Charge Distribution in Fission: Fractional Cumulative Fission Yields of ^{143}Ba and ^{144}Ba from Thermal-Neutron-Induced Fission of ^{235}U†

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Rapid chemical separations of Cs and Ba from La and Ce have been used to measure half-lives and cumulative yields of ^{143}Ba and ^{144}Ba from thermal-neutron-induced fission of ^{235}U. The half-lives are 13.2±0.3 sec for ^{143}Ba and 11.9±0.3 sec for ^{144}Ba. The yields, each expressed as a fraction of the total chain, are 0.88±0.06 and 0.78±0.06 for ^{143}Ba and ^{144}Ba, respectively. Charge distribution for $A=144$ can be represented by a Gaussian distribution with width parameter $\sigma=0.52\pm0.02$ and $Z_p=56.09\pm0.05$. For $A=143$, $\sigma=0.58\pm0.05$, and $Z_p=55.87\pm0.05$, in agreement with previously reported values. Evidence is presented that charge distribution as determined from yields of several isotopes of the even-Z elements Xe, Ba, Kr, and Sr can be satisfactorily represented by a single Gaussian distribution with $\sigma=0.56$. A possible even-odd effect in nuclear charge distribution from fission of ^{235}U is noted.

INTRODUCTION

RELATIVE yields of isobaric fission products have been shown to fit a Gaussian curve by Wahl and his co-workers,[1,2] who found the width parameter σ of the Gaussian to be approximately constant for several mass chains. Niece[3] suggested, however, that there is a small, but statistically significant, decrease in σ with increasing mass in both the light- and heavy-mass peaks. Strom *et al.*[4] suggest that σ also decreases with increasing mass in the mass range $A=131$–133.

To determine whether this trend continues in the heavy peak, we measured the fractional cumulative fission yields of ^{144}Ba, the second yield measured for the mass-144 chain, and of ^{143}Ba, the third yield for the mass-143 chain.

The fractional cumulative yields of ^{143}Ba and ^{144}Ba were measured in a series of nine experiments, each consisting of a brief irradiation of a U solution followed by a rapid precipitation and filtration of $La(OH)_3$ from the irradiated solution. After the shorter-lived members of the mass-143 and mass-144 chains had decayed to ^{143}Ce and ^{144}Ce, $La(OH)_3$ was dissolved; equal amounts of Ce were added to this solution and to the filtrate, and Ce was purified from both solutions. By comparison of the specific activities of ^{143}Ce in the two solutions, the fraction of the mass-143 chain existing as ^{143}Ba and its precursors at the time of the initial Ba-La separation was calculated. From the results of this series of separations at varying time intervals after irradiation, the fraction of the total chain existing as ^{143}Ba and its precursors at the time of irradiation was calculated. The fraction of the mass-144 chain existing as ^{144}Ba and its precursors was similarly calculated from the ^{144}Ce activities of the same samples.

EXPERIMENTAL

Irradiations

Irradiations were made at the Oak Ridge Research Reactor in the pneumatic tube facility where the thermal-neutron flux density was $\sim6\times10^{13}$ neutrons cm^{-2} sec^{-1}. A cadmium ratio for ^{235}U fission of ~60 had been measured previously.[5] 1-ml solutions containing 1 mg of U (93% ^{235}U) and 2 mg of natural La were irradiated in high-density polyethylene capsules for periods of 2 sec.

Chemical Procedures

After irradiation the polyethylene capsule was punctured with a hypodermic needle and the contents drawn into a fritted-glass filtering funnel on which was suspended 10 mg of Ba carrier in concentrated ammonia solution. The reactant mixture was vacuum-filtered and the precipitated $La(OH)_3$ washed. The time interval τ between the midpoint of filtration and the mean irradiation time was taken to be the time between occurrence of fission and the separation of La from its precursors. Each filtration required ~3–4 sec. Tracer experiments showed that ~1.1% of the Ba was coprecipitated with $La(OH)_3$ and that ~0.4% of the La was in the filtrate.

