Nuclear Charge Distribution in Fission: Y^{92}, Y^{93}, Y^{94}, and Y^{95} Independent Yields*

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The fractional independent yields of Y^{92}, Y^{93}, Y^{94}, and Y^{95} from thermal-neutron fission of U^{235} have been determined to be 0.0013 ± 0.0002, 0.016 ± 0.004, 0.08 ± 0.02, and 0.29 ± 0.08, respectively. The experimental method involved rapid separation of yttrium and strontium fission products by precipitation of yttrium hydroxide and subsequent radiochemical determination of the yttrium isotopes. Newly determined half-life values are 84 ± 5 sec for Sr^{94} and 33 ± 6 sec for Sr^{95}. Gaussian curves which satisfactorily represent the nuclear charge dispersion for $A=92$, 93, 94, and 95 have standard deviations $\sigma=0.58\pm0.02$, 0.56 ± 0.02, 0.56 ± 0.04, and 0.53 ± 0.02, respectively. The average value of σ for ten mass numbers ($A=91$–95 and 139–143) is 0.59 ± 0.02. For the individual mass numbers, a single Gaussian curve with $\sigma=0.59\pm0.06$ (full width at half-maximum $=1.56\pm0.12$ charge units) gives a satisfactory representation of nuclear charge dispersion.

I. INTRODUCTION

THE distribution of nuclear charge among fission products with the same mass number after neutron evaporation—called nuclear charge dispersion—has been investigated[1] for five mass numbers among the heavy products, but for only one mass number among the light products from thermal-neutron fission of U^{235}. Since the fractional cumulative yields of Kr^{92}, Kr^{94}, Kr^{94}, and Kr^{95} have been measured,[1,2] determination of the fractional independent yields of Y^{92}, Y^{93}, Y^{94}, and Y^{95} should give information about the charge dispersion for four more mass numbers of light products. This article describes these independent yield measurements. The next article[3] describes related measurements of the independent yields of Rb^{92} and Rb^{93}.

The radioactive decay chains of interest are shown in Fig. 1,[4–9] together with the fractional yields that have been determined.

II. METHODS

A. Experimental Procedure

Solutions of uranium (VI) enriched to 20 or 93% in U^{235} were irradiated in the thermal-neutron flux produced at the Washington University cyclotron; then yttrium was separated chemically from strontium, purified from other fission products, and measured for radioactivity with beta-proportional counters.

A solution, after irradiation in a pneumatic rabbit cartridge,[1] was divided into two portions; the separation from strontium was carried out immediately on one portion and much later on the other, after substantial decay of a precursor strontium isotope. The separation involved precipitation and filtration of $Y(OH)_3$ from an ammonia solution containing $(Sr^{85})^{2+}$ tracer, Sr^{2+} carrier, and NH_3OHCl, the last reagent preventing precipitation of U^{VI}. The procedures used are similar to the one used for separating lanthanum from barium[1]; they are described in detail elsewhere.[10] Ten to twenty percent of the strontium remained with the $Y(OH)_3$ through the filtration. Immediately afterward an ammonia solution was used to wash out the bulk of the remaining strontium. About 1% of the original strontium remained with the $Y(OH)_3$ after the wash, and correction for this amount was made through the use of the Sr^{85} tracer. The effective time of the strontium-yttrium separation was taken to be the time when the filtration, which required about 10 sec, was approximately 60% complete. The details of the separation-time estimates are given elsewhere[10]; the uncertainty in the estimates is believed to be about 2 sec.

