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Fission of ^{234}U and ^{236}U with 14.8-MeV Neutrons*

D. R. Nethaway and B. Mendoza
University of California, Lawrence Livermore Laboratory, Livermore, California 94550
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We determined mass-yield distributions from fission of ^{234}U and ^{236}U with 14.8-MeV neutrons. We measured the yields of 24 products covering the mass range 66–161 for ^{234}U and for 30 products covering the mass range 66–172 for ^{236}U. We estimated total chain yields by correcting for the effects of nuclear-charge dispersion in fission. The mass-yield curves are similar to those for fission of ^{233}U, ^{235}U, and ^{238}U with 14.8-MeV neutrons. We measured the independent fractional chain yields of the following nuclides: for ^{234}U: ^{96}Nb, $(1.54\pm0.10)\times10^{-3}$; ^{136}Cs, 0.095 ± 0.006; for ^{236}U: ^{96}Nb, $(2.7\pm0.3)\times10^{-4}$; ^{126}Sb, 0.096 ± 0.010; ^{136}Cs, 0.022 ± 0.002.

INTRODUCTION

In this study of the mass-yield distribution for fission of ^{234}U and ^{236}U with 14.8-MeV neutrons, the ^{234}U products that were measured ranged from ^{66}Ni to ^{161}Tb and, for ^{236}U, from ^{66}Ni to ^{172}Er. No previous measurements have been reported for fission of these isotopes with 14-MeV neutrons. These measurements are part of a series that include ^{235}U and ^{238}U fission[1] and ^{233}U fission[2] with 14-MeV neutrons. This paper deals specifically with ^{234}U and ^{236}U; we reported elsewhere[3] on a detailed examination of the fission distribution for all five uranium isotopes. The ^{234}U measurements were made as relative yields and then converted to absolute fission yields by adjusting the total area under each half of the mass-yield curve to unity. The ^{236}U calculations are based both on that method and on the measurement of the number of fissions.

EXPERIMENTAL DETAILS

Our experimental details are similar to those reported previously[1,2] and are described here only briefly. The 14.8-MeV neutron irradiations were made at the insulated-core-transformer (ICT) accelerator at the Lawrence Livermore Laboratory. The uranium targets were placed at 0° to the source of neutrons produced by the reaction of a 400-keV deuteron beam on a rotating titanium tritide target. The 14-MeV D-T neutron source strength was typically 1×10^{12} sec^{-1} or greater; the flux density in the uranium target was about 2×10^{10} cm^{-2} sec^{-1}, with very little decrease, if any, during an 8-h irradiation.

The ^{234}U target assembly consisted of about 48 mg U_3O_8 (99.33% ^{234}U, with $<5\times10^{-4}$% ^{233}U, 0.36% ^{235}U, 0.11% ^{236}U, and 0.20% ^{238}U) wrapped in aluminum foil and sealed in a thin polyethylene bag. The assembly was placed in a 30-mil cadmium can to reduce the low-energy neutron background. Five such targets were prepared from 240 mg of the uranium oxide, which was obtained from the Oak Ridge National Laboratory.

The ^{236}U targets were prepared in a similar fashion from 480 mg U_3O_8 (99.21% ^{236}U, with $<2\times10^{-3}$% ^{233}U, 5×10^{-3}% ^{234}U, 0.57% ^{235}U, and 0.21% ^{238}U) obtained from Oak Ridge National Laboratory. Six targets were used, ranging in mass from about 40 to 150 mg each. In addition, two 10-mil uranium metal foils weighing 200 mg each were used as targets.[4] The isotopic composition was 99.76% ^{236}U, 0.20% ^{235}U, and 0.03% ^{238}U. These foils were wrapped in aluminum and then sandwiched between monitor foils before being sealed in polyethylene bags. The monitor foils were niobium in one case and yttrium in the other.

The uranium targets were irradiated for either 8 or 16 h, producing up to 7×10^{11} fissions in the ^{234}U and 1.5×10^{12} fissions in the ^{236}U. The target foil was dissolved together with the aluminum wrapping, so that recoil losses were negligible. The dissolving was delayed for about 17 h, so that the bulk of the short-lived products could decay

TABLE I. Radioactivity and counting-efficiency measurements.