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[1] A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1963).
[2] A. E. Norris and A. C. Wahl, Phys. Rev. **146**, 926 (1966).
[3] L. H. Niece, Oak Ridge National Laboratory Report No. ORNL-TM-1333, 1965 (unpublished).
[4] P. O. Strom, D. L. Love, A. E. Greendale, A. A. Delucchi, D. Sam, and N. E. Ballou, Phys. Rev. **144**, 984 (1966).
[5] D. E. Troutner, R. L. Ferguson, and G. D. O'Kelley, Phys. Rev. **130**, 1466 (1963).

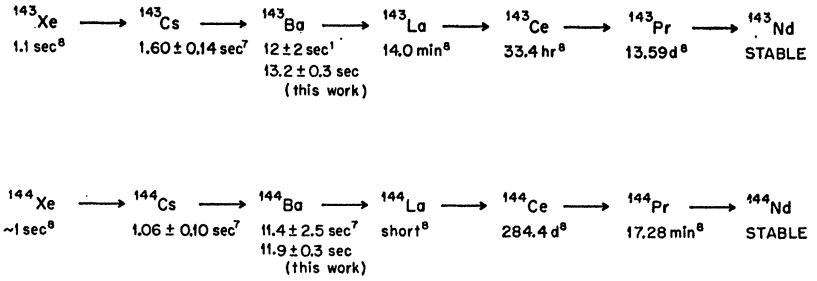

FIG. 1. β decay chains for mass numbers 143 and 144. Superscripts refer to references.

Equal aliquots of a Ce carrier solution were added to the filtrate and to the filter funnel. The mixture on the funnel was acidified with HNO_3 and the resulting solution collected. After a delay of ~3 h to allow decay of ^{143}La to ^{143}Ce, the Ce in each of the two solutions was purified by a ceric-iodate precipitation followed by a methylisobutyl-ketone extraction as described by Glendenin *et al.*[6]

Counting

Each sample was precipitated as $Ce_2(C_2O_4)_3$, filtered, dried, and weighed on a tared filter paper circle $\frac{7}{8}$ in. in diameter. The circles were mounted on Al counting plates.

Decay of the 33-h ^{143}Ce was followed in some samples with a thin-window Geiger-Mueller tube, which required graphical resolution of the resulting decay curves into components representing 33.4-h ^{143}Ce, 13.7-day ^{143}Pr, 32.5-day ^{141}Ce, and 285-day ^{144}Ce. In other samples, decay curves were obtained with a single-channel γ spectrometer set to discriminate against Ce fission-product activities other than ^{143}Ce. These curves consisted of the 33.4-h component and about 1% of a longer-lived component, identified by comparison with samples of ^{143}Ce and ^{144}Ce as primarily daughter ^{143}Pr and ^{144}Ce. This long-lived component was subtracted from the total decay curve. The decay of each sample was followed for at least seven half-lives. At least three separate measurements, each with a statistical standard deviation of 1%, were made during each half-life.

After decay of the ^{143}Ce, the 2.98-MeV β activity of ^{144}Pr in equilibrium with the ^{144}Ce was counted through a 214-mg/cm^2 Al absorber. The detector was a continuous-flow Geiger-Mueller counter. Calculation of the activity ratio for each pair of samples was made by averaging four or five separate measurements, each resulting from activity measurements of both samples during a single day. In each individual measurement, the statistical standard deviation was 1% for both the sample count and the background count.

Because the long half-life of ^{144}Ce made following the decay of the samples impractical, errors resulting from incomplete radiochemical purifications were estimated by purifying, dissolving, and reprecipitating triplicate Ce fission-product samples. The specific activities of the three samples agreed to within 0.35% after our normal purification procedure and were unchanged by the additional purification step.

[6] L. E. Glendenin, K. Flynn, R. F. Buchanan, and E. P. Steinberg, Anal. Chem. **27**, 59 (1955).

CALCULATIONS

The ratio of ^{143}Ce radioactivity recovered from the filtrate to the sum of the ^{143}Ce radioactivities recovered from the filtrate and from $La(OH)_3$, corrected for chemical yield of $Ce(IO_3)_4$, for incomplete separation of Ba from La, and to the same time after irradiation is represented here by Q. Since La and its decay products were in the precipitate and Ba and its precursors were in the filtrate, Q is the fraction of the chain present as ^{143}Ba or its precursors at the time of separation. Q can be defined in a similar manner for the mass-144 chain.