The yttrium purification procedures involved extraction of Y^{III} into a toluene solution of di-(2-ethylhexyl)-orthophosphoric acid and a series of precipitations of YF_3, $Y(OH)_3$, and $Y_2(C_2O_4)_3$. The procedures were patterned after the one developed by Wolfsberg[11]; they are described in detail elsewhere.[10]

Yttrium samples were mounted for radioactive measurement either as the oxalate or oxide; chemical yields were determined by weighing the oxide either before or after the radioactivity measurements. Samples from the two portions of an irradiated solution were

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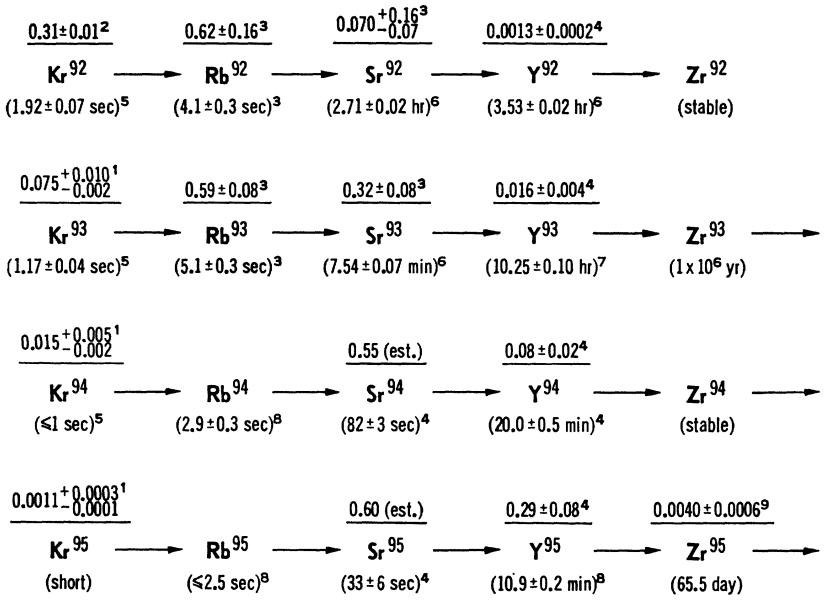

FIG. 1. Radioactive decay chains. Underlined quantities over nuclide symbols are experimental fractional cumulative yields for Kr isotopes and fractional independent yields for other nuclides. Quantities in parentheses under symbols are half-lives. Superscripts refer to the footnotes.

mounted in the same way and measured with the same beta-proportional counter. Empirically determined self-absorption and coincidence corrections were applied when appropriate.

B. Yttrium-Decay Data

Radioactive-decay data were resolved by the method of least squares using Cumming's program[12] modified for use with an IBM 7072 computer. The half-life of at least one component was allowed to vary; agreement with published values was taken as evidence both that the resolution was properly carried out and that the samples were radiochemically pure. A comparison of half-life values is given in Table I.

The published half-life values were used in subsequent calculations, except that for Y^{94} the value 20.0±0.5 min was adopted. An unpublished value of 19.5 min[11] helps to support a somewhat lower value than the published one.

C. Independent-Yield Calculations

Calculations of independent yields were made with equations previously developed,[1] except that provision was made throughout to correct for the contamination of yttrium with strontium after the initial separation. Thus, for determination of both the half-life of Sr^{94} or Sr^{95} and the independent yield of Y^{94} or Y^{95}, suitable approximations are applied to Eq. (1) of Ref. 1 to give the following equation:

$$\ln(1-Q)\exp(-\lambda_C\tau)=\ln(S)-\lambda_B\tau, \quad (1)$$

$$S=\frac{(\lambda_A-z\lambda_A-y\lambda_B)}{\lambda_A-\lambda_B}\{1-R+R\exp(-\lambda_B\Delta t)\}. \quad (1a)$$

[12] J. B. Cumming, U. S. At. Energy Comm. NAS-NS-3107, 1963, p. 25.

The definitions of the symbols are the same as in Ref. 1, with subscripts A, B, and C applying, respectively, to the Rb, Sr, and Y members of a radioactive decay chain. The definitions are repeated here for convenience:

λ = radioactive decay constant.

y, z = fractional independent yields of B and C, respectively, based on the cumulative yield of C.

T = duration of irradiation producing fissions at a constant rate.

t, t' = time intervals between the end of an irradiation and the separation of adjacent chain members for the two samples from the same irradiation.

$\tau=0.5T+t$; $\tau'=0.5T+t'$.

Q = ratio of activity of C separated after t to activity of C separated after a longer time interval t', corrected for chemical yield, to the same number of fissions, and for decay to the same time.

R = fraction of B remaining with nuclide C after initial rapid separation of C from B.

Δt = time interval between initial rapid separation of B from C and subsequent separation (assumed to be complete).

The approximation is made that krypton nuclides and their precursors can be neglected, i.e., that the history of growth of an yttrium nuclide can be described

TABLE I. Yttrium half-life values.

Isotope	No. of determinations	Weighted average half-life	Published half-life	Reference
Y^{92}	20	3.54±0.01 h	3.53±0.02 h	6
Y^{93}	5	10.25±0.01 h	10.25±0.10 h	7
Y^{94}	21	19.9 ±0.5 min	20.35±0.20 min	8
Y^{95}	21	10.9 ±0.1 min	10.9 ±0.2 min	8

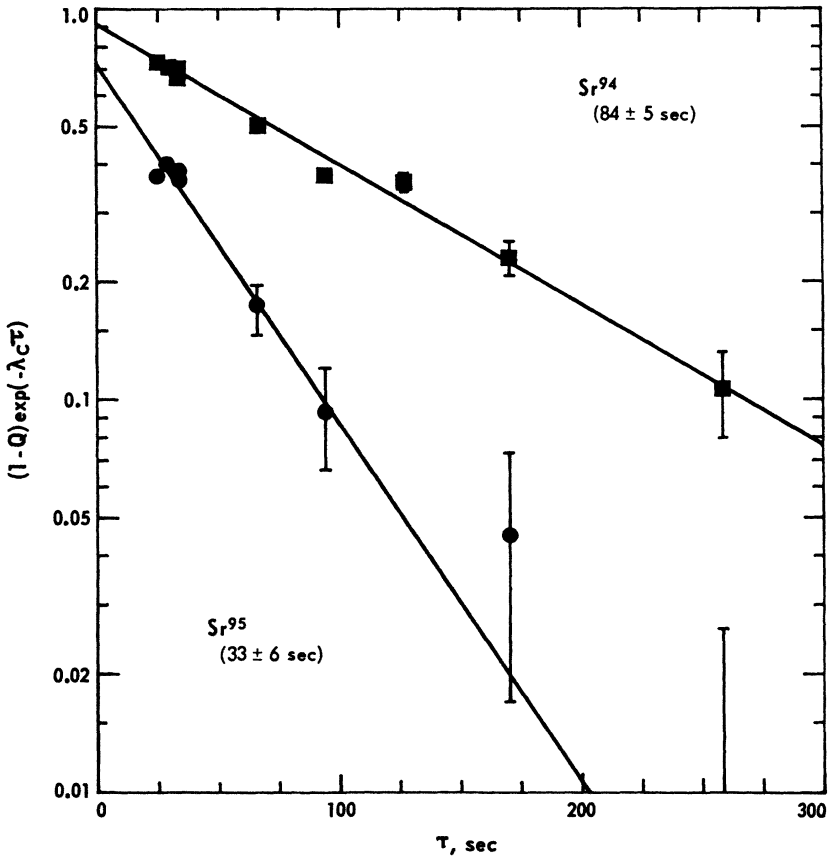

FIG. 2. Half-lives of Sr^{94} and Sr^{95}. Points are experimental; error bars represent an estimated uncertainty in Q of $\pm 3\%$. The lines represent Eq. (1) with parameters derived from the least-squares analysis. (See Refs. 13 and 14.)

adequately by the equation for a three-membered chain. Tests of the approximation with the equation for a four-membered chain showed that assuming zero for the half-lives of the krypton nuclides introduces no appreciable error. We believe that making a similar assumption for krypton precursors has the same result, because their half-lives are expected to be short and their yields low.

III. RESULTS

A. Strontium Half-Lives

The half-live values for Sr^{92} and Sr^{93} reported in Refs. 6 and 7 appear to be sufficiently reliable to eliminate any need for their redetermination in this work. The Sr^{94} and Sr^{95} half-lives are relatively more important in calculating yttrium yield values. Therefore, some Y^{94} and Y^{95} independent-yield data were taken at timed intervals to permit checking the strontium half-lives. The data appropriate for Eq. (1) are shown in Fig. 2.[13,14] The 83.6 ± 5.2 sec half-life of Sr^{94} calculated from the slope of the line agrees with the published values, 81.6 ± 3.6 sec[8] and 78 ± 12 sec.[7] The three values were weighted by the square of the reciprocal of their uncertainties, and the weighted average value of 82 ± 3 sec is used in subsequent calculations. For Sr^{95}, the half-life calculated from the yield data is 33 ± 6 sec, appreciably shorter than the published values of 48 ± 9 sec[8] and ~40 sec.[15] Since more timed separation data were used in calculating the 33-sec value than were reported in the other references, this value is used subsequently for the Sr^{95} half-life.

B. Yttrium Independent Yields

The independent-yield data were used in two ways, depending on whether the strontium half-life values were to be measured as well as the yields and whether the time interval t' was long enough for the approximations to be valid that were made in deriving Eq. (1).

Method A. The intercepts at $\tau=0$ of the lines in Fig. 2 are $S=0.911\pm0.018$ for Y^{94} and $S=0.71\pm0.09$ for Y^{95}. The fractional independent-yield values calculated from the intercepts and Eq. (1a) are 0.095 ± 0.030 and 0.29 ± 0.10 for Y^{94} and Y^{95}, respectively.[16] If the average

[13] Points for Y^{95} at $\tau=127.5$ and 259.0 sec are not shown because the Q values are negative, -0.036 and -0.014, respectively. These values were included in the least-squares analysis (see Ref. 14).

[14] W. R. Busing and H. A. Levy, Oak Ridge National Laboratory Report No. ORNL-TM-271, 1962 (unpublished).

[15] D. C. Hoffman, J. D. Knight, D. L. Frasco, and B. J. Dropesky (unpublished); D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. **30**, 661 (1958).

[16] Other parameters used in the calculation are: half-lives and uncertainties from Fig. 1, y for $Sr^{92}=0.55_{-0.55}{}^{+0.37}$ (estimated), y for $Sr^{93}=0.6_{-0.6}{}^{+0.1}$ (estimated), $R=0.010_{-0.007}{}^{+0.016}$ (average value and limits), $\Delta t=193_{-40}{}^{+131}$ sec (average value and limits). An uncertainty in τ of 2 sec is assumed.

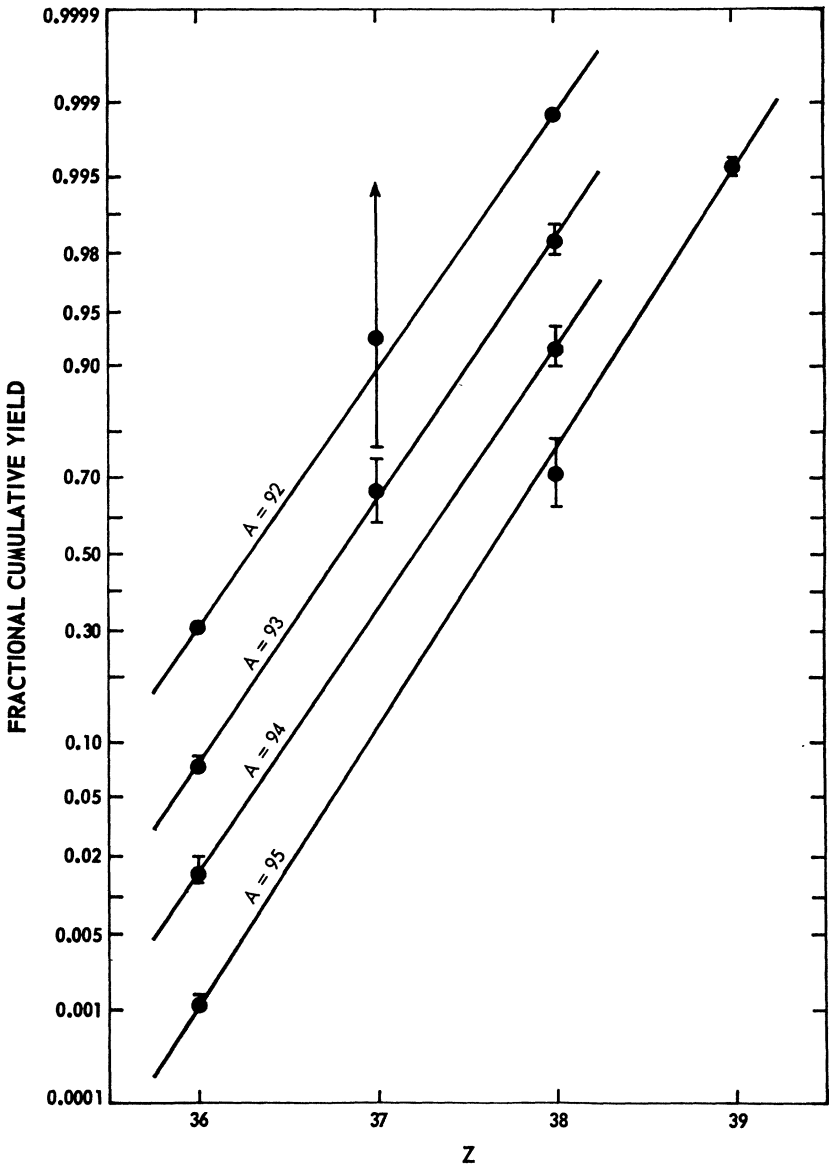

FIG. 3. Charge dispersion curves for mass numbers 92, 93, 94, and 95. Data are listed in Table III.

half-life value of 82±3 sec is used for Sr^{94}, the intercept at $\tau=0$ of the line representing the least-squares analysis of the data is $S=0.916\pm0.021$, and the calculated independent yield is 0.090 ± 0.030.

Method B. Equation (1) of Ref. 1 was employed with the half-lives and the Sr independent yields shown in Fig. 1. The Sr^{94} and Sr^{95} independent yields had to be estimated, but allowance was made in the calculations for the maximum possible uncertainty ($y=0$ to $1-z$). The averages of the calculated values are given in Table II; the uncertainties given include all known experimental uncertainties and are intended to approximate standard deviations.

The selected values for Y^{94} and Y^{95} are averages of the values calculated in Methods *A* and *B*; the values used from Method *B* in the averaging were derived only from data not treated by Method *A*.

It should be noted that the value calculated for the Y^{95} independent yield is very dependent on the value of the Sr^{95} half-life used. If the 48±9 sec half-life value[8] is used, the calculated fractional independent yield is $0.42_{-0.07}{}^{+0.05}$, a value which is appreciably higher than the value of 0.23 derived from the Gaussian charge dispersion curve for $A=95$ (see discussion).

IV. DISCUSSION

Data concerning nuclear charge dispersion for thermal-neutron fission of U^{235} are summarized in Table III, and data for mass numbers 92, 93, 94, and 95 are plotted in Fig. 3. The data are interpreted on the

TABLE II. Fractional independent yields.

Fission product	Method	Percent independent yield ($z\times100$) Calculated[a]	Selected
Y^{92}	*B*	0.127±0.021[b]	0.13±0.02
Y^{93}	*B*	1.59±0.37[c]	1.6±0.4
Y^{94}	*A*	9.0±3.0	8±2
	B	6.1±2.8[d]	
Y^{95}	*A*	29±10	29±8
	B	$29_{-14}{}^{+9e}$	

[a] The uncertainties are estimated to include all known experimental uncertainties, such as those in timing and in half-lives, as well as the standard deviation of the average value. The estimates were made by varying each parameter individually and recalculating the value of z for a typical experiment. The difference between each new z value and the original value was squared, the resulting values were added to the variance of the average value, and the square root of the total was taken to give the uncertainty listed.

[b] The average value of $z\times100$ for 17 determinations is 0.127±0.012. Data for a typical experiment are: $T=18.0$ sec, $t=33.1$ sec, $t'=11\,890$ sec, $\Delta t=406$ sec, $Q=0.0052$, and $R=0.0098$; the resulting value of $z\times100$ is 0.113. Uncertainties of 2 sec in t, 0.002 in R, 0.16 in y, 0.3 sec in the Rb^{92} half-life, and 0.02 h in the Y^{92} half-life cause uncertainties in $z\times100$ of 0.014, 0.006, 0.007, 0.003, and 0.003, respectively. Uncertainties in other parameters cause uncertainties in $z\times100$ of <0.001.

[c] The average value of $z\times100$ for 20 determinations is 1.59±0.16. Data for a typical experiment are as in footnote b, except $Q=0.0733$; the resulting value of $z\times100$ is 1.56. Uncertainties of 2 sec in t, 0.002 in R, 0.08 in y, 0.3 sec in the Rb^{93} half-life, and 0.07 min in the Sr^{93} half-life cause uncertainties in $z\times100$ of 0.30, 0.09, 0.09, 0.05, and 0.06, respectively. Uncertainties in other parameters cause uncertainties in $z\times100$ of <0.01.

[d] The average value of $z\times100$ for 3 determinations that are not treated by Method *A* is 6.1±0.8; for 7 determinations including 4 treated by Method *A*, the average value of $z\times100$ is 7.9±0.7. Data for a typical experiment are: $T=12.6$ sec, $t=32.8$ sec, $t'=252.1$ sec, $\Delta t=458$ sec, $R=0.0026$, $Q=0.340$, $y=0.55$ (estimated); the resulting value of z is 0.059. Uncertainties of 2 sec in t, 2 sec in t', 0.002 in R, 0.55 in y, 0.3 sec in the Rb^{94} half-life, and 3 sec in the Sr^{94} half-life cause uncertainties in $z\times100$ of 1.7, 0.1, 0.2, 2.0, 0.2, and 0.7, respectively. Uncertainties in other parameters cause uncertainties in $z\times100$ of <0.1.

[e] The average value of $z\times100$ for 2 determinations that are not treated by Method *A* is 29±2; for 6 determinations including 4 treated in Method *A*, the average value of $z\times100$ is also 29±2. Data for a typical experiment are as in footnote d, except that $Q=0.675$ and $y=0.60$ (estimated). Uncertainties of 2 sec in t, 6 sec in the Sr^{95} half-life, and 0.60 in y cause uncertainties in $z\times100$ of 2.8, +7.7 or −13.0, and 3.7, respectively. Uncertainties in other parameters cause uncertainties in $z\times100$ of <0.2.

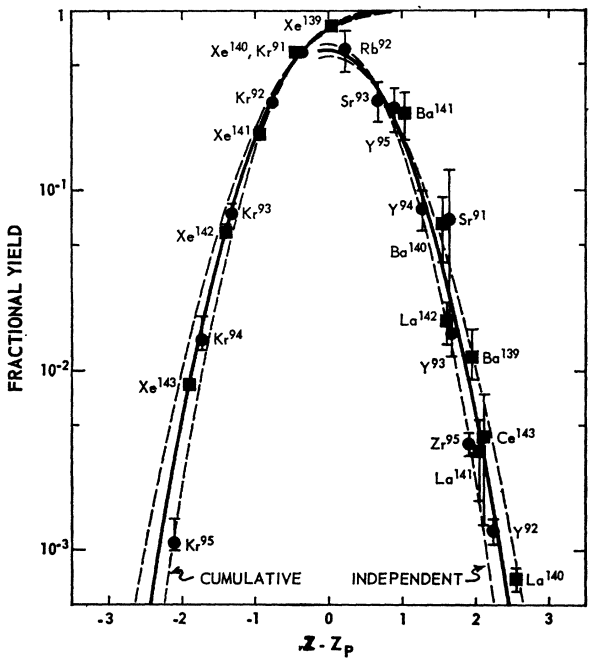

FIG. 4. Charge dispersion data for ten mass numbers. The line is a Gaussian distribution curve with $\sigma=0.59\pm0.06$. Z_P values are chosen to give the best correlation of the curve with the data and are listed in the last column of Table III.

assumption that the charge dispersion is Gaussian, an assumption that is supported by the linearity of the plots in Fig. 3 for $A=92$, 93, and 95 and of similar plots[1] for $A=140$ and 141, the five mass numbers for which three fractional yields have been measured. The values of σ and Z_P listed in Table III are constants that appear in the cumulative form of the Gaussian expression.[1] The data may also be represented by the equation:

$$P(Z)=(c\pi)^{-1/2}\exp[-(Z-Z_P)^2/c], \qquad (2)$$

in which $P(Z)$ is the fractional independent yield of a

TABLE III. Charge dispersion curves.

Fission product	Fractional cumulative yield[a]	σ	Z_P	Z_P[b]
$A=91$		0.81±0.27[c]	36.32±0.09	36.37
Kr^{92}	0.31±0.01[d]			
Rb^{92}	$0.93_{-0.16}{}^{+0.07e}$	0.58±0.02	36.78±0.02	36.78
Sr^{92}	0.9987±0.0002			
Kr^{93}	$0.075_{-0.002}{}^{+0.010c}$			
Rb^{93}	0.664±0.080[e]	0.56±0.02	37.29±0.04	37.32
Sr^{93}	0.984±0.004			
Kr^{94}	$0.015_{-0.002}{}^{+0.005c}$	0.56±0.04	37.71±0.07	37.73
Sr^{94}	0.92±0.02			
Kr^{95}	$0.0011_{-0.0001}{}^{+0.0003c}$			
Sr^{95}	0.706±0.080	0.53±0.02	38.10±0.03	38.15
Y^{95}	0.9960±0.0006[f]			
$A=139$		$0.75_{-0.10}{}^{+0.14c}$	$53.82_{-0.20}{}^{+0.14g}$	54.05
$A=140$		0.68±0.03[c]	54.34±0.03[c]	54.50
$A=141$		0.59±0.03[c]	54.97±0.04[g]	54.97
$A=142$		0.55±0.03[c]	55.36±0.04[e]	55.40
$A=143$		0.60±0.04[c]	$55.92_{-0.10}{}^{+0.08c}$	55.90

Average σ for 5 light mass numbers: 0.57±0.01
Average σ for 5 heavy mass numbers: 0.62±0.03
Average σ for all 10 mass numbers: 0.59±0.02

[a] It is assumed that independent yields are negligible for late chain members, those with Z two units or more larger than the Z of the last member listed.

[b] Z_P value chosen to give the best correlation of the data with the Gaussian curve $\sigma=0.59\pm0.06$.

[c] From Ref. 1.

[d] From Ref. 2.

[e] From Ref. 3.

[f] From Ref. 9.

[g] From Ref. 1, uncertainty corrected.

fission product with atomic number Z, Z_P is the value of Z (not necessarily integral) for which the function $P(Z)$ is a maximum, and c is a constant

$$(c\approx2[\sigma^2+1/12]).^1$$

As can be seen in Table III, the values of σ are approximately constant, which suggests that a single charge dispersion curve might be used to represent all charge dispersion data. The variations in σ that are observed between individual values or between the average values for light and heavy mass numbers are

neither large nor statistically significant. The average value of σ is 0.59 with a standard deviation ± 0.02. For an individual mass number, the uncertainty would be larger; we choose ± 0.06 as a reasonable value. Thus for a general representation of charge dispersion data, a Gaussian curve with $\sigma = 0.59 \pm 0.06$ or $c = 0.86 \pm 0.15$ might be used; such a curve has a full width at half-maximum of 1.56 ± 0.12 charge units. Figure 4 shows that the curve represents very well the fractional-yield data for the ten mass numbers studied.

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Nuclear Charge Distribution in Fission: Rb^{92} and Rb^{93} Independent Yields*

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The fractional independent yields of Rb^{92} and Rb^{93} from thermal-neutron fission of U^{235} have been determined to be 0.62 ± 0.16 and 0.59 ± 0.08, respectively. These values agree with those predicted from Gaussian representations of nuclear charge dispersion for $A = 92$ and $A = 93$. Newly determined half-life values are 4.1 ± 0.3 sec for Rb^{92} and 5.1 ± 0.3 sec for Rb^{93}. The experimental method involved rapid separation of rubidium and strontium fission products by adsorption of strontium on solid strontium carbonate and subsequent radiochemical determination of yttrium descendants.

I. INTRODUCTION

THE distribution of nuclear charge among fission products with the same mass number has been assumed to be Gaussian.[1] The assumption is supported experimentally for thermal-neutron fission of U^{235} by three measured yields for each of the two mass numbers 140 and 141. Additional evidence is desirable, especially for light fission products. Therefore, we undertook measurement of the fractional independent yields of Rb^{92} and Rb^{93} from thermal-neutron fission of U^{235}. These yields with the measured[1-3] fractional yields of Kr^{92}, Kr^{93}, Y^{92}, and Y^{93} give detailed information about the charge dispersion for mass numbers 92 and 93. The decay chains and the yields determined are summarized in Fig. 1 of the preceding article.[3]

II. METHODS

A. Experimental Procedure

Solutions containing U^{235} were irradiated with thermal neutrons in the Oak Ridge Research Reactor through the use of a fast pneumatic rabbit system[4]; then fission-product rubidium isotopes were separated from strontium isotopes by passing the solution through a filter bed of $SrCO_3$.[5] The irradiations lasted 1.2 to 1.3 sec, and separation of Rb^+ from Sr^{2+} occurred in times as short as 6.1 sec after the end of an irradiation.

The irradiated solution, in a 1-ml volume, contained 10 mg of Rb^+ carrier, $(Sr^{85})^{2+}$ tracer, 10 μg of U^{235} as UO_2^{2+}, 0.6M NH_3, and 0.3M $(NH_4)_2CO_3$. The times required for passage of the solution through a filter bed ranged from 1.1 to 2.4 sec; the effective time of a rubidium–strontium separation was taken to be the mean filtration time.

Approximately 70% of the Rb^+ and 1% of the Sr^{2+} passed through a strontium carbonate filter bed (30 mg of Sr) supported on an M-porosity fritted disk in a 15-ml filter funnel. The presence of Sr^{85} tracer in the solution allowed accurate determination of the fraction of strontium passing through the filter bed. The fraction of rubidium passing through the filter was determined by measuring the amount of rubidium carrier.

After addition of yttrium carrier, strontium carrier, and Rb^{86} tracer to the filtered solution and yttrium carrier and Rb^{86} tracer to an hydrochloric acid solution of the $SrCO_3$, the yttrium in each fraction was separated from strontium and rubidium three to four hours after the initial rubidium–strontium separation, and rubidium and strontium were separated a few days later.

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