Yields of fission products produced by thermal-neutron fission of 229 Th

J. K. Dickens and J. W. McConnell Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830 (Received 24 May 1982)

Absolute yields have been determined for 47 gamma rays emitted in the decay of 37 fission products representing 25 mass chains created during thermal-neutron fission of 229 Th. Using a Ge(Li) detector, spectra were obtained of gamma rays emitted between 15 min and 0.4 yr after very short irradiations by thermal neutrons of a $15-\mu$ g sample of 229 Th. On the basis of measured gamma-ray yields and known nuclear data, yields for cumulative production of 37 fission products were deduced. The absolute overall normalization uncertainty is $<$ 8%. The results are compared with fission-product yields previously measured, with generally good agreement. On this basis, and using other measurements for masses not observed in the present experiment, a complete mass distribution for A between 76 and 152 was deduced. The measured A-chain cumulative yields from the present program make up 84% of the total light-mass $(A < 115)$ yield and 77% of the total heavy-mass yield. The data were analyzed to obtain values of most-probable charge (Z_p) and charge-dispersion (σ) parameters. Based upon insight gained from study of similar data obtained for thermalneutron fission of ²³⁵U, we postulate a simple functional dependence $\sigma = \sigma(Z_p)$, and using this dependence obtain values of $Z_{p}(A)$ for 15 mass chains created during fission of ²²⁹Th. Values of $Z_p(A)$ were estimated for other mass chains based upon results of a recent study of $Z_{p}(A)$. Charge distributions determined using the deduced mass distribution and the deduced sets of $Z_n(A)$ and $\sigma(Z_n)$ are in very good agreement with recent measurements, exhibiting a pronounced even-odd effect in elemental yields. These results may be used to predict unmeasured vields for 229 Th fission.

NUCLEAR REACTIONS Fission $^{229}Th(n_{th},f)$, measured fission product gamma-ray yields; deduced fission-product yields and element and mass yields.

INTRODUCTION

The isotope 229 Th is the lightest of the long-lived fissioning actinides available in sufficient quantity to study mass and charge distributions. Within the last year, results of two measurements have been reported for thermal-neutron fission of 229 Th; one by Gindler et al ¹ obtaining cumulative yields for individual fission products and deducing a complete mass distribution from their own and earlier²⁻⁴ data, and the other by Mariolopoulos et $al⁵$ obtaining the overall charge distribution for low-Z fission products $(Z=32-40)$ essentially by total x-ray measurements. The charge distribution which they obtained exhibits a very pronounced even-odd effect in measured yields of the light elements.⁵ The mass distribution obtained by Gindler et al .¹ exhibits a nominal fine structure and agrees reasonably well with a recent evaluation for the light-mass $(A < 115)$ distribution, but does not agree very well with most of the evaluated⁶ distribution for the heavy masses.

As part of an overall program⁷⁻¹⁰ to determin fission-product yields for a variety of fissioning systems, we obtained data for thermal-neutron fission of 229 Th. In our first report¹¹ of these data (which will be designated by the label I in the remainder of the present report), yields of 39 short-lived fission products for 2^{29} Th fission were given. An attempt was made to utilize these data in conjunction with the other reported¹⁻⁴ fission yields to determine charge distributions, but without success, as reported in I. What was determined was that the experimental data reported in I were in only moderate agreement with evaluated⁶ yields and, in particular, in rather substantial disagreement (by \sim 50%) for $A = 87, 91, 136, 144, 145, and 146$. The primary difficulties encountered in trying to deduce charge distribution parameters, as had been done for data for Cm fission⁹ and ²⁴⁹Cf fission,¹⁰ lay partly in the lack of sufficient fission-product yield data for $n_{\text{th}} + {}^{229}\text{Th}$ to try a two-parameter "best-fit" analysis for any mass, coupled with the pronounced even-odd effect⁵ in Z which rendered meaningless

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attempts to ascertain overall trends by, e.g., plotting independent yields as functions of $[Z-Z(A)]$ on probability paper where $Z(A)$ might be calculated using simple assumptions.¹² In this respect, there fore, while many new data were reported in I, the analysis was incomplete. In particular, there was no satisfactory procedure to relate the data in I with the results of Gindler et al ¹ nor with the results of Mariolopoulos et al ⁵ nor the latter two with each other.