Nuclide	Half-life (days)	Type of counter [a]	Discriminator settings (keV)	Basis for counting efficiency	E_γ (I_γ) [b]	Uncertainty in efficiency (%)
^{66}Ni	2.29	β	...	β efficiency curve	...	10
^{67}Cu	2.56	β	...	β curve and NaI PHA	184(0.43)	10
^{72}Zn	1.942 [c]	β	...	Ge PHA [d]	835(0.96, ^{72}Ga)	3
^{89}Sr	50.6	β	...	4π counter	...	5
^{88}Y	106.6	NaI	1600–1910	e	...	3
^{91}Y	58.8	β	...	β efficiency curve	...	10
^{93}Y	0.427	β	...	β efficiency curve	...	10
^{95}Zr	65.0	NaI	600–900	Ge PHA	765(0.99, ^{95}Nb)	4
^{97}Zr	0.701	NaI	600–900	Ge PHA	743(0.94)	6
^{92}Nb	10.16	Ge	...	NaI and Ge PHA	935(0.99)	4
^{96}Nb	0.975	Ge	...	Ge PHA	778(0.97)	5
^{99}Mo	2.752	NaI	600–900	NaI and Ge PHA	739(0.124)	9
^{105}Rh	1.476	NaI	200–400	NaI and Ge PHA	319(0.19)	8
^{112}Pd	0.875	NaI	520–700	NaI and Ge PHA	617(0.435, ^{112}Ag)	5
^{111}Ag	7.43	NaI	200–400	$4\pi\beta$-γ	...	4
^{115g}Cd	2.208	NaI	50–580	NaI and Ge PHA	336(0.50, ^{115m}In)	10
^{115m}Cd	43.0	β	...	$4\pi\beta$-γ	...	4
^{126}Sb	12.5	Ge	...	Ge PHA	666(1.00)	4
^{127}Sb	3.87	Ge	...	Ge PHA	685(0.368)	6
^{132}Te	3.24	NaI	400–1000	NaI and Ge PHA	668(0.98, ^{132}I)	9
^{136}Cs	13.0	NaI	400–2200	Ge PHA	818(1.00)	4
^{137}Cs	10 960.0	NaI	400–2200	Ge PHA	662(0.85)	4
^{140}Ba	12.80	NaI	1100–1700	NaI and Ge PHA	1596(0.96, ^{140}La)	4
^{141}Ce	32.4	β	...	4π and Ge PHA	145(0.49)	8
^{143}Ce	1.379	NaI	200–400	NaI and Ge PHA	293(0.47)	5
^{144}Ce	284.0	β	...	Ge PHA	134(0.11)	6
^{147}Nd	11.04	NaI	470–650	NaI and Ge PHA	531(0.132)	7
^{153}Sm	1.928	β	...	β curve and NaI PHA	103(0.28)	8
^{156}Eu	15.19	NaI	800–2200	Ge PHA	812(0.108)	5
^{159}Gd	0.773	β	...	β efficiency curve	...	10
^{161}Tb	6.96	β	...	4π counter	...	5
^{166}Dy [f]	3.40	β	...	4π counter	...	5
^{169}Er	9.5	β	...	4π counter	...	5
^{172}Er [f]	2.08	β	...	4π counter	...	5

[a] β refers to gas-flow β proportional counters, NaI refers to NaI(Tl) detectors with lower and upper discriminator settings given in the next column, and Ge refers to Ge(Li) detectors used with pulse-height analyzers.

[b] The energy of the γ ray is given in keV, followed by the absolute photon intensity (photons/disintegration) in parentheses.

[c] The value of the ^{72}Zn half-life was erroneously given as 2.45 day in Ref. 2.

[d] By direct comparison with the 835-keV photon ($I_\gamma = 1.00$) in a standard sample (Ref. 7) of ^{54}Mn.

[e] By direct counting of a standard sample (Ref. 7) of ^{88}Y.

