

as the activity of the krypton from the irradiated sample as measured at the midpoint of the first counting period. Corresponding to Eq. 11, the decay correction is made to the end of the irradiation, where $T_{II} = 1800.7$ days.

TABLE IX. High-burnup Sample: ^{85}Kr Yield
from Irradiated Sample

Dilution Sequence	Observed Specific Activity in Final Dilution, (d/m)/mL ^a	Dilution Factor ^b ($\times 10^{-7}$)	Total d/m of ^{85}Kr ^c ($\times 10^{-9}$)
E	94.48 ± 0.10	8.8180	8.155 ± 0.009
F	87.42 ± 0.11	9.2965	8.127 ± 0.010
G	77.64 ± 0.14	10.4680	8.127 ± 0.015
Average			8.136 ± 0.009^d

^{a,b,c}See footnotes in Table VII.

^dThe propagated error is 0.0067, whereas the estimated standard error of the mean, $s/\sqrt{3}$, is 0.009. As in Table VII, the latter is used.

The correction for decay during the irradiation period is more complex than in the low-burnup case, because (1) a large portion of the ^{235}U was burned out, and (2) the neutron flux varied considerably, indeed being zero for a large portion of the irradiation time. It was necessary to solve for the correction by numerical integration, using Eqs. 14 and 15. The function $\phi(t)$ in Eq. 13 was evaluated (within a constant factor) through the use of the power chart of the reactor. (We are grateful to W. Maeck of Allied Chemical Corporation for providing us with a copy of this chart.) The function $N_{25}(t)$ was evaluated, as described in Sec. XII below, through the mass-spectrometric measurement of the total ^{235}U burnup and calculation using $\phi(t)$.

From the Mean-value Theorem, the correction for any arbitrary $\phi(t)$ can be replaced by a single decay correction to an "effective" median time. For a constant flux, this time would lie in the earlier part of the irradiation period. However, since the reactor was down more frequently in the first half of the period, the effective median time turned out (coincidentally) to be almost exactly the midpoint of the irradiation period.

The numerical integration gave the value (corresponding to Eq. 17)

$$\frac{K}{K_{\infty}} = \frac{1}{1.0291} \quad (19)$$

Hence, the total decay correction provides the activity of the ^{85}Kr atoms formed as

$$\frac{dN_{85}}{dt} = C_{II} = (8.136 \times 10^9) \times 1.3742 \times 1.0291 = (11.506 \pm 0.13) \times 10^9 \text{ d/m}$$

and from Eq. 10, the number of ^{85}Kr atoms formed was

$$N = (9.386 \pm 0.011) \times 10^{16} \text{ atoms.}$$

XI. BRANCHING-FRACTION RESULTS

From the decay scheme in Fig. 1, the branching fraction BF may be defined as

$$BF = \frac{N_{Kr}}{N_{Rb} + N_{Kr}}, \quad (20)$$

where

N_{Rb} = Number of atoms of ^{85}Rb formed from the β decay of ^{85m}Kr

and

N_{Kr} = Number of atoms of ^{85}Kr formed from internal transition of ^{85m}Kr .

In using the measured results of Secs. IX and X, we require one important correction. The effect of the decay lag of the ^{85m}Kr on the results may be neglected, since its half-life is short compared to the times involved. However, correction must be made for the fact that some of the ^{85}Rb observed arose from the β decay of the 10.75-yr ^{85}Kr , and not from the direct decay of ^{85m}Kr . Hence, we may write Eq. 20 as

$$BF = \frac{N_{Kr}}{(N'_{Rb} - N^*_{Rb}) + N_{Kr}} \quad (21)$$

where N_{Kr} is defined as above and is the value calculated in Sec. X, N'_{Rb} is the value calculated in Sec. IX, and N^*_{Rb} is the amount of ^{85}Rb grown from the long-lived ^{85}Kr . This decay occurred throughout the irradiation and until the end of the dissolution period (when the carrier krypton gas was separated from the solution), after which ^{85}Kr decay no longer contributed to the amount of ^{85}Rb measured. As mentioned in Sec. X, the effect of ^{85}Kr decay could be accurately corrected for by using the midpoint of the irradiation, for both the low- and high-burnup samples. The waiting periods involved (Δt_i) are given in Table I. We have, then,

$$N^*_{Rb} = N_{Kr}[1 - \exp(-\lambda\Delta t_i)] \quad (22)$$

or

$$BF = \frac{N_{Kr}}{N'_{Rb} + N_{Kr} \exp(-\lambda\Delta t_i)} = \frac{1}{\frac{N'_{Rb}}{N_{Kr}} + \exp(-\lambda\Delta t_i)} \quad (23)$$

