

Independent fission yields of Rb and Cs from ^{238}U induced by fission-spectrum neutrons*

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The relative independent yields of Rb and Cs from ^{238}U fission induced by fission-spectrum neutrons have been determined by means of a mass spectrograph on line with a nuclear burst reactor. The spectrograph ion source is a tantalum oven containing a target of high purity ^{238}U in a graphite matrix. After irradiation, the radioactive fission products are deposited on the focal plane of the spectrograph and each mass position is analyzed quantitatively from its β activities. Independent yields were calculated by normalizing the measurements using chain yield values and fractional independent yields that were calculated from recently measured cumulative yields for Kr and Xe. The results indicate a charge deviation from an unchanged charge distribution of 0.41 ± 0.07 nuclear charge units, a scaling factor of 1.27 for the number of neutrons associated with ^{238}U fission over the prompt number from thermal-neutron fission of ^{235}U for these fission products, and values of σ for the isobaric charge distribution of 0.49 ± 0.04 and 0.60 ± 0.02 for Rb and Cs, respectively. An odd-even Z effect is noticeable in that the fractional independent yields are generally lower by $(20 \pm 10)\%$ for both Rb and Cs than the mean values estimated from a smooth theoretical model. The sum of the estimated independent yields for the Rb isotopes agrees within error with that for the isotopes of the complementary element Cs.

NUCLEAR REACTIONS $^{238}\text{U}(n, f)$, $E =$ fission spectrum; measured relative abundances; deduced independent fission yields, charge deviation, odd-even Z effect. Mass spectrograph, burst reactor.

INTRODUCTION

The systematics of fission product yields were developed originally in terms of mass-chain yields from fission of ^{235}U , ^{239}Pu , and ^{233}U by slow neutrons.^{1,2} This was encouraged by the fact that radiochemical data first became available in the form of cumulative yields of stable and long-lived nuclides from their shorter-lived precursors. As more data became available, the systematics were extended to describe the distribution of charge within each mass chain³⁻⁷ as Gaussian about a most probable charge Z_p , and with a standard deviation σ that is more or less constant for all mass chains. The value of Z_p is shifted from the value Z_{ucd} for an unchanged charge distribution in which the ratio of protons to atomic mass number for the fission fragment before neutron emission is the same as in the original fissioning nucleus.

With the advent of recent on-line mass spectrographic techniques,⁸⁻¹³ data are becoming available for the independent yields of short-lived isotopes of certain elements formed from various nuclear reactions. This work reports a method for measuring relative independent fission yields on-line which employs a burst reactor and the Bernas on-line surface-ionization technique. The relative abundances of Rb and Cs isotopes produced from fission of ^{238}U induced by essentially fission-spec-

trum neutrons have been measured by this method. A set of values for the independent yields of these isotopes have been calculated from these ratios.

Since it is still popular to regard the fission product yield systematics principally in terms of isobaric concepts, the results of this experiment are analyzed with the aid of a theoretical model that relates these isotopic results to isobaric parameters. Rb and Cs are complementary elements in binary fission of uranium, and therefore the combined data is more significant than the data from either element alone.

EXPERIMENTAL

The on-line fission yield measurement method employed at the Los Alamos Scientific Laboratory uses a mass spectrograph equipped with a modified Bernas type surface ionization source⁹ placed near the Godiva-IV critical assembly;¹⁴ see Fig. 1. The reactor is a portable assembly using about 66 kg of uranium highly enriched in ^{235}U . It produces a short but intense burst of up to 10^{17} neutrons in a $30\mu\text{s}$ interval. The neutron energy distribution is only slightly degraded from that of a pure fission spectrum. For this work, the reactor was placed about 30 cm from the fissionable target material in the spectrograph source.