[f] The ^{166}Dy and ^{172}Er were measured as their daughter nuclides ^{166}Ho (1.116 day) and ^{172}Tm (2.65 day).

away. Aliquots of the target solution were added to 10- to 20-mg amounts of carriers for the various products. Conventional radiochemical procedures were used for the purification of each of the products.[5,6]

The rare-earth elements were separated on ion-exchange columns, with α-hydroxyisobutyric acid as the eluant. The lowest-yield rare-earth nuclides (^{160}Tb and ^{169}Er) were further purified by a second column step. The nuclides ^{166}Dy and ^{172}Er were determined by separating and measuring the ^{166}Ho and ^{172}Tm daughters. The chemical separation of strontium-yttrium and samarium-europium was delayed for several days following the irradiations in which ^{91}Y and ^{156}Eu were to be measured. This was done to allow complete decay of the precursor nuclides ^{91}Sr (9.5 h) and ^{156}Sm (9.4 h).

Details of the radioactivity measurements are given in Table I. The counting efficiencies are based on several methods: (1) comparison with 4π and 4π β-γ coincidence counters, (2) comparison with calibrated NaI(Tl) and Ge(Li) detectors with pulse-height analyzers (PHA), (3) the use of calibrated standard solutions,[7] and (4) the use of an experimentally determined curve of β counting efficiency versus mean β energy.

TABLE II. Yields of products from fission of ^{234}U with 14.8-MeV neutrons.

Product nuclide	Ratio to ^{147}Nd	Measured fission yield [a]	Estimated total chain yield [b]
^{66}Ni	3.49×10^{-4}	$(4.6\pm0.5)\times10^{-6}$	
^{67}Cu	8.3×10^{-4}	$(1.09\pm0.11)\times10^{-5}$	
^{72}Zn	8.1×10^{-3}	$(1.06\pm0.06)\times10^{-4}$	1.07×10^{-4}
^{93}Y	3.92	0.052 ± 0.005	
^{95}Zr	3.98	0.052 ± 0.002	
^{97}Zr	3.84	0.0506 ± 0.003	0.0514
^{96}Nb	6.05×10^{-3}	$(8.0\pm0.3)\times10^{-5}$ [c]	
^{99}Mo	3.34	0.044 ± 0.004	
^{105}Rh	1.68	0.0221 ± 0.0018	
^{111}Ag	1.03	0.0136 ± 0.0005	
^{112}Pd	1.15	0.0152 ± 0.0010	0.0153
$^{115m+g}$Cd	1.02	0.0134 ± 0.0013 [d]	
^{132}Te	3.28	0.043 ± 0.004	0.050
^{136}Cs	0.373	$(4.91\pm0.20)\times10^{-3}$ [c]	
^{137}Cs	3.89	0.0512 ± 0.0023	0.0515
^{140}Ba	3.00	0.0395 ± 0.0016	0.0401
^{141}Ce	2.91	0.038 ± 0.003	
^{143}Ce	2.58	0.0339 ± 0.0017	
^{144}Ce	2.10	0.0277 ± 0.0020	
^{147}Nd	1.00	0.0132 ± 0.0009	
^{153}Sm	0.143	$(1.88\pm0.15)\times10^{-3}$	
^{156}Eu	0.0369	$(4.86\pm0.22)\times10^{-4}$	
^{159}Gd	9.10×10^{-3}	$(1.20\pm0.12)\times10^{-4}$	
^{161}Tb	3.60×10^{-3}	$(4.7\pm0.5)\times10^{-5}$	

[a] The experimental standard deviations given here do not include a systematic 5–10% uncertainty in the normalization of the mass-yield curve.

[b] Values of the estimated total chain yield are given where they differ appreciably from the measured fission yield.

[c] The ^{96}Nb and ^{136}Cs values are independent fission yields.

[d] The measured ^{115m}Cd/^{115g}Cd ratio is 0.067.