The results calculated from this relation are shown in Table X. From theoretical considerations, it would not be expected that the BF value would vary with the amount of burnup, nor even with the neutron spectrum, should the two samples have differed somewhat in this exposure. It is gratifying, therefore, that the two results check quite well (within 0.5%). That the difference exceeds the estimated error by a factor of two indicates that the propagated errors from the measurements of N_{Rb} and N_{Kr} do not completely describe the random-error sources.

TABLE X. Calculation of Branching Fraction

	Low-burnup Sample	High-burnup Sample
Measured ^{85}Rb Yield	$231.1 \pm 0.5 \times 10^{15}$	$364.8 \pm 0.34 \times 10^{15}$
Measured ^{85}Kr Yield	$6.063 \pm 0.008 \times 10^{16}$	$9.386 \pm 0.011 \times 10^{16}$
N_{Rb}/N_{Kr}	3.813	3.887
$\exp(-\lambda \Delta t_i)$	0.8037	0.7549
BF	0.21659 ± 0.00046	0.21545 ± 0.00028
BF (Low) - BF (High) = 0.00114 ± 0.00054		
BF _{av} = 0.2160 ± 0.0006		

The two results are averaged as

$$BF = 0.2160 \pm 0.0006;$$

where the error is taken as $s/\sqrt{2} = (x_1 - x_2)/2$.

The error estimates given cover only the random errors within the experiment and do not include the major sources of systematic error. These are (1) the error in the ^{85}Kr half-life, which impacts most directly in the calculation of N_{85} from the relation $N_{85} = (dN_{85}/dt)/\lambda$ in Sec. X, and (2) the error in calibrating the standardized gas, which impacts the evaluation of dN_{85}/dt in Sec. X (see Eq. 9). These errors are discussed further in Sec. XIII.

XII. BURNUP EFFECTS IN HIGH-BURNUP EXPERIMENT

A. Burnup of ^{85}Rb and ^{85}Kr

In the following, only the thermal-neutron capture is considered. For the accuracy required, resonance neutron capture may be neglected.

From Refs. 6 and 7, the thermal-neutron capture cross sections needed are: $\sigma(^{85}Rb) = 0.46$ b, $\sigma(^{85}Kr) = 1.66$ b, and $\sigma_a(^{235}U) = 681$ b.

From Table I, the ^{235}U burnup was 0.418, hence $\exp(-\sigma_{235}\phi t) = (1 - 0.418)$ and $\phi t = 7.9 \times 10^{20}$ neutrons/cm² (ϕt = total flux). Since neutron capture by both ^{85}Rb and ^{85}Kr are second order reactions, the fractions of these that are burnt out are

$$f(^{85}\text{Rb}) = \frac{1}{2}\sigma(^{85}\text{Rb})\phi t = 0.00018$$

and

$$f(^{85}\text{Kr}) = \frac{1}{2}\sigma(^{85}\text{Kr})\phi t = 0.00066.$$

Were these corrections entered into Eq. 23, BF would change by the factor 1.0003, which is negligible.

It is evident that the secondary burnup is also negligible in the low-burnup experiment.

B. Measurement of ^{235}U Burnup and Its Original Mass

To correctly evaluate the effect of ^{85}Kr decay during its period of formation, it is necessary to know the total burnout of ^{235}U during the irradiation. Because the initial amount of ^{235}U placed in the capsule was not accurately known, it was calculated by measurement of (1) the residual amount of ^{235}U in the dissolver solution, (2) the number of fissions, and (3) the amount of ^{236}U formed by the reaction $^{235}\text{U}(n, \gamma)^{236}\text{U}$. Items (1) and (3) were evaluated by isotope-dilution mass spectrometry and a small correction for the neutron burnup of both initial and formed ^{236}U .