From the measured values of Q the fractional cumulative yields of ^{143}Ba and ^{144}Ba were calculated by the following method. The fission-product decay chains for $A=143$ and $A=144$, which are shown in Fig. 1,[1,2,7,8] can be represented by the following expression:

$$\begin{array}{ccc} x & y & z \\ \downarrow & \downarrow & \downarrow \\ A & \rightarrow B & \rightarrow C, \end{array}$$

where A, B, and C represent Cs, Ba, and La, respectively. The yields, all expressed as fractional chain yields, are defined as follows: x is the cumulative yield of A, y is the independent yield of B, z is the sum of the independent yields of C and its decay products, and $(x+y)=(1-z)$ is the cumulative yield of B.

For $\lambda_A \gg \lambda_B$, and for τ much longer than the half-life of A, the following equation derived from the equations of radioactive decay is applicable:

$$Q=\{1-z+x[\lambda_B/(\lambda_A-\lambda_B)]\}e^{-\lambda_B\tau}.$$

The half-lives of ^{143}Cs and ^{144}Cs were taken to be 1.60±0.14 and 1.06±0.10 sec, respectively, as reported by Amarel *et al.*[7] Since the uncertainties for all points are approximately equal, an unweighted least-squares

[7] I. Amarel, R. Bernas, R. Foucher, J. Jastrzebski, A. Johnson, J. Teillac, and H. Gauvin, Phys. Letters **24B**, 402 (1967).

[8] G. Hermann, Habilitationsschrift, Universität Mainz, 1964 (unpublished).

TABLE I. Fraction of chain present as Ba and precursors at separation time.

Separation time (sec)	Q ^{143}Ba	^{144}Ba
12.0	0.492	0.371
13.3	0.432	0.352
18.9	0.349	0.276
20.3	0.335	0.255
26.8	0.212	0.167
31.0	0.179	0.127
39.5	0.121	0.069
49.7	0.073	0.054
55.1	0.047	0.029

analysis was used to fit a straight line to each set of values. The half-lives of ^{143}Ba and ^{144}Ba given in Fig. 1 were calculated from the slopes of these lines and are in agreement with published results.

RESULTS

Separation times and Q values for ^{143}Ba and ^{144}Ba are given in Table I. Plots of $\log_{10}Q$ values as a function of separation time are shown in Fig. 2. The error limits shown represent an estimated 5% error in the radiochemical measurements upon which each point is based.

Values of the term $1-z+x[\lambda_B/(\lambda_A-\lambda_B)]$ taken from the intercepts of the lines with the ordinate (Fig. 2) are 0.917 ± 0.035 and 0.797 ± 0.035 for ^{143}Ba and ^{144}Ba, respectively. Since the independent yields of ^{143}Cs and ^{144}Cs have not been measured, the value of x for each chain was evaluated by successive approximations. Assuming that the intercepts are approximately equal to the fractional cumulative yields of ^{143}Ba and ^{144}Ba, that the charge distribution for these chains is Gaussian, and using previously reported[1] cumulative yields of ^{143}Xe and ^{144}Xe, x was estimated to be 0.32 and 0.13 for $A=143$ and $A=144$, respectively. The values of $1-z$ calculated with these values of x are improved estimates of the fractional cumulative yields of ^{143}Ba and ^{144}Ba and were used for a second estimate of values of x. These values were 0.27 and 0.125 for $A=143$ and $A=144$, respectively. A third such approximation did not change the values of x. Using these final values of x and the values of the term $1-z+x[\lambda_B/(\lambda_A-\lambda_B)]$ given above, the fractional cumulative yields of ^{143}Ba and ^{144}Ba were calculated to be 0.880 ± 0.035 and 0.785 ± 0.035, respectively.

The error limits above are standard deviations of the intercepts of the lines in Fig. 2 and include random errors associated with the timing, sample weighing, measurement of radioactivity, and resolution of decay curves. The magnitudes of the standard deviations are consistent with the estimated 5% uncertainty in each point. The standard deviations of the intercepts do not include systematic errors due to uncertainties in times of separation and in half-lives and yields of Cs precursors.