FISSION YIELD MEASUREMENTS

^{234}U Results

Results of the ^{234}U fission yield measurements are summarized in Table II. Each result is the average of about three separate measurements. The results were first calculated as the ratio to ^{147}Nd and plotted as a relative mass-yield curve. The absolute fission yield of ^{147}Nd (and hence also the other products) was then obtained by adjusting the total yield under each half of the mass-yield curve to unity. The mass-yield curve was then replotted and is shown in Fig. 1. The accuracy of this normalization technique is probably about ±5–10%. Corrections have been applied to the fission yields for independent formation of later members

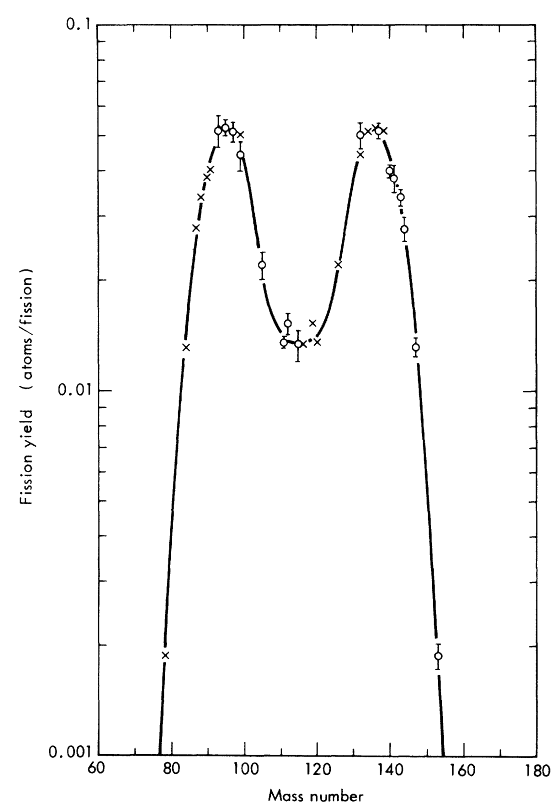

FIG. 1. Mass-yield curve for fission of ^{234}U with 14.8-MeV neutrons. Measured values are indicated by open circles with error bars; mirror points reflected about mass 115.5 are shown by crosses.

of the isobaric decay chains. The corrected mass yields are given in Table II when they differ significantly from the measured fission yield (the corrections are discussed later in this paper).

The low-yield products on the wings of the mass-yield curve are shown separately in Fig. 2. A Gaussian curve was fitted to these data and is also shown in Fig. 2. A least-squares procedure was used in which the data were weighted by the reciprocal of the square of their standard deviations. The Gaussian curve is given by yield = area $\times \exp\{-0.5[(A-A_0)/\sigma]^2\}/\sigma(2\pi)^{0.5}$, with area = 89 $\pm$ 19, $A_0 = 115.64 \pm 0.07$, and $\sigma = 9.55 \pm 0.10$.

^{236}U Results

The ^{236}U fission yield measurements are summarized in Table III. Each result is the average of about four or five separate measurements. The results were first calculated as the ratio of each product to ^{147}Nd. Two separate methods were used to obtain the absolute fission yield of ^{147}Nd.

The first method was the graphic technique used previously for the ^{234}U results. The ^{147}Nd fission yield was found to be 0.0172 by adjusting the yields of both halves of the mass-yield curve to unity.

The second method involves the measurement of the total number of fissions in the two irradiations in which monitor foils were used. The cross sections of the ^{89}Y$(n, 2n)^{88}$Y and ^{93}Nb$(n, 2n)^{92}$Nb reactions at 14.8 MeV are 1060 $\pm$ 50 and 487 $\pm$ 24 mb, respectively.[8] The integrated neutron fluence in the target foil was obtained by averaging the results from the monitor foils in front and back of the uranium. The fission cross section of ^{236}U at 14.8 MeV is 1.59 $\pm$ 0.06 b.[9] The number of fissions was calculated from the product of the number of ^{236}U target atoms, the fission cross section, and the neutron fluence. The fission yield of ^{147}Nd was found to be 0.0197, based on the yttrium monitor, and 0.0185 based on the niobium monitor.