The isotope dilution measurements gave the results:

1. Mass ^{235}U left = 21.223 mg, corresponding to $N_{25}^1 = 5.437 \times 10^{19}$ atoms.
2. The $^{236}\text{U}/^{235}\text{U}$ atom ratio is $\left(\frac{N_{26}}{N_{25}}\right)_{\text{obs}} = 0.10824$.

The dependence of the ^{236}U content on the irradiation time is given by the relation

$$\left(\frac{N_{26}}{N_{25}}\right)_{\text{obs}} = \left\{ \frac{N_{26}^0}{N_{25}^0} + \Theta[1 - \exp(-\phi t \sigma_{a6})] \right\} \exp(\phi t \sigma_{a6}), \quad (24)$$

where $N_{26}^0/N_{25}^0 = 0.0063$ = initial atom ratio of ^{236}U and ^{235}U , $\sigma_{25} = \sigma_a(^{235}\text{U})$, $\sigma_{a6} = \sigma_{25} - \sigma_\gamma(^{236}\text{U})$, ϕt = integrated flux, $\psi = \sigma_\gamma(^{236}\text{U})/\sigma_a(^{235}\text{U})$, and

$$\Theta = \frac{\sigma_c(^{235}\text{U})}{\sigma_{a6}} = \left(\frac{\alpha}{1 + \alpha} \right) / (1 - \psi) \quad (25)$$

and

$$\alpha = \frac{\frac{\Theta}{1 - \psi}}{1 - \psi - \Theta}, \quad (26)$$

with $\alpha = \sigma_c/\sigma_f$ for ^{235}U , while $\psi = 5.2/680.8 = 0.00764$ and $\sigma_{a6} = 680.8 - 5.2 = 675.6$ from Refs. 6 and 7.

The fractional burnup of ^{235}U is given by

$$\text{FB} = \frac{(N_{85}/\text{FY}) + N_{26}^*}{N_{25}^0} = \frac{(N_{85}/\text{FY}) + N_{26}^*}{N_{25}^0 + (N_{85}/\text{FY}) + N_{26}^*}, \quad (27)$$

where N_{25}^0 = initial amount of ^{235}U , N_{85} = amount of 85 chain formed, FY = fission yield of 85 chain, N_{26}^* = total amount of ^{236}U formed by neutron capture, and N_{85} = sum of ^{85}Rb and ^{85}Kr present at one time = 4.356×10^{17} atoms. Given the value FY , N_{26}^* may be iteratively calculated from Eq. 24 setting the preliminary value of FB to $[1 - \exp(-\phi t \sigma_{25})]$ and then calculating $(\phi t \sigma_{a6}) = (\sigma_{a6}/\sigma_{25})(\phi t \sigma_{25})$, with $\sigma_{a6}/\sigma_{25} = 0.9924$ (Refs. 6 and 7).

Two values of FY were considered. One is from the national compiled values^{8,9}

$$\text{FY}_1 = 0.01314.$$

The other is an alternative value (not consistent, within the assigned errors, with FY_1) which is taken from Ref. 10:

$$\text{FY}_2 = 0.01333.$$

These differ by only 1.4%, hence yield similar values:

$$\text{From } \text{FY}_1, \text{FB}_1 = 0.418, \alpha = 0.176, {}^{235}\text{U}_{\text{orig}} = 36.48 \text{ mg.}$$

$$\text{From } \text{FY}_2, \text{FB}_2 = 0.415, \alpha = 0.179, {}^{235}\text{U}_{\text{orig}} = 36.26 \text{ mg.}$$

The value entered in Table I was taken from FY_1 , but the impact of the difference in FY on the "decay correction during irradiation" was negligible, since the correction itself was only 2.9% (see Eq. 19).

Note that both α values calculated from Eq. 26 are consistent within the errors of the estimates, but deviate somewhat from the evaluated thermal value in Refs. 6 and 7 which is $\alpha = 0.169 \pm 0.002$.

XIII. OTHER MEASUREMENTS OF THE BRANCHING FRACTION

(Note that the errors discussed in this section are 1 - σ values.)