The error introduced by the assumptions made in deriving the equation for Q is less than 0.001. Uncertainties of ±1 sec in τ, ±0.14 sec in the ^{143}Cs half-life, and ±0.05 in x cause uncertainties of ±0.05, ±0.004, and ±0.007, respectively, in the fractional cumulative yield of ^{143}Ba. The total uncertainty in the yield, taken to be the square root of the sum of the squares of all the uncertainties, including the standard deviation, is ±0.06. Since the total correction of the ^{144}Ba yield for decay of ^{144}Cs is small, uncertainties in that correction can be neglected. The uncertainty caused by an error of ±1 sec in τ is ±0.05. The total uncertainty, calculated in the same manner as for the ^{143}Ba yield, is ±0.06. Values resulting from these experiments and including the uncertainties described above are 0.88 ± 0.06 and 0.78 ± 0.06 for ^{143}Ba and ^{144}Ba, respectively.

For a single isobaric chain, measured fractional cumulative yields may be plotted as a function of atomic number on probability graph paper, as described by Terrell.[9] A Gaussian distribution appears as a straight line from which Z_p, the most probable charge for the chain, and the width parameter σ may be obtained graphically.

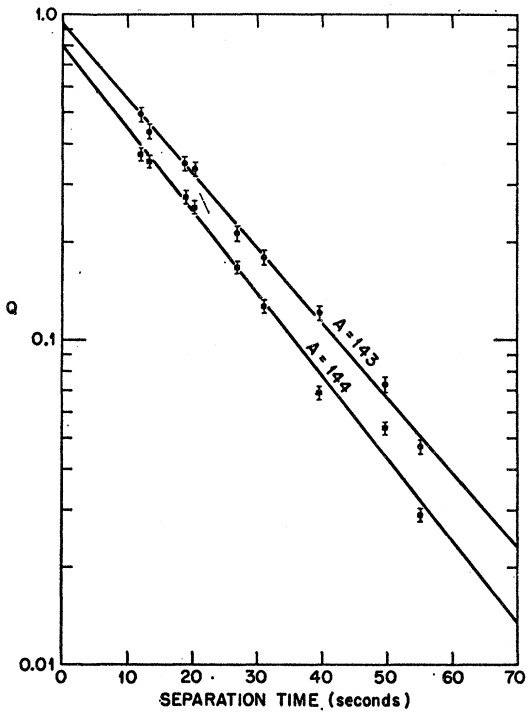

FIG. 2. $\text{Log}_{10}Q$ as a function of τ.

[9] J. Terrell, Phys. Rev. 108, 783 (1957).

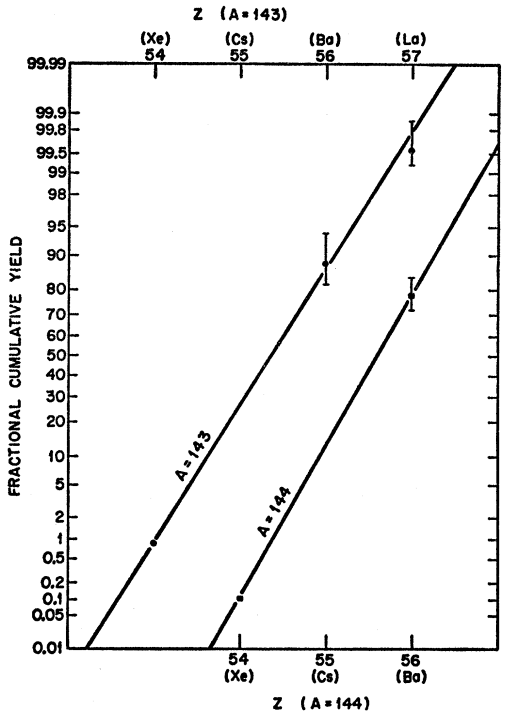

FIG. 3. Charge dispersion curves for mass numbers 143 and 144. The probability scale is given in percent.