The discrepancy between the results for the two

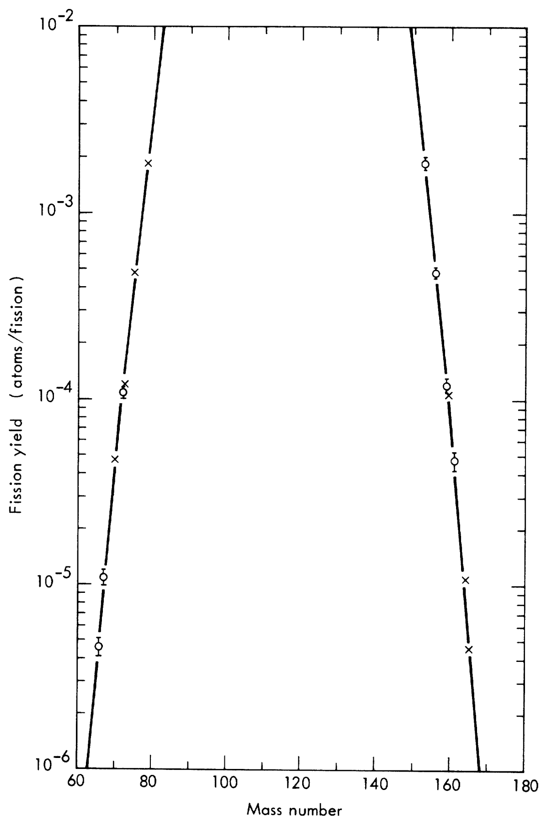

FIG. 2. Low-yield products from fission of ^{234}U with 14.8-MeV neutrons. Measured values are indicated by open circles with error bars; mirror points reflected about mass 115.6 are shown by crosses.

TABLE III. Yields of products from fission of ^{236}U with 14.8-MeV neutrons.

Product nuclide	Ratio to ^{147}Nd	Measured fission yield [a]
^{66}Ni	1.07×10^{-4}	$(1.91 \pm 0.19)\times 10^{-6}$
^{67}Cu	2.35×10^{-4}	$(4.2 \pm 0.4)\times 10^{-6}$
^{72}Zu	2.70×10^{-3}	$(4.82 \pm 0.15)\times 10^{-5}$
^{89}Sr	2.11	0.0378 $\pm$ 0.0019
^{91}Y	2.47	0.044 $\pm$ 0.005
^{93}Y	2.92	0.052 $\pm$ 0.005
^{95}Zr	3.04	0.0544 $\pm$ 0.0022
^{97}Zr	3.11	0.056 $\pm$ 0.004
^{96}Nb	8.10×10^{-4}	$(1.45 \pm 0.12)\times 10^{-5}$ [b]
^{99}Mo	2.96	0.053 $\pm$ 0.005
^{105}Rh	1.45	0.0259 $\pm$ 0.0021
^{111}Ag	0.626	0.0112 $\pm$ 0.0005
^{112}Pd	0.672	0.0120 $\pm$ 0.0006
$^{115m+g}$Cd	0.584	0.0105 $\pm$ 0.0010 [c]
^{132}Te	2.63	0.047 $\pm$ 0.005 [d]
^{136}Cs	0.0671	$(1.20 \pm 0.05)\times 10^{-3}$ [b]
^{137}Cs	2.96	0.0531 $\pm$ 0.0022
^{140}Ba	2.37	0.0425 $\pm$ 0.0018
^{141}Ce	2.35	0.042 $\pm$ 0.003
^{143}Ce	2.28	0.0408 $\pm$ 0.0021
^{144}Ce	2.02	0.0361 $\pm$ 0.0024
^{147}Nd	1.00	0.0179 $\pm$ 0.0010
^{153}Sm	0.162	$(2.90 \pm 0.24)\times 10^{-3}$
^{156}Eu	0.0409	$(7.3 \pm 0.3)\times 10^{-4}$
^{159}Gd	9.5×10^{-3}	$(1.69 \pm 0.17)\times 10^{-4}$
^{161}Tb	3.76×10^{-3}	$(6.7 \pm 0.4)\times 10^{-5}$
^{166}Dy	2.43×10^{-4}	$(4.3 \pm 0.3)\times 10^{-6}$
^{169}Er	7.0×10^{-5}	$(1.24 \pm 0.26)\times 10^{-6}$
^{172}Er	7.8×10^{-6}	$(1.4 \pm 0.6)\times 10^{-7}$

[a] The experimental standard deviations given here do not include a systematic uncertainty of about 6% in the normalization of the mass-yield curve.

[b] The ^{96}Nb and ^{136}Cs results are independent fission yields.

[c] The measured ^{115m}Cd/^{115g}Cd ratio is 0.0735.

[d] The estimated total chain yield for mass 132 is 0.049.

neutron monitors is probably due to the difference in excitation functions and the effect of lower-energy (scattered and fission-spectrum) neutrons. The $^{89}Y(n, 2n)^{88}Y$ reaction has a high threshold (11.6 MeV), and the cross-section curve does not resemble that of the ^{236}U fission reaction as well as does that for the $(n, 2n)$ reaction on ^{93}Nb. For this reason we discount the results based on ^{88}Y and use only those based on ^{92}Nb and on the graphic method. These results, 0.0185 and 0.0172, both have uncertainties of 5% or more; we chose to use the average, 0.0179 ± 0.0010, for the fission yield of ^{147}Nd. The yields of the other products were calculated from the measured ratio to ^{147}Nd.

Corrections for independent formation of later members of the isobaric decay chains have been estimated and found to be negligible in each case except for mass 132 (these corrections are discussed later). The mass-yield curve is shown in Fig. 3, and the low-yield products on the wings are shown separately in Fig. 4. The parameters for the Gaussian curve that was fitted to the latter data are as follows: area $= 113 \pm 16$, $A_0 = 116.80 \pm 0.05$, and $\sigma = 9.34 \pm 0.06$.

In addition to the yields given in Table III, the ratio of yields $^{126}Sb/^{127}Sb$ was also measured to be 0.083 ± 0.006. Difficulties with carrier-tracer exchange prevented us from measuring the absolute yields of both isotopes. The measured ^{126}Sb is that formed independently in fission, as it is shielded from β decay of precursors by 10^5-yr ^{126}Sn. We estimate the independent fractional chain yield of ^{126}Sb to be 0.096 ± 0.010, by using interpolated values for the mass-126 and -127 chain yields from the mass-yield curve.

Charge-Distribution Corrections

In another paper[3] we describe an empirical method for estimating charge-distribution corrections that we have used to infer total chain yields from measured fission yields. The corrections are made by estimating the value of Z_p, the most probable charge for each mass chain, from the equation $Z_p = Z_p(\text{ref}) + \Delta Z_p$. $Z_p(\text{ref})$ is the reference Z_p function for fission of ^{235}U with thermal neutrons. The values of ΔZ_p found for 14.8-MeV fission of ^{234}U are 0.42 (light fragment) and 0.69 (heavy frag-

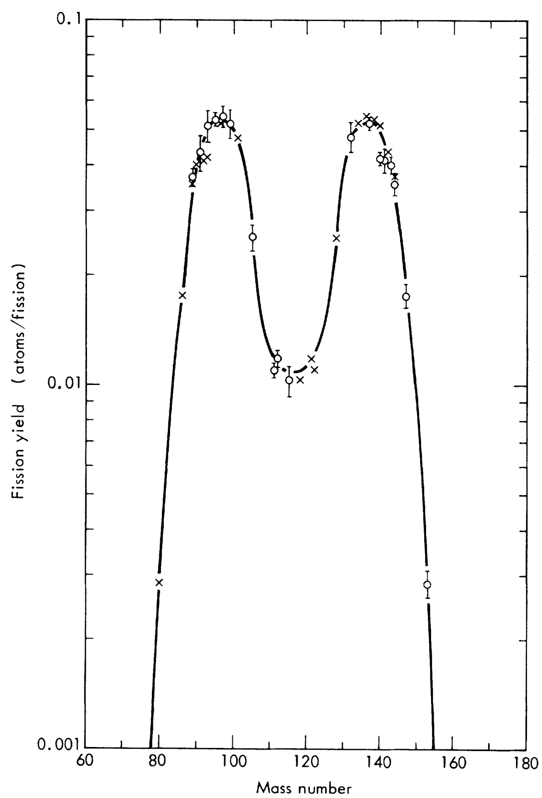

FIG. 3. Mass-yield curve for fission of ^{236}U with 14.8-MeV neutrons. Measured values are indicated by open circles with error bars; mirror points reflected about mass 116.5 are shown by crosses.