A. From Gamma-ray Intensity Measurements

It is evident from Fig. 1 that the BF of Eq. 13 can be written as

$$BF = \frac{I_{Tr}(305)}{I_{Tr}(305) + I_{Tr}(151)}, \quad (28)$$

where $I_{Tr}(305)$ is the transition intensity of the decay of ^{85m}Kr to ^{85}Kr and $I_{Tr}(151)$ is the transition intensity from the first excited state of ^{85}Rb to its ground state. Essentially all the β decay from ^{85m}Kr to ^{85}Rb passes through the 151-keV excited state. These transition intensities are determinable by measurement of the gamma-ray and conversion-electron intensities. Then $I_{Tr}(305) = I_{\gamma}(305) + I_e(305) = I_{\gamma}(305)(1 + \alpha_{305})$, where $\alpha_{305} = I_e(305)/I_{\gamma}(305)$ = conversion coefficient of the 305-keV transition.

Equation 28 is expressed as

$$BF = \frac{1}{1 + \chi}, \quad (29)$$

with

$$\chi = \frac{I_{\gamma}(151)}{I_{\gamma}(305)} \frac{1 + \alpha_{151}}{1 + \alpha_{305}} = R_{\gamma} R_{\alpha}. \quad (30)$$

Three measurements are available for R_{γ} and one for R_{α} :

$$R_{\gamma} = 5.59 \pm 0.31 \quad (\text{Ref. 11}),$$

$$R_{\gamma} = 5.38 \pm 0.28 \quad (\text{Ref. 12}),$$

$$R_{\gamma} = 5.35 \pm 0.18 \quad (\text{Ref. 13}),$$

and

$$R_{\alpha} = 0.693 \pm 0.011 \quad (\text{Ref. 12}).$$

The weighted average for R_{γ} is 5.39 ± 0.14 . Then Eq. 29 gives

$$BF = 0.211 \pm 0.005$$

B. From Mass Spectrometric Determination of ^{85}Kr and ^{85}Rb

Two such measurements have been made at the same laboratory,^{10,14} in which highly irradiated ^{235}U samples were dissolved and the recovered ^{85}Kr and ^{85}Rb were determined by mass-spectrometric isotope-dilution analysis.

The measurement from Ref. 14 gave the value

$$\text{BF} = 0.2176 \pm 0.0024.$$

The error is the standard error of the mean based on three samples (i.e., $s/\sqrt{3}$) and "includes the random and systematic errors." (Presumably the systematic errors contain the uncertainties in the spike analyses.)

The measurement from Ref. 10, as corrected,¹⁵ is reported to give

$$\text{BF} = 0.2194 \pm 0.0008.$$

The error includes the standard error of the mean based on the scatter among six samples (i.e., $s/\sqrt{6}$) and some systematic errors, but does not contain allowance for the systematic error involved in the mass-spectrometer bias correction for the ^{85}Kr isotope-dilution analysis.

C. Present Work: From Mass-spectrometric Determination of ^{85}Rb and Absolute Gas Counting of ^{85}Kr

The value given in Sec. X was

$$\text{BF} = 0.2160 \pm 0.0006$$

The error is the standard error of the mean, based on the scatter of two samples (i.e., $s/\sqrt{2}$) and includes allowance for various systematic errors, the most important of which are the spike calibration error and the mass-spectrometer bias correction.

Two systematic errors not included in this value are the errors in $T_{1/2}$ (^{85}Kr) and the error in the calibration of the standardized gas. There are no strong data on which to base either error value, so only judgmental estimates will be used. The situation relative to $T_{1/2}$ (^{85}Kr) was described in Sec. X.A. A more important error is that in the calibration of the standardized gas, since this enters into the GM-counter efficiency calibration; the error is estimated to be ~1%.

The errors are combined to give

$$\text{BF} = 0.2160 \pm 0.0019.$$

All measurements are summarized in Table XI.

TABLE XI. Summary of Branching-fraction Measurements

Method	Value	References
Gamma-ray intensity measurements, corrected with conversion-coefficient measurements	0.211 ± 0.005	11-13
Mass-spectrometer measurement of ^{85}Kr and ^{85}Rb	0.2176 ± 0.0024	14
Mass-spectrometer measurement of ^{85}Kr and ^{85}Rb	0.2194 ± 0.0008^a	10
Mass-spectrometer measurement of ^{85}Rb ; absolute gas counting of ^{85}Kr	0.2160 ± 0.0019	Present work

^aNo value was provided for an important systematic error that arises in the ^{85}Kr isotope-dilution mass-spectrometry measurement.

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