After completing the data reduction of the remainder of our 229 Th fission yield data, it seemed that it would be necessary to adapt in some manner the characteristics of charge-distribution parametrizations evaluated¹³ to represent the (much more complete) data set for $2^{35}U$ fission. After some study we discovered a definite, albeit imperfectly defined, relationship between the two standard charge-distribution parameters, with the net result that an A-dependent charge distribution can be reduced to one parameter. On the assumption of the correctness of this observation, and using almost exclusively data obtained for this report and for I, we were able to perform a completely independent analysis, resulting in the desired comparisons among all of the various data^{$1-5$} for 229 Th fission.

EXPERIMENTAL DETAILS

The 229 Th sample for these studies was obtained from an ORNL nuclear materials stock that had been obtained by milking a sample of 233 U. At the time of our measurements, the sample was 60% by mass 229 Th (in the form of thorium nitrate) with the remainder being primarily 209 Bi. The 228 Th content was $\sim 7 \times 10^{-6}$; the ²³³U content was less than 1%. Several gamma-ray spectra were obtained prior to irradiation¹⁴; these indicated that the sample was free of fission products.

For our experiment, \sim 25 μ g of material (=15 μ g of 229 Th) was deposited and dried in a small polyethylene container of wall thickness ~ 0.5 kg/m² and covered with a lid of similar thickness. This container was then placed permanently inside a polyethylene capsule designed for pneumatic transfer to and from an irradiation position at the Oak Ridge Research Reactor. The neutron flux at the irradiation position was $\sim 5 \times 10^{13} n/cm^2$ s, and the ratio of thermal neutrons to resonance energy neutrons was \sim 30:1, measured using gold and manganese foils and calculated assuming an E^{-1} epithermal flux.

Data in the present paper were obtained from three irradiations of this sample. The first was for 150 s, and the taking of gamma-ray spectra commenced 15 min after the end of the irradiation. A well-shielded 90-cm³ Ge(Li) detector in a low background was used. The sample-to-detector distance was 0.2 m. Thirty-eight spectra were obtained over a period of six days. The second irradiation was for 1200 s. Following this irradiation, 45 gamma-ray spectra were obtained using the same 90 -cm³ detector and the same geometry; these measurements extended to five months after the irradiation. The third irradiation was for 120 s. Nineteen gammaray spectra were obtained using a high-resolution intrinsic germanium detector especially set up to study the low-energy portion of the gamma-ray spectrum with much better resolution than could be obtained with the 90 -cm³ detector. These last measurements were initiated 10 min after the end of the irradiation and were terminated within 20 h.

DATA REDUCTION

The primary experimental information deduced from the measured spectra are photon activities of particular gamma-ray transitions from decay of fission products produced during the irradiation or subsequently from decay of other fission products as a function of time following the irradiation. These were extracted from the spectral data using tech-
nigure discussed in datail in prior reports 9,10,15 niques discussed in detail in prior reports.^{9,10,15} A given gamma ray was assigned to the decay of a particular fission product by matching the observed gamma-ray energy with the expected energy. The assignment was corroborated (in most cases) by matching the apparent decay constant of the gamma-ray yield as a function of time with that of the assigned fission product. In some cases it was necessary to include a growth term due to decay of the parent fission product. To determine fissionproduct yields (C), both the efficiency $\epsilon(E_{\nu})$ and the fraction of the decay of the fission product giving the desired gamma ray (B) are required, as well as the number of fissions (n_f) determined for each run the number of fissions (n_f) determined for each run using known fission cross sections, ^{16, 17} sample mass and total neutron fluence to an accuracy of $\pm 13\%$. The branching ratios (B) are discussed in the next section since they are not part of the experimental program. The measured intensities for 47 gamma rays are collected in Table I. Tabulated uncertainties include statistical (random) uncertainties, $\Delta \epsilon(E_{\nu})$, and counting-rate associated uncertainties, but not the overall uncertainty in n_f .

RESULTS

Determination of the yield of a given fission product required knowledge of its nuclear properties, particularly the gamma-ray branching ratio, the half-life of the fission product, and the half-life of its parent. These are also tabulated in Table I.

TABLE I. Intensities of gamma rays associated with decay of fission products created by thermal-neutron fission of 229 Th and deduced fission-product yields.