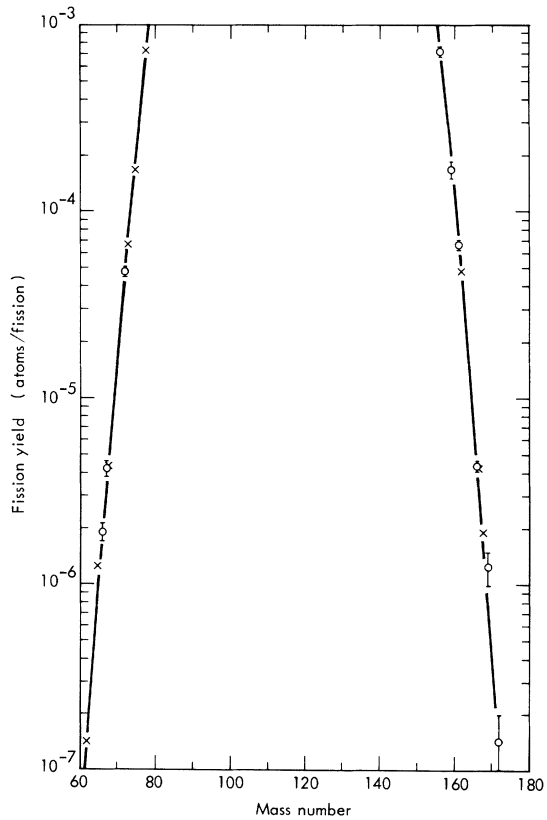

FIG. 4. Low-yield products from fission of ^{236}U with 14.8-MeV neutrons. Measured values are indicated by open circles with error bars; mirror points reflected about mass 116.8 are shown by crosses.

TABLE IV. Comparison of measured and calculated independent yields.

Uranium target	Product	Independent fractional chain yield Measured	Calculated
^{234}U	^{96}Nb	$(1.54 \pm 0.10) \times 10^{-3}$	2.0×10^{-3}
	^{136}Cs	0.095 ± 0.006	0.069
^{236}U	^{96}Nb	$(2.7 \pm 0.3) \times 10^{-4}$	2.8×10^{-4}
	^{126}Sb	0.096 ± 0.010	0.016
	^{136}Cs	0.022 ± 0.002	0.014

ment) and, for ^{236}U fission, 0.10 (light fragment) and 0.29 (heavy fragment). We used these values of ΔZ_p, together with the Z_p(ref) values of Wahl, Norris, Rouse, and Williams,[10] to estimate independent fractional chain yields of later members of each isobaric chain. We used a charge-dispersion curve with $\sigma = 0.56$.[10]

For the fission of ^{236}U, the corrections are all negligible except for mass 132. We estimated the independent yield of ^{132}I to be 3.7% of the total chain yield. For the fission of ^{234}U the corrections are larger, of course, and we estimated the independent yield of ^{132}I to be 14% of the total chain yield. The results of applying these corrections are included in Table II. In most cases the uncertainties in the Z_p(ref) function, the value of ΔZ_p, and the value of σ have a negligible effect on the estimated total chain yields.

We measured a number of independent yields for fission of ^{234}U and ^{236}U (see Tables II and III). The independent fractional chain yields for these nuclides were calculated by estimating the total chain yields from the smooth mass-yield curves in Figs. 1 and 3. Table IV compares these yields to those calculated from the estimated Z_p values and the standard charge-dispersion curve with $\sigma = 0.56 \pm 0.06$.[10] The calculated independent yields have an uncertainty of a factor of 2 in most cases (when the yield is small), because of the uncertainty in Z_p and in σ. The ^{96}Nb and ^{136}Cs yields are in good agreement with the measured values, while the estimated ^{126}Sb yield is much too low.

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