The spectrograph is a 51-cm radius, 90° broad-range instrument of the type first described by

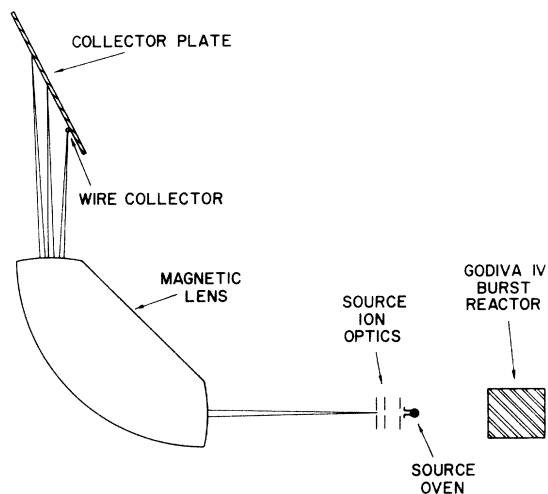


FIG. 1. Schematic of the on-line mass analysis experiment.

Browne and Buechner.¹⁵ The source chamber is water cooled and designed to use the Nier-type thick lens ion optics as modified by Dietz¹⁶ to include Z focus.

The ion source is pictured in Fig. 2. It is an oven which is constructed from 51- μm thick tantalum sheet and heated by an alternating electric current. The oven encloses the target which is a solid right circular cylinder. The orifice of the oven forms a channel with a large depth-to-width ratio to enhance the probability of ionization from contact with tantalum. The structure is spot welded to two 2.03-mm-diam molybdenum electrodes.

The target is a 3.6-mm-diam cylinder 9.5-mm long. It contains approximately 300 mg of high purity ^{238}U ($< 0.025\%$ ^{235}U) in a graphite matrix. It was fabricated by first extruding a mixture of uranium oxide, graphite, and furfuryl alcohol as a binder through a circular die and then curing it at high temperature in an inert atmosphere. The result was a target material in the form of a hard, thin rod that was subsequently cut to the proper length for the oven.

The collector plate at the focal surface of the spectrograph is lined with a 13- μm -thick, 5-cm-wide Mylar strip before each irradiation. A fiducial point on the focal plane is provided by an ion-beam-collector electrode consisting of a straight wire suitably located flat against the Mylar and oriented normal to the drawing in Fig. 1. A mass-marker solution containing equal molecular concentrations of BaCl_2 and SrCl_2 was introduced into the source oven and dried. The number of natural Ba and Sr atoms introduced was over 10 orders of magnitude greater than that produced from any

single irradiation. The purpose was to focus a 4.5- to 5-kV ion beam of natural Ba or Sr nuclides of desired mass onto the wire collector.

The reactor and spectrograph are located in a building especially constructed to provide radiation shielding and located 200 m from the control room. Closed-circuit television is used to read meters near the equipment and to view the operation of the reactor.

The procedure for an irradiation and mass analysis is the following: The source oven temperature is raised and maintained at about 1880°C by regulating the primary current of the isolation transformer. The power dissipated in the spectrograph-source chamber at this temperature is over 200 W. The accelerating voltage is regulated to direct an ion beam current of known mass at the wire collector (using $^{87}\text{Sr}^+$ or $^{137}\text{Ba}^+$ at this time). The reactor is brought to critical. The nuclides of alkali elements produced from fissions in the target quickly diffuse out of the graphite and ionize by contact with tantalum. The ions are mass separated by the magnetic lens and deposited onto the Mylar that lines the collector plate. Collection is terminated by switching off the accelerating voltage at a short, preset time after the irradiation. The collection time was varied from 0.1 to 20 s for 14 irradiations in all. The collector plate is retrieved about 10 min later, when the radiation around the reactor has decreased to a safe level. The Mylar strip is removed and cut

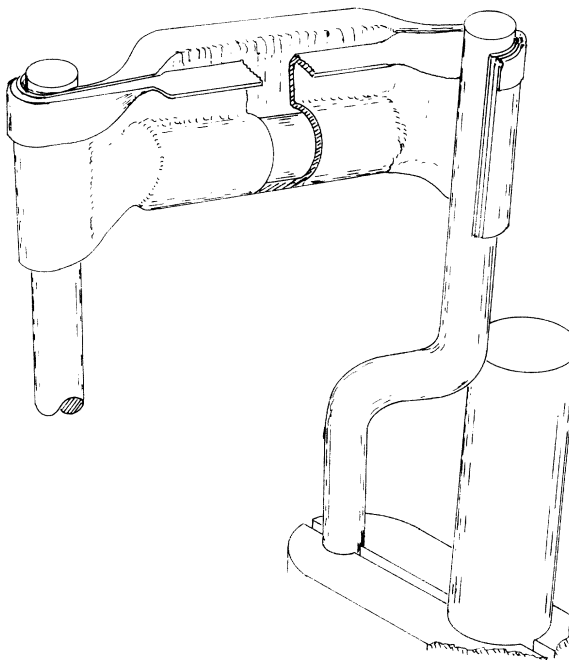


FIG. 2. Ion source oven containing the target.

TABLE I. Summary of relative and absolute independent fission yields of Rb and Cs from ^{238}U fission induced by fission-spectrum neutrons.

Nuclide	Relative abundance	Fractional independent yield ^a (%)	Chain yield ^b (%)	Independent fission yield (%)
^{89}Rb	0.040 ± 0.021	0.4 ^{+0.5} _{-0.4}	3.02 D	0.113 ± 0.063
^{91}Rb	0.338 ± 0.086	15.6 ^{+1.0} _{-2.3}	4.51 E	0.95 ± 0.30
^{92}Rb	0.404 ± 0.087	40.2 ^{+2.7} _{-5.3}	3.91 G	1.14 ± 0.32
^{93}Rb	1.00 ± 0.15	58.9 ^{+5.8} _{-6.8}	4.83 G	2.82 ± 0.65
^{94}Rb	0.91 ± 0.17		5.14 G	2.56 ± 0.66
^{95}Rb	0.60 ± 0.11		5.58 D	1.69 ± 0.43
^{97}Rb	0.0331 ± 0.0066 ^c		5.96 E	0.093 ± 0.025
^{138}Cs	0.033 ± 0.024		5.92 E	0.058 ± 0.042
^{139}Cs	0.198 ± 0.034	5.18 ^{+0.50} _{-0.81}	5.32 F	0.340 ± 0.087
^{141}Cs	1.00 ± 0.11	40.0 ^{+1.7} _{-5.7}	5.45 G	1.72 ± 0.38
^{142}Cs	1.10 ± 0.14		4.70 F	1.90 ± 0.43
^{143}Cs	1.06 ± 0.17		4.53 D	1.82 ± 0.45
^{145}Cs	0.298 ± 0.034 ^c		3.74 D	0.51 ± 0.11
^{146}Cs	0.065 ± 0.014 ^c		3.39 D	0.112 ± 0.033

^a Computed from the cumulative fractional yield of the parent nuclide (Ref. 23). A complete list appears in Table II.

^b Values and precision levels taken from Ref. (25). The precision used in the computation was taken as the middle of the range indicated by each letter, i.e., for C, 2%; D, 3%; E, 6%; F, 12%; and G, 24%.

^c These values are uncorrected for delayed neutron emission (see section on "Data analysis").

midway between mass positions; the β -decay activity in each mass position is followed with proportional counters for several days. A set of relative isotopic abundance ratios is obtained from analysis of the data.

DATA ANALYSIS

A function of activity versus time was constructed for each mass chain from decay data¹⁷ to represent the count rate per parent atom of Rb and Cs as follows: If a number N of parent atoms are formed at burst time, which decay by a series of β emissions, one can compute the number $N_i(t)$ of atoms of the i th descendant that will be present at a time t later.¹⁸ This descendant decays at a rate $\lambda_i N_i(t)$, where λ_i is the respective decay constant. The β -ray detection efficiency of a proportional counter is energy dependent at lower energies and increases to an asymptotic maximum at higher energies.¹⁹ If the weighted average efficiency for the β emissions from the i th descendant is ϵ_i , the contribu-

tion to the count rate from this source is $\epsilon_i \lambda_i N_i(t)$, and the total count rate one observes is the sum from all the contributing descendants that are present at time t . (In those decay processes where the β -decay schemes are not yet known, the efficiency of the counter was estimated as that for β decay to the ground level.) Each function is a sum of products of exponentials with constant coefficients, and it differs in appearance, fortunately, from that of its neighboring mass chains.

In constructing the functions for the specific activities from ^{94}Rb and ^{95}Rb , the effect from branching due to delayed neutron emission from these precursors was also included.²⁰ The available, measured values for delayed neutron emission probability from the other Rb and Cs precursors were considered too small for their effects to be included. Delayed neutron probabilities for ^{145}Cs and ^{146}Cs are unknown but could be appreciable. There is evidence that it is appreciable for ^{97}Rb although not known precisely.²¹ Effects from these precursors were not included.

If perfect mass resolution existed, it would be

sufficient to fit the count-rate data from one mass location by adjusting the amplitude of the respective function for that mass chain, correcting for decay during the time for each count. However, the ionization produced by the reactor burst causes the source electrode voltages to change momentarily. These recover quickly, but the result at the collector is a noticeable spill of ions of one mass onto the mass position of the next lower mass. Therefore, where necessary, the data from each mass position was analyzed by including the function of the next higher mass.²² Having the contribution to the activity by a mass chain at its mass location and also in the next lower location, the net abundance was taken as the sum of the two amplitudes for that mass chain. Spillage displaced by two mass locations (i.e., ^{91}Rb at the ^{89}Rb location) was negligible.

The assays for ^{145}Cs and ^{146}Cs showed some mass-145 activity at the mass-146 position. The respective amplitudes for each of these mass chain functions were added regardless of location. However, the presence of the mass-145 activity at mass 146 may have been due more likely to delayed neutron emission from the mass-146 chain precursors than to poor resolution or to uncertainty of mass location so far from the fiducial point.

The collector is also designed to move at a steady rate in a direction transverse to the beam spectrum. The reactor burst initiates the motion. The resulting beam trace at the collector describes the ion emission from the source as a function of time. Sections of traces obtained in this way were analyzed, and the information made minor corrections possible for in-growth and decay of Cs in the source during the finite collection time. Data from long collection times that required large ingrowth corrections for Rb were not used. No decay corrections were necessary in the case of Rb since it was found to diffuse very quickly.

RESULTS

The averaged relative isotopic abundances of Rb and Cs from ^{238}U fission induced by fission-spectrum neutrons are listed in Table I. They are normalized to the ^{93}Rb and ^{141}Cs abundances. The listed errors include the statistical standard deviations of the averages (from seven irradiations for each element with different collection times) plus 10% of the average value to include possible systematic errors. From the method of analysis, only the relative abundances of ^{97}Rb , ^{145}Cs , and ^{146}Cs are uncorrected for delayed neutron effects.²⁰ No information was available for corrections from further branching due to delayed neutron emission along the decay chains concerned in

this work, but such branching is likely to be smaller from nuclides closer to stability. Values for the isotopes with masses 90, 96, 140, and 144 are missing because the β decay rates in those mass chains were too small to yield information.

The estimates for the independent fission yields in Table I, or those fractions of the fissions that result in the formation of these isotopes after prompt neutron emission, are made by normalizing the relative abundances to radiochemical information as follows:

(1) The fractional cumulative yield, i.e., the probability of forming a particular nuclide and all of its precursors, has recently been measured for several Kr and Xe isotopes from ^{238}U fission induced by fission spectrum neutrons.²³ Fractional independent yields (FIY), or the probability of forming only that particular nuclide in a given mass chain, were calculated for Rb and Cs from Ref. 23 in cases where the contribution to the mass chain by the Rb and Cs daughters is small. The fractional cumulative yield of the parent nuclide was subtracted from unity, and the difference was corrected for the comparatively small fractional part contributed by the daughter nuclide which was inferred from systematics.²⁴ For example, the measured fractional cumulative yield of ^{91}Kr is $0.84_{-0.01}^{+0.02}$. Therefore the maximum possible FIY for ^{91}Rb is $0.16_{-0.02}^{+0.01}$. If one assumes a Gaussian charge distribution with a standard deviation of 0.56 ± 0.06 (Ref. 24), one computes that only slightly more than 2% of this difference is attributable to ^{91}Sr and nuclides of higher Z in this mass chain. Therefore, Table I shows a value of 15.6% for the FIY of ^{91}Rb . The accuracy is estimated from error propagation. These FIY results are included in Table I (a more complete list is provided in Table II).

(2) Several independent yield estimates for each element were obtained by multiplying each FIY by its mass-chain yield,²⁵ also included in Table I. These were used to normalize the relative abundances. The normalized independent fission yields are listed in the last column of Table I.

DISCUSSION

Current isobaric systematics describe the probability of forming a fission product of mass number A and nuclear charge Z as Gaussian along that mass chain and centered about a most probable charge Z_p . The fractional cumulative yield for this nuclide has therefore been expressed⁴ by:

$$\sum_0^Z (P_n) = \sigma^{-1} (2\pi)^{-1/2} \int_{-\infty}^{Z+0.5} \exp[-0.5(n - Z_p)^2 \sigma^{-2}] dn \quad (1)$$

Experimental data have been more readily available from thermal-neutron fission of ^{235}U than from ^{238}U fission (Ref. 7). In thermal-neutron fission, most of the observed values for σ lie in a range that is small within a fragment group and centers about 0.56 nuclear charge units. The values for Z_p far from symmetric fission show a shift of about 0.45 from Z_{ucd} such that the lighter fragment tends to be richer in protons than the heavier one. An odd-even effect has also been reported where the yield of nuclides of even- Z elements is enhanced and that of odd- Z elements is reduced.^{6,7,12} According to a recent survey by Amiel and Feldstein,²⁶ the yields deviate from the smooth function of Eq. (1) by $\pm 25\%$. This refinement is now included in calculated tabulations of fractional fission yields.²⁴

The model

The systematics for ^{238}U fission are adopted to analyze the abundance data from this experiment. If odd-even effects are ignored (they will be shown later to exist), the FIY of a nuclide can be expressed from Eq. (1):

$$F(Z, A) = \sigma^{-1} (2\pi)^{-1/2} \int_{Z-0.5}^{Z+0.5} \exp[-0.5(n - Z_p)^2 \sigma^{-2}] dn \quad (2)$$

When considering the fission yields of nuclides of the same element, the integrand of Eq. (2) can be regarded as a function of A only, if Z_p (and strictly speaking also σ) is regarded as a function of A . We note also that Eq. (2) is normalized so that the sum of all the FIY's in a mass chain is unity.

The Z_p function is approximated by:

$$Z_p = \frac{Z_f}{A_f} [A + \nu(A)] + C, \quad (3)$$

where Z_f/A_f is the charge-to-mass ratio of the fissioning nucleus, and $\nu(A)$ is the average number of neutrons emitted from the fragment of post-neutron-emission mass A . The quantity C is the charge deviation and is assigned the subscripts l or h to denote the light or heavy fragment group. We assume in this analysis that σ and C are constant over a short mass range and also that the value of C_l that is observed for Rb is the reflection of C_h from the complementary nucleus, Cs. Detailed values for $\nu(A)$ are not yet available for the separate ^{239}U fission fragments. If one assumes that neutron emission is similar to that from thermal-neutron fission of ^{235}U , this process can be described approximately by

$$\nu(A) = g \nu^{\text{ref}}(A), \quad (4)$$

where $\nu^{\text{ref}}(A)$ is the average number of neutrons that are emitted by the fragment of post-neutron-emission mass A from thermal fission of ^{235}U (see Ref. 27 and Table II of Ref. 7), and g is a scaling factor.

Analysis

A set of *relative* FIY values $Y(A)$ can be generated for each element by dividing the relative abundance by the mass chain yield and normalizing to a chosen isotope. If the FIY value of the chosen isotope is denoted by f , the values for the others become the products $fY(A)$. Estimates for σ , C , and f are obtained by minimizing the summation²²

$$S = \sum P(A) [fY(A) - F(Z, A)]^2 \quad (5)$$

over all values of $Y(A)$. $P(A)$ is the weight assigned to $Y(A)$ and estimated from the propagated errors.

A series of solutions of Eq. (5) were carried out for Rb and Cs separately in which the scaling factor was varied from $g=1.0$ to $g=1.5$. The values for f and σ were nearly insensitive to the scaling factor, but the values for C depended strongly upon it, as shown in Fig. 3. Since Rb and Cs are

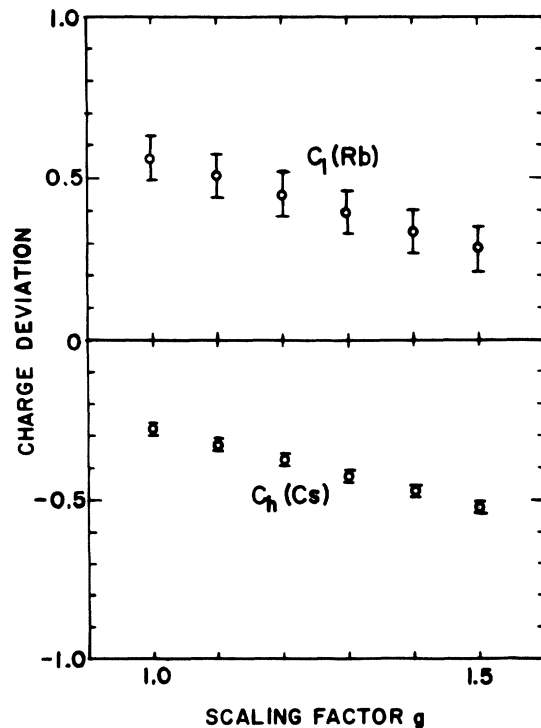


FIG. 3. Dependence of the value for charge deviation C on the scaling factor g ; see Eq. (4).

TABLE II. Comparison of fractional independent yields with the adjusted products $fY(A)$ and the fitted model.

Nuclide	$fY(A)$ ^a (%)	From the fitted model ^b (%)	From experiment ^c (%)
^{89}Rb	0.045 ± 0.024	0.008 ± 0.007	$0.4^{+0.5}_{-0.4}$
^{91}Rb	25.5 ± 6.7	21.4 ± 5.9	$15.6^{+1.0}_{-2.3}$
^{92}Rb	35 ± 11	48.9 ± 8.0	$40.2^{+2.7}_{-5.8}$
^{93}Rb	70 ± 20	69 ± 11	$58.9^{+5.6}_{-6.8}$
^{139}Cs	8.7 ± 1.6	7.82 ± 0.92	$5.18^{+0.50}_{-0.61}$
^{140}Cs		21.9 ± 1.7	$13.8^{+1.0}_{-2.2}$
^{141}Cs	43 ± 11	42.6 ± 2.2	$40.0^{+1.7}_{-5.7}$

^a Adjusted products $fY(A)$ from Eq. (5).

^b $F(Z, A)$ values of the fitted model obtained from Eq. (2) using the estimates for C , σ , and g , Eqs. (6).

^c Computed from the fractional cumulative yields of the parent nuclide (Ref. 20).

complementary, it is assumed that the value for g is that for which $C_h = -C_l$. The adjusted parameters for the fitted model become:

$$\begin{aligned}
 g &= 1.27 \pm 0.07, \\
 |C| &= 0.41 \pm 0.7, \\
 \sigma_l &= 0.49 \pm 0.04 \text{ for Rb}, \\
 \sigma_h &= 0.60 \pm 0.02 \text{ for Cs}.
 \end{aligned}
 \tag{6}$$

The listed errors are statistical 1- σ values. Similar solutions with the yields of ^{97}Rb , ^{145}Cs , and

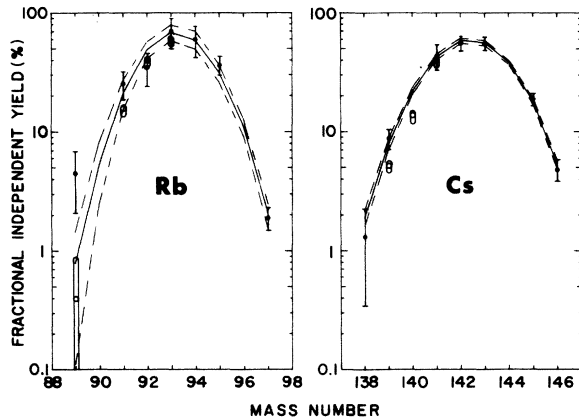


FIG. 4. Comparison of the lower Rb and Cs FIY values computed from radiochemical data (open bars) with the adjusted products $fY(A)$ and the $F(Z, A)$ values from the fitted model. The adjusted products are plotted as dots. The solid curve is the fitted model, and the dashed curves indicate the associated standard deviations of the mean.

TABLE III. Estimates using systematics of independent fission yields for those Rb and Cs nuclides that could not be observed by this experimental technique.

Nuclide	Fractional independent yield ^a (%)	Chain yield ^b (%)	Estimated independent yield (%)
^{90}Rb	4.3 ± 2.2	3.28 E	0.141 ± 0.073
^{96}Rb	8.8 ± 1.7	5.50 G	0.48 ± 0.11
^{140}Cs	$13.8^{+1.0}_{-2.2}$	5.95 C	$0.82^{+0.06}_{-0.13}$
^{144}Cs	30.8 ± 4.1	4.54 D	1.40 ± 0.19

^a The value for ^{140}Cs is taken from Table II. The others are 0.8 times the $F(Z, A)$ values computed from the fitted model.

^b See footnote b, Table I.

^{146}Cs altered to represent estimated corrections for effects from delayed neutrons (see Refs. 20 and 28) show no appreciable difference in these results.

The charge deviation $C = Z_p - Z_{\text{ucd}}$ and the average value for σ above conform with the values observed for fission yields from ^{235}U . However, these results show an asymmetry in that the distribution of charge for the light fragments is smaller than for the heavy fragments. The value for g seems high. An average effective fission-spectrum neutron energy of about 2.8 MeV is obtained by weighting the neutron energy by the product of the fission-neutron energy spectrum (Ref. 5, p. 39) and the fission cross section for ^{238}U at that energy.²⁹ According to Davey,³⁰ the average number of neutrons $\bar{\nu}(E)$ from all the fission products is 2.68 per fission for this neutron energy. For thermal-neutron fission of ^{235}U , this number is 2.41, and the value one would expect for g is 1.11. The discrepancy between this measurement (1.27) and the expected value, if real, concerns the over-all average number of neutrons per fission compared with that from Rb and Cs, specifically. Direct inspection of the mass values where the relative abundances peak (Table I) also suggest that the number of neutrons emitted per fission is nearer to 3.1 than to 2.68 for this fission-product pair.

The values for the adjusted products $fY(A)$ and those from the fitted model are listed in Table II. These two sets of values are related to each other by Eq. (5) and systematics that assume no odd-even Z effect. The latter are computed from Eq. (2) using the estimates for C , σ , and g from Eqs. (6). The FIY values computed from radiochemical measurements²³ are also listed. The values deduced from measurement are generally lower by about $(20 \pm 10)\%$ for both elements. Therefore, there appears to be a suppression of the indepen-

dent yields of Rb and Cs (odd- Z elements), also from fission of ^{238}U by neutrons near threshold. The effect is visible in Fig. 4. The adjusted $fY(A)$ products are plotted as dots, the solid curve is the fitted model, and the dashed curves are the associated standard deviations of the predicted mean. The open circles and error bars are the FIY values computed from radiochemical measurements and which were used to normalize the independent fission yields.

As a check for the method used to normalize the abundances in Table I, the sum of the yields of the isotopes of these complementary elements should be equal. The independent fission yields for the missing isotopes were estimated as follows: (1) When unavailable from radiochemical information, the FIY for the missing isotope was computed from the fitted model and then multiplied by 0.8 for the odd-even effect. (2) The FIY was multiplied by the mass-chain yield to obtain the independent fission yield. These calculations and propagated error estimates are summarized in Ta-

ble III. The sums of the independent fission yields from Tables I and III are $(10.0 \pm 1.1)\%$ for Rb and $(8.7 \pm 0.8)\%$ for Cs. This is good agreement in view of the uncertainties involved in the estimates.

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