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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[4] J. D. Rylander, Oak Ridge National Laboratory Report CF 61-3-104, 1961 (unpublished).

[5] R. S. Rai, D. R. Nethaway, and A. C. Wahl, Radiochimica Acta (to be published).

The separations involved precitipations of $Y(OH)_3$ and $SrCO_3$ from ammoniacal solutions.

Yttrium and strontium were purified,[3,6] weighed as Y_2O_3 and $SrCO_3$, respectively, and their radioactivities determined—Sr^{85} with a sodium-iodide scintillation spectrometer—Y^{92} and Y^{93} by following their decay with a beta-proportional counter and resolving the resulting decay data as described in the preceding article.[3] Rubidium in the solution remaining from the $Y(OH)_3$ and $SrCO_3$ precipitations was determined by flame photometry, and the radioactivity of Rb^{86} in the solution was determined with a sodium-iodide scintillation spectrometer.

B. Treatment of Data

It was necessary to consider four-membered radioactive decay chains and partial separation of various chain members at three different times. Equations were derived from standard equations of radioactive transformation[7] to describe the numbers of atoms of each species present as a function of time. Then approximations were made that were appropriate for the $A=92$ and 93 chains (see Fig. 1 or the preceding paper[3]) and for our experimental conditions. The simplified equations were then combined and arranged in the form

$$\phi=\left[y+x\frac{\lambda_A}{\lambda_A-\lambda_B}\right]\exp(-\lambda_B\tau), \qquad (1)$$

which is suitable for calculating both the decay constant λ_B and the fractional independent yield y of the rubidium nuclide. The quantities λ_A and x represent, respectively, the decay constant and fractional cumulative yield of the Kr nuclide, and τ represents the interval between the mean irradiation time and the mean time of the rubidium–strontium separation. The quantity ϕ is a complex function defined in the Appendix; ϕ represents approximately the fraction of atoms of a given mass number that was present as rubidium or krypton at the time of the rubidium–strontium separation.

Since the function ϕ contains both measured quantities and quantities to be determined, it was evaluated by successive approximations. First, measured and known or estimable quantities were substituted in Eq. (2) (see Appendix) along with preliminary estimates of λ_B and y, and a value of ϕ for each experiment was calculated.[8] A least-squares method[9] was used to analyze these ϕ values and their associated, measured τ values in the relationship expressed in Eq. (1) to obtain new values of λ_B and y, since x and λ_A were known. Data were weighted equally. The new values of λ_B and y were then inserted into Eq. (2), and the cycle was repeated until input and output values of λ_B and of y were consistent. The values of ϕ thus derived are plotted in Fig. 1; the lines represent Eq. (1) with values of λ_B and y derived from the least-squares analysis.[10]

The final ϕ values and their associated τ values were also analyzed using a logarithmic form of Eq. (1) and Hamilton's least-squares method.[11,12] In this method uncertainties in two parameters, ϕ and τ, are treated simultaneously, and the quantity minimized is the sum of the squares of the perpendicular distances from points to a line. The results obtained are essentially the same as those obtained with Eq. (1) and the conventional least-squares method.[9] In the conventional method the uncertainty in only one parameter ϕ is treated, and the quantity minimized is the sum of the squares of vertical distances from points to a line (in a plot such as Fig. 1).

The uncertainties in λ_B and y were calculated from the uncertainties in the slope and intercept obtained from the least-squares analysis and from the uncertainties in parameters other than λ_B and y appearing in Eqs. (1) and (2). The uncertainties in the slope and intercept depend on the uncertainties in ϕ and τ used in the analysis; these were estimated to be $\pm 0.07\phi$ and ± 0.7 sec, respectively, values which are reasonable both experimentally and statistically.[13] The effect of uncertainties in other parameters on the uncertainties in λ_B and y are small (see Appendix).

III. RESULTS AND DISCUSSION

The results of the measurements and the calculations are that the half-lives of Rb^{92} and Rb^{93} are 4.1 ± 0.3 and 5.1 ± 0.3 sec, respectively, and the fractional independent yields of Rb^{92} and Rb^{93} are 0.62 ± 0.16 and 0.59 ± 0.08, respectively, based on the cumulative yields of Y^{92} and Y^{93}. The half-life values are appreciably lower than those reported in the literature,[14] 5.3 ± 0.5 sec for Rb^{92} and 5.6 ± 0.5 sec for Rb^{93}, but we believe, after considering the two methods of measurement, that the new values are the more reliable.

As shown in Fig. 3 of the previous paper, the in-

[6] A. E. Norris, Ph.D. thesis, Washington University, 1963, University Microfilms (Ann Arbor, Michigan), L. C. Card No. Mic. 64-2325.

[7] See, for example, W. Rubinson, J. Chem. Phys. 17, 542 (1949).

[8] R. A. Rouse wrote the computer program for evaluation of ϕ.

[9] W. R. Busing and H. A. Levy, Oak Ridge National Laboratory Report No. ORNL-TM-271, 1962 (unpublished). The program was modified for use on an IBM-7072 computer by R. A. Rouse.

[10] Data for $\tau=50.1$ sec were not included in the analysis and are not shown in Fig. 1. Values of ϕ obtained from the data and Eq. (2) are $(7\pm 10)\times 10^{-4}$ and $(18\pm 10)\times 10^{-4}$ for $A=92$ and 93, respectively; values predicted from Eq. (1) are $(3\pm 1)\times 10^{-4}$ and $(8\pm 4)\times 10^{-4}$. The agreement and the closeness of the values to zero show that Sr^{92} and Sr^{93} are not formed appreciably by a third, slower path (e.g., by decay of an hypothetical rubidium isomer).

[11] W. C. Hamilton, Acta Cryst. 14, 185 (1961).

[12] G. J. Atta and A. R. Jenkins wrote the program for Hamilton's least-squares method for use on a CDC 1604A computer.

[13] Statistical reasonableness was judged by the χ^2 criterion; it was required that the goodness of fit parameter λ in Hamilton's least-squares method not differ significantly from 8.3, which is the value of χ^2 for probability 0.5 and 9 degrees of freedom (11 experiments, 2 parameters to be determined).

[14] K. Fritze and T. J. Kennett, Can. J. Phys. 38, 1614 (1960).

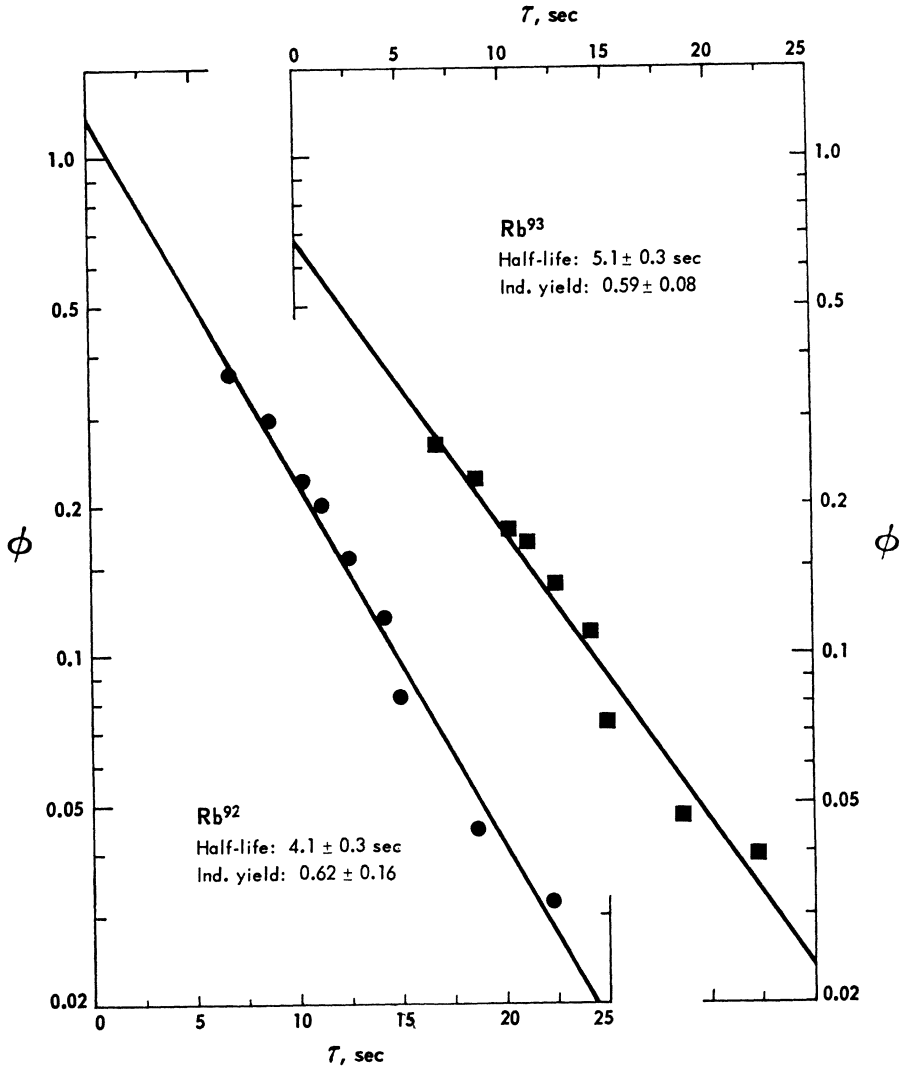

FIG. 1. Least-squares analysis of data with Eq. (1). The highest point for each curve represents the average of three measurements.

dependent-yield values are consistent with a Gaussian representation of nuclear charge dispersion for $A=92$ and $A=93$; therefore, the hypothesis that Gaussian representations of charge dispersion are generally applicable is supported.

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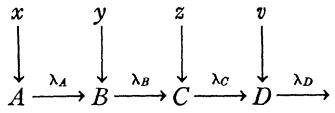
APPENDIX

Consider the following fission-product decay chain and definitions, which are consistent with previous usage[1,3] when $v=0$. (In this paper A, B, C, and D refer, respectively, to Kr, Rb, Sr, and Y isobars.)



x=fractional cumulative yield of A, based on the cumulative yield of D.

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

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

t=time interval between the end of an irradiation and the separation of B and C.

t_1=time interval between the end of an irradiation and the opening of the rabbit cartridge, when some A may have been lost.

t_2 = time interval between the end of an irradiation and the separation of C and D.

$\Delta = t - t_1$.

$\tau = 0.5T + t$.

μ = fraction of A lost at the end of time interval t_1.

$\alpha, \beta, \gamma, \delta$ = fractions of A, B, C, and D, respectively, in the filtered solution resulting from separation of B from C, based on the sum of the amounts in the precipitate and solution.

N = total number of atoms formed with a given mass number.

A_{t_1}, $B_{t_2}^{\mathrm{I}}$, C_t^{II}, etc. = number of atoms of the designated element (A,B,C) present at the end of the designated time interval (subscripts t_1, t_2, t) and in the designated fraction (superscripts I, the precipitate, and II, the filtered solution). The absence of a superscript denotes the sum of the numbers of atoms in the two fractions, e.g., $D_t = D_t^{\mathrm{I}} + D_t^{\mathrm{II}}$.

$$J = [1 - \exp(-\lambda_A T)]\exp(-\lambda_A t).$$

$$K = [1 - \exp(-\lambda_B T)]\exp(-\lambda_B t).$$

$$L = [1 - \exp(-\lambda_C T)]\exp(-\lambda_C t).$$

$$J_1 = [1 - \exp(-\lambda_A T)]\exp(-\lambda_A t_1).$$

$$a = \frac{\lambda_B \lambda_C}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)}, \quad b = \frac{\lambda_A \lambda_C}{(\lambda_A - \lambda_B)(\lambda_C - \lambda_B)},$$

$$c = \frac{\lambda_A \lambda_B}{(\lambda_A - \lambda_C)(\lambda_B - \lambda_C)}.$$

$$\bar{b} = \frac{\lambda_C}{\lambda_C - \lambda_B}, \quad \bar{c} = \frac{\lambda_B}{\lambda_B - \lambda_C}.$$

$$\phi = \frac{1}{\beta}\left\{\frac{D_{t_2}^{\mathrm{II}}}{D_{t_2}}\left(1 - \frac{\mu x J_1}{\lambda_A T}\right) - \alpha\frac{A_t}{N} - \gamma\frac{C_t}{N} - \delta\frac{D_t}{N}\right\}$$
$$+ \frac{Jx}{T(\lambda_A - \lambda_B)}\{1 - \mu + \mu \exp[(\lambda_A - \lambda_B)\Delta]\}. \quad (2)$$

$$\frac{A_t}{N} = \frac{(1-\mu)xJ}{\lambda_A T}. \quad (2a)$$

$$\frac{C_t}{N} = \frac{1}{\lambda_C T}\{L(z + y\bar{c} + xc)$$
$$+ K[y\bar{b} + xb[1 - \mu + \mu \exp((\lambda_B - \lambda_C)\Delta)]\}$$
$$+ Jxa[1 - \mu + \mu \exp((\lambda_A - \lambda_C)\Delta)]\}. \quad (2b)$$

$$\frac{D_t}{N} = v \exp(-\lambda_D \tau) + (1 - v)\left(\frac{\lambda_C}{\lambda_C - \lambda_D}\right)$$
$$\times [\exp(-\lambda_D \tau) - \exp(-\lambda_C \tau)]. \quad (2c)$$

The evaluation of Eq. (1) was programmed[8] for the Washington University IBM-7072 computer; the programming was checked by hand calculation. The quantities $D_{t_2}^{\mathrm{II}}$, D_{t_2}, β, and γ were determined radiochemically, and all time intervals were measured in each experiment. The quantities x and v and all decay constants except λ_B were known (see Fig. 1 of preceding article). Some quantities were estimated: $\mu = 0.5 \pm 0.5$, because of lack of information about Kr escape; $\alpha = \beta \pm (1-\beta)$, because $1-\beta$ is determined mainly by hold-up of solution in the filter disk; and $\delta = \gamma \pm \gamma$, because tracer experiments had shown that very little strontium or yttrium tracer passes through a $SrCO_3$ filter bed, and the little that does may be due to channeling. The values to be determined, y and λ_B, were estimated initially and then revised after each calculation. By definition, $z = 1 - x - y - v$.

The first term in Eq. (2) is the important one and involves only the measured quantities β, $D_{t_2}^{\mathrm{II}}$, and D_{t_2}; the contribution of the other terms to ϕ is less than 25%.

The effect of the uncertainty in a quantity on the final results was investigated by adding the uncertainty to each quantity, one at a time, calculating a new set of ϕ values, and performing the least-squares calculation. The effects were all small; the largest was that due to the ± 0.5 uncertainty in μ on the Rb^{92} half-life and fractional independent yield, 0.06 sec and 0.06, respectively. Addition of the squares of these uncertainties, as well as the squares of all other uncertainties determined, to the variance from the least-squares analysis increased the uncertainties in the Rb^{92} half-life and yield from 0.22 to 0.23 and 0.14 to 0.16, respectively. There was no change in uncertainties for the Rb^{93} parameters.