Such plots for the mass-143 and mass-144 chains are shown in Fig. 3. Other yields shown are taken from Ref. 1. Values of σ and Z_p estimated from these plots are shown in Table II.

DISCUSSION

Two or more cumulative yields have been measured for each of six consecutive chains in the heavy mass peak from thermal-neutron fission of ^{235}U. Figure 4 is a plot of σ values, obtained as indicated above, for those chains as a function of A. The slope of a straight line fit to these points by a weighted least-squares procedure is -0.035 ± 0.008. Considering the calculated uncertainty, this value is different from zero at the 99.999% confidence level. These results appear to support the suggestion of Niece[3] that σ decreases with mass in this region. Norris and Wahl[2] conclude, however, on the basis of the same data, that a single σ, of width 0.59 ± 0.06, gives a satisfactory representation of nuclear-charge dispersion.

TABLE II. Values of σ and Z_p for $A=143$ and $A=144$.

A	σ	Z_p
143	0.58 ± 0.05	55.87 ± 0.05
144	0.52 ± 0.02	56.09 ± 0.05

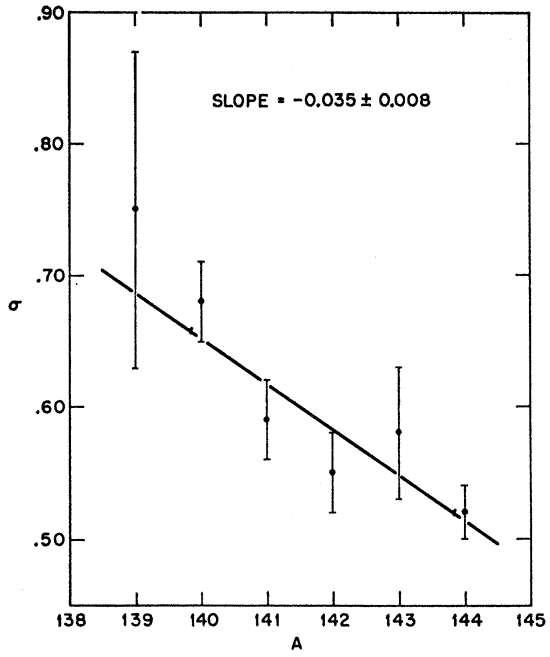

FIG. 4. Width parameter σ as a function of A.

In an attempt to find the cause of this apparent variation in σ, we examined the manner in which σ was calculated for each of these mass chains. If a Gaussian distribution adequately represents the variation of yield with nuclear charge for a given mass, then σ can be deduced from any two known cumulative yields. For example, see Fig. 5, which is a plot on a probability scale of fractional cumulative yields as a

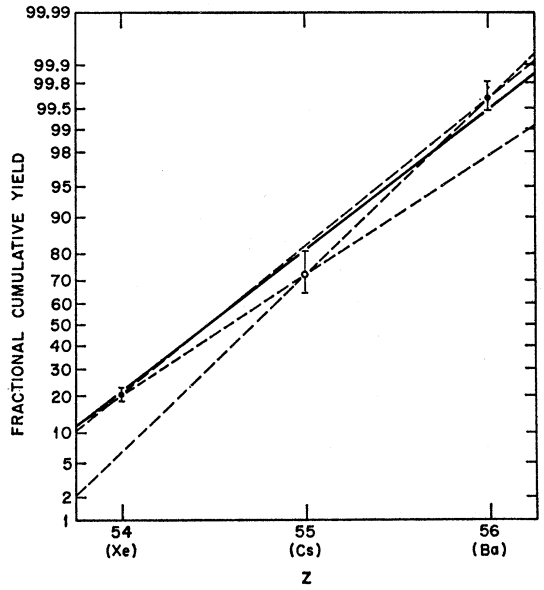

FIG. 5. Charge dispersion curve for mass number 141. The probability scale is given in percent.

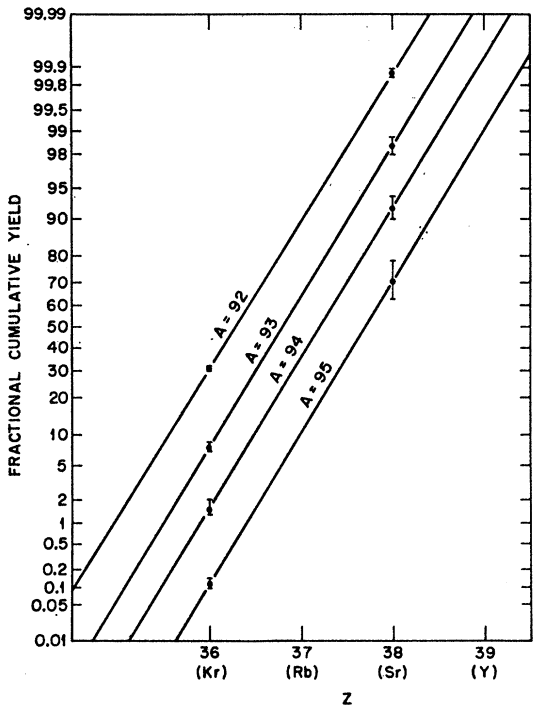

FIG. 6. Charge dispersion curves for mass numbers 92, 93, 94, and 95. The probability scale is given in percent.

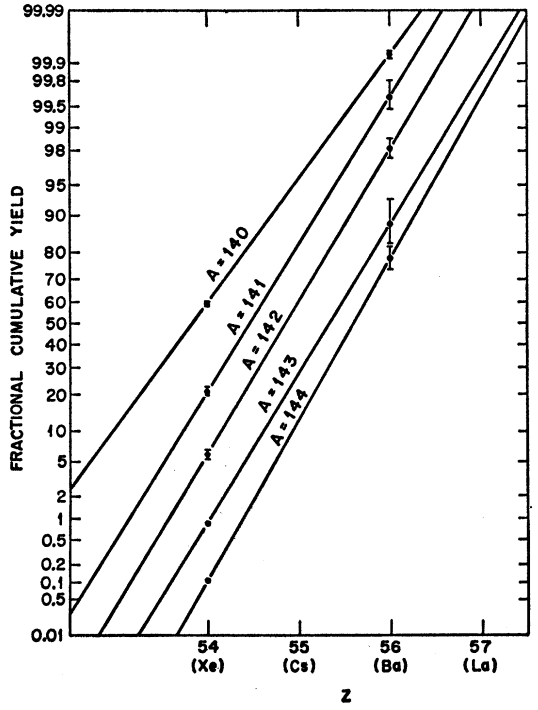

FIG. 7. Charge dispersion curves for mass numbers 140, 141, 142, 143, and 144. The probability scale is given in percent.

TABLE III. Values of σ for $A=92$–95 and $A=140$–144.

A	σ
92	0.57
93	0.55
94	0.56
95	0.56
140	0.68
141	0.57
142	0.55
143	0.57
144	0.52

function of Z for $A=141$. The solid line was fitted by Wahl *et al.*[1] to all three of the known yields for the chain. From this line, values of 0.59 for σ and 54.97 for Z_p are calculated. However, if only two instead of three yields were known for this chain, the apparent value of σ would be changed. If only the yields of Xe and Cs, Xe and Ba, or Cs and Ba were known, the σ calculated would have been 0.67, 0.57, or 0.47, respectively. Lines connecting each of these three possible pairs of points are shown as dashed lines in Fig. 5.

If the calculated value of σ depends systematically on which two of the yields in a chain have been measured, the apparent variation of σ with mass might be

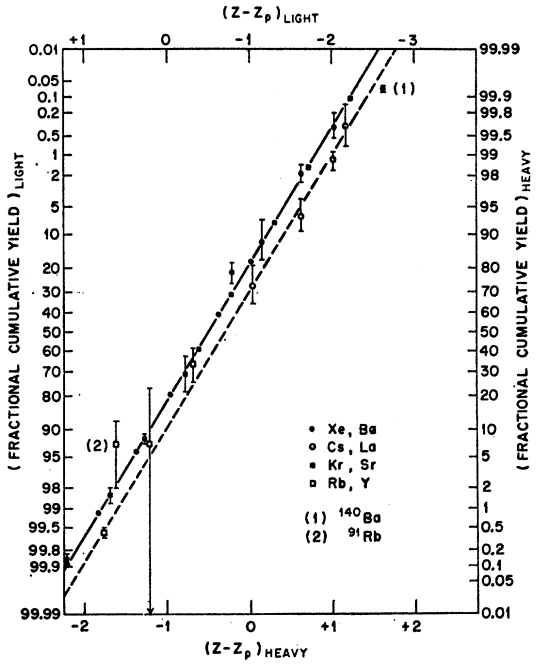

FIG. 8. Probability-scale plot for fractional cumulative yields for mass numbers 91–95 and 139–144. The probability scale is given in percent. Plot is discussed in text.

investigated by comparing σ values for several chains in which yields of the same two elements were used for calculation. We searched, therefore, for a pair of elements with yields known for as many chains as possible. The yields of both Xe and Ba, or their proton complements Kr and Sr, have been measured for a total of nine chains in this mass region and the nearly complementary mass region $A=92$–95. For each of these chains the sum of the fractional independent yields of Xe, Cs, and Ba (or Kr, Rb, and Sr) is greater than 0.70. Since there are no other pairs of elements for which so many data are available, and because the yields of Xe and Kr are reported with low limits of error, we chose to compare σ values based on Xe-Ba and Kr-Sr yields. Figures 6 and 7 are plots on probability scales of fractional cumulative yields as a function of Z for these nine chains. All yields except those for ^{143}Ba and ^{144}Ba are taken from Refs. 1 and 2.

The values of σ for these chains are summarized in Table III. Only yields of Xe and Ba and Kr and Sr have been considered in estimating these σ's. The unweighted average value of σ for the nine chains is 0.57. Only σ for $A=140$, 0.68, is far from this average. If it is neglected, the average value for the other eight chains is 0.56.

The general agreement for the charge dispersion in chains $A=91$–95 and $A=139$–144 is shown in Fig. 8. All yields from these chains are plotted on a probability scale as a function of $Z-Z_p$. The solid line was drawn through $Z-Z_p=0$ with the slope equivalent to $\sigma=0.56$. Yields of Xe and Kr were plotted on the line, and Z_p calculated from each of these yields and the solid line. Measured yields of Ba, Sr, Cs, Rb, La, and Y were then plotted at the corresponding values of $Z-Z_p$. For example, the yields of ^{141}Xe, ^{141}Cs, and ^{141}Ba are plotted at $Z-Z_p$ values of -0.97, $+0.03$, and $+1.03$, respectively.

The agreement of the Xe-Ba and Kr-Sr yields with the solid line (only the yield of ^{140}Ba falls far from the line) indicates that charge dispersion for these pairs of elements can be satisfactorily represented by a single Gaussian curve. The width parameter σ, however, appears to be $\sim$0.56, slightly smaller than the earlier values of 0.59 and 0.62 reported by Norris and Wahl[2] and by Wahl *et al.*,[1] respectively.

Yields of Rb, Y, Cs, and La fall along a line which is parallel to the solid line but displaced by $\sim$0.2 charge unit. Only the yield of ^{91}Rb is displaced in the opposite direction. Note that the points on the solid line are for even-Z nuclides and the displaced points are for odd-Z nuclides. Although the vertical displacement from the solid line is less than the uncertainties in the points for four of the eight odd-Z nuclides, the distribution around the line is clearly not a random one. Further, for the six chains discussed here ($A=92$, 93, 95, 140, 141, and 143), for which three fractional cumulative yields have been measured, odd-Z points for each chain are displaced from a line connecting even-Z points for the chain.

The previously noted[3] change in σ with mass for the region $A=139$–144 seems then to be the result of the way in which σ was calculated. The highest σ, for $A=139$, is the result of a calculation based on one even-Z and one odd-Z nuclide, and the lowest σ, for $A=144$, on two even-Z nuclides.

We believe, therefore, that nuclear-charge dispersion in the regions $A=91$–95 and $A=139$–144 can be satisfactorily represented by a single Gaussian curve, but that there are slight deviations from this curve of an odd-even nature. The possibility of the existence of this effect should be considered in estimating σ for any chain for which only two fractional cumulative yields are known.

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