E_{γ}	Yield per	Assigned	Gamma-ray	$T_{1/2}$	$T_{1/2}$ (parent)	Cumulative fission
(keV)	100 fissions ^a	fission product	branching ratio $(\%)^b$	(s)	(s)	product yield (%)
57.4	0.95 ± 0.05	143 Ce	12.2 ± 1.3	1.15E5	840	± 0.9 7.7
114.3	0.111 ± 0.006	149Nd	19.0 ± 2.2	6100	140	0.60 ± 0.07
127.9	0.165 ± 0.009	^{147}Pr	8.1 ± 0.4	834	11	2.02 ± 0.15
145.45	3.49 \pm 0.12	141 Ce	48.2 ± 0.3	2.81E6	1.4E4	7.24 ± 0.25
149.8	0.300 ± 0.017	131 Te	68. ± 1	1500	1380	0.44 ± 0.03
151.2	7.20 \pm 0.22	85 Kr*	78.3 ± 5.2	16130	172	9.20 ± 0.67
165.85	1.99 ± 0.08	139 Ba	23.8 ± 0.3	4962	570	8.37 ± 0.35
168.5	0.88 ± 0.05	93 Sr	19.7 ± 2.9	444	6	4.5 ± 0.7
181.1	0.90 ± 0.05	134 Te	18.0 ± 1.3	2508	10	5.02 ± 0.46
190.3	3.46 ± 0.10	141 Ba	± 3 46.	1080	25	7.53 \pm 0.54
196.3	1.91 ± 0.14	${}^{88}\text{Kr}$	26.3 ± 1.4	10200	16	7.24 ± 0.64
218.3	0.76 ± 0.04	146 Ce	20.5 ± 3.2	834	11	3.7 ±0.6
228.33	1.039 ± 0.024	132 Te	88.2 ± 0.2	2.75E5	168 (252)	1.18 ± 0.03
255.1	1.54 ± 0.08	142 Ba	21.2 ± 1.2	636	2	7.27 \pm 0.56
258.3	2.27 ± 0.23	$^{138}\mathrm{Xe}$	31.5 ± 1.3	850	7	7.21 \pm 0.79
266.9	0.342 ± 0.008	^{93}Y	6.8 ± 0.4	36900	446	5.02 ± 0.32
293.28	3.77 ± 0.09	143 Ce	43.4 ± 2.0	1.19E5	840	8.68 ± 0.43
312.1	1.03 ± 0.06	133 Te	72.6 ± 0.8	744	160	1.42 \pm 0.14
314.7	0.402 ± 0.027	147 Pr	± 1 22.	816	56	1.93 ± 0.16
316.76	1.80 ± 0.06	146Ce	52.5 \pm 5.7	834	11	3.44 \pm 0.39
343.7	1.00 ± 0.05	141 Ba	14.2 ± 1.2	1080	25	7.0 ± 0.7
356.6	2.35 ± 0.06	${}^{83}Se$	68.6 ± 0.5	1344	13	3.42 ± 0.09
364.5	0.503 ± 0.011	^{131}I	82.5 ± 0.4	6.94E5	(1500) 1.08E5	0.609 ± 0.014
402.7	3.16 ± 0.09	87 Kr	49.5 ± 1.6	4579	56	6.38 ± 0.27
487.0	3.68 ± 0.08	140 La	45.9 ± 1.4	1.15E5	1.11E6	8.01 ± 0.34
529.5	2.69 ± 0.06	133 I	87.3 ± 0.2	74880	745(133245	3.08 ± 0.06
531.0	0.253 ± 0.006	147 Nd	13.1 ± 0.8	9.56E5	816	1.93 ± 0.13
537.6	1.95 ± 0.05	140 Ba	24.4 ± 0.2	1.11E6	66	7.98 ± 0.20
555.63	3.60 ± 0.07	$91Y*$	95.1 ± 0.1	3000	34680	3.79 ± 0.08
566.0	0.900 ± 0.026	134 Te	18.9 ± 1.3	2508	10	4.76 ± 0.36
590.2	2.67 ± 0.10	93 Sr	$\pm\,8$ 73.	446	6	3.66 ± 0.42
647.5	0.189 ± 0.013	$^{133}Te*$	22.1 ± 2.4	3324	162	0.86 ± 0.11
657.9	0.673 ± 0.030	97Nb	98.34 ± 0.11	432	60840	0.685 ± 0.031 °
667.76	1.233 ± 0.033	^{132}I	98.7 ± 0.1	8222	2.75E5	1.25 ± 0.04^c
743.5	0.647 ± 0.014	^{97}Zr	92.8 ± 0.3	60840	4	0.697 ± 0.015
756.9	1.477 ± 0.033	95Zr	54.6 ± 0.5	5.53E6	618	2.70 ± 0.07
793.4	0.173 ± 0.018	130Sb	99.8 ± 0.1	2400	102	0.174 ± 0.018
881.0	4.08 ± 0.11	84Br	42. ± 3	1908	209	9.72 ± 0.56
884.31	3.89 ± 0.10	134 I	65.3 ± 1.0	3156	2508	5.95 ± 0.18
918.24	1.92 ± 0.05	$^{94}{\rm Y}$	$\pm 5^d$ 49.	1122	75	3.91 \pm 0.39
934.4	0.845 ± 0.022	92Y	13.9 ± 1.0	12600	9756	6.08 ± 0.47
1024.25	2.092 ± 0.042	91 Sr	33.5 ± 0.7	34680	59	6.25 ± 0.18
1131.5	1.135 ± 0.026	135 I	22.8 ± 0.4	23796	18	4.98 ± 0.14
1260.4	1.460 ± 0.031	135 I	29.0 ± 0.4	23796	18	5.03 ± 0.13
1383.9	4.89 ± 0.10	^{92}Sr	90. ± 10	9756	4.5	5.43 ± 0.61
1596.6	1.946 ± 0.044	$\rm ^{140}La$	95.4 ± 0.1	1.15E5	1.11E6	8.16 ± 0.18
1836.0	1.646 ± 0.037	88Rb	21.4 ± 1.2	1067	10200	7.69 ± 0.47

 $\overline{^{4}$ Does not include $\pm 13\%$ absolute normalization uncertainty.

 bV alues taken from *Table of Isotopes*, Ref. 18, or our previous evaluations, Refs. 7–10, except as noted.

"Yield of parent $(^{97}Zr$ and ^{132}Te).

 d Glendenin et al., Ref. 19.

Most of these data had been evaluated for our previous measurements, $7-10$ and the remaining data were obtained from the Lederer and Shirley compilation's or from a more recent measurement.¹⁹ The last column of Table I gives the deduced cumulative fission yields for the fission products listed in column 3. All of the required nuclear data were completely reviewed and details of these studies have been given.^{$7-10$} For most of the fission-product yields (C) obtained in this experiment, errors in $T_{1/2}$ contribute very little to an error in C. However, assigned uncertainties ΔB are very important in the evaluation of the assigned uncertainties ΔC .

Fission-product yields were obtained primarily for nuclides having half-lives between 7 min and 65 d

and having yields $> 0.4\%$. Yields were not obtained for all nuclides within this defined set, particularly those that decay almost entirely by beta-ray emission, but also for a few for which the primary gamma ray had nearly the same E_{γ} as a more intense gamma ray from another nuclide having a similar half-life, or for which the primary gamma ray was nearly degenerate with a strong gamma ray emanating from the decay of 229 Th and its daughters.¹⁴ In fact, the ²²⁹Th decay gamma rays were the primar interference for $E_\gamma < 300$ keV.

There were gamma rays observed for which photon yield per fission results were not obtained, even though the gamma ray could have been ascribed to decay of a specific fission product. We were satis-

TABLE II. Comparisons of measured cumulative fission product yields for thermal-neutron fission of ²²⁹Th. Measured

product	Present	ORNL (1982) ^a	ANL $(1981)^b$	ANL $(1966)^c$	McMaster $(1965)^d$	USSR (1968) ^e
${}^{83}Se$	3.42 ± 0.09	3.29 ± 0.15	2.54 ± 0.33			
84Br	9.72 ± 0.56		10.01 ± 0.72			
85 Kr*	9.20 ± 0.67		9.22 ± 0.30		8.40 ± 0.70	
87 Kr	6.38 ± 0.27	6.46 ± 0.35	7.21 ± 0.33		6.80 ± 0.57	
88 Kr	7.24 ± 0.64		7.51 ± 0.30		7.49 ± 0.63	
91 Sr	6.25 \pm 0.18		6.35 ± 0.20	5.7 ± 1.2		
^{92}Sr .	5.43 ± 0.61		5.40 ± 0.62			
$^{92}{\rm Y}$	6.08 ± 0.47					6.40 \pm 0.54
93 Sr	3.98 ± 0.42 ^f	3.91 ± 0.31				
93Y	5.02 ± 0.32		5.11 ± 0.20			4.40 \pm 0.27
94Y	3.91 ± 0.39		4.51 ± 0.43			
95Zr	2.70 ± 0.07		2.77 ± 0.08	2.6 ± 0.5		2.60 ± 0.20
^{97}Zr	0.689 ± 0.014 ^f		0.73 ± 0.03			0.61 ± 0.05
131 _T	0.609 ± 0.014		0.56 ± 0.02	$0.87 + 0.18$		0.430 ± 0.045
132 Te	1.21 ± 0.03 ^f		1.19 ± 0.04	1.23 ± 0.25		0.870 ± 0.054
133 Te	1.42 ± 0.14	1.49 ± 0.12				
133 I	3.08 ± 0.06		2.95 ± 0.09			4.00 \pm 0.96
134 Te	4.87 ± 0.33 ^f	5.72 ± 0.46	5.46 ± 0.43			
134 I	5.95 ± 0.18					5.30 ± 0.66
135 I	5.01 \pm 0.12 ^f		5.06 ± 0.16			
138 Xe	7.21 \pm 0.79	7.38 ± 0.64				
^{139}Ba	8.37 ± 0.35					8.96 ± 0.23
140 Ba	7.98 ± 0.20					
140 La	8.09 \pm 0.20		8.57 ± 0.25	7.2 ± 1.4		8.70 ± 0.65
^{141}Ba	7.53 ± 0.54	7.29 ± 0.72				
141 Ce	7.24 ± 0.25		7.51 ± 0.38	7.8 ± 1.6		7.83 ± 0.36
142 Ba	7.27 ± 0.56	7.25 ± 0.52				
143 Ce	8.36 ± 0.32		8.22 ± 0.25			8.87 ± 0.27
146 Ce	3.54 \pm 0.43	3.97 ± 0.24				
^{147}Nd	1.93 ± 0.13		2.40 ± 0.07			1.83 ± 0.27

^aReference 7.

 $149Nd$

Reference 1.

'Reference 2. We assign 20% uncertainties to quoted yields based upon discussion given in the reference.

^dReference 3. An average normalization of 7.77 ± 0.65 was applied to the reported mass spectrometric data.

 0.65 ± 0.03

Reference 4. We assign 7.5% uncertainties to results given without uncertainties in the reference.

Weighted average of several deduced yields.

 0.60 ± 0.07

Fission

fied with analyzing one or two gamma rays associated with decay of a given fission product, usually but not always the gamma ray with the largest branching ratio (B) . We attempted to obtain information for additional fission products, particularly for
⁹⁹Mo, 103 Ru, 127,129 Sb, and 136 Cs. However, the extracted information for each expected gamma ray was not adequate to determine an unambiguously correct assignment.

DISCUSSION

The cumulative yields for 31 fission products are compared with prior results^{1-4,7} in Table II. The agreement with our data in I is very good, not unexpectedly, since there was extensive cross checking between the two experiments. The agreement with the recent Argonne National Laboratory' (ANL) experiment is also good with 14 out of 21 comparisons within the combined quoted uncertainties. The earlier ANL data² in the fifth column have large uncertainties, and the only disagreement is for 131 I. The data for krypton isotopes in the sixth column³ are from mass spectrometric results relative to 86 Kr. We normalized these data to our own using an average normalization to give the closest overall comparison for the three krypton isotopes. The analysis is not as satisfactory as it may seem in the table since this normalization results in a yield for the spectrometric 84 Kr of (8.47±0.71)%, which is clearly low. The present results are in good agreement with the results⁴ in the last column. The primary conclusion from comparisons of all of the data in this table is that the present results are in reasonable agreement with all prior measurements. As a consequence the present yields will be used (rather than "average" values for these fission products) in the further analyses to obtain chain yields.

The charge distributions for all mass chains are necessary to complete the task of determining the mass distribution. For this purpose one would need to determine independent yields for all fission products created during fission of 229 Th, a very difficult task which has not yet been done. To make the maximum use of the available data, we utilize a systematic semiempirical description of measured fission yields, for there is not yet a satisfactory theory of fission which can be used to predict fission yields.

It has been known for some time^{12,20} that independent yields are approximately represented by Gaussian charge dispersions of the form

$$
P(A,Z) = (2\pi\sigma^2)^{-1/2}
$$

×exp(-0.5{[Z-Z_p(A)]/σ}²), (1)

where A is the mass number and Z the atomic number of nuclide (A,Z) . $Z_p(A)$ is the most probable nuclear charge for the fission-product decay chain of mass number A and σ the charge-dispersion parameter which may also be a function of A . The fractional independent yield for nuclide (A, Z) is then

$$
F^{i}(A, Z) = \int_{Z-1/2}^{Z+1/2} P(A, z) dz
$$
 (2)

and the fractional cumulative yield for nuclide (A, Z) is

$$
F^{c}(A, Z) = \int_{-\infty}^{Z+1/2} P(A, z) dz .
$$
 (3)

Analyses^{9, 10, 12, 21} of representative data sets for several fissioning systems indicate a constant $\sigma\simeq0.62$ ($\pm10\%$ depending upon the fissioning system) for all A will give satisfactory results when used in Eq. (2). These analyses are incomplete, however, because for any fissioning system [except ²³⁵U(*n*, *f*)] there are insufficient data to obtain Z_p and σ values from Eq. (2) for each A. Instead, an approximation to $Z_p(A)$ is computed using, for example, the unchanged charge distribution (UCD) hypothesis,¹² which is a hypothesis that assumes that no redistribution of charge occurs during fission (first suggested to explain high-energy fission²²). Then such fractional cumulative yields as have been determined are plotted versus $(Z - Z_{\text{UCD}})$, where Z_{UCD} is computed for each mass number A, including estimates for neutron emission as a function of A. Data plotted as probability graphs in this fashion for thermal-neutron fission of 245 Cm (Ref. 9) and 249 Cf (Ref. 10) exhibit a simple systematic behavior which can be represented by a single value (or at most two values) for σ .

However, for thermal-neutron fission of ^{235}U (and also for ²³³U) it has become evident that σ is not independent of A and that, where sufficient experimental data exist for a given decay chain, 2^{3-25} both $Z_n(A)$ and $\sigma(A)$ can be found approximately by the plotting of the cumulative yields against A on probability paper. The values of $Z_p(A)$ and $\sigma(A)$ thus obtained do not exhibit any well-defined functional dependence on A . It has been anticipated that these deduced variations in $Z_n(A)$ and $\sigma(A)$ must be related to odd-even effects, i.e., those (pairing) effects which tend to enhance even- Z (and perhaps even- N) nuclide production at the expense of odd- Z (odd- N) nuclide production observed in thermal-neutron fission of 235 U by Amiel and Feldstein.²⁵ It is clear from the measurements of Mariolopoulos et al.⁵ that there is a pronounced enhancement of the total yields for even-Z nuclides for Z between 32 and 40 for thermal-neutron fission of 229 Th. Thus, to obtain the needed charge distributions for this reaction, it will be necessary to rely upon analyses of the much more complete set of data for thermal-neutron fission of ^{235}U , and to attempt to adapt what can be deduced from such analyses to the present data for 229 Th.

In 1977, Crouch¹³ reported an evaluation of fission yields, including $n_{\text{th}} + {}^{235}U$. To account for the fact that the independent yields found experimentally for even-Z nuclides are larger than predicted yields obtained using Eqs. (1) and (2) and that the odd-Z nuclide yields are smaller than the predicted yields, the experimental results were represented by a modification²⁶ of Eq. (1) ,

$$
P(A,Z) = N(A)(1+K)(2\pi\sigma^2)^{-1/2}
$$

×exp(-0.5{[Z-Z_p(A)]/σ}²), (4)

where K is positive for even-Z radionuclides and negative for odd-Z radionuclides and $N(A)$ is a normalization factor which ensures that

$$
\sum_{Z} F^{i}(A, Z) = 1.0
$$
 (5)

for every A. All of the available data were subjected to a least-squares analysis to obtain the best results for $Z_p(A)$ and $\sigma(A)$, with a tabulation given¹³ for $Z_p(A)$ and $\sigma(A)$. As noted above, there are no obvious relationships between Z_p and A or σ and A. However, plotting $\sigma(A)$ as a function of $Z_p(A)$, as shown in Fig. 1(a), produces a remarkable relationship between σ and Z_p ; σ is "large" for odd Z_p and "small" for even Z_p , a relationship independent of A. This apparent odd-even effect is in addition to that already included in the analysis represented by the factor K.

As a check on the possibility (however small it might seem) that the relationship between σ and Z_n exhibited in Fig. 1(a} was a fortuitous result of Crouch's analysis, 13 Strittmatter's data and analysis²⁷ for the light-mass yields obtained in 1978 (and not included in Crouch's evaluation¹³) are shown in Fig. 1(b). Strittmatter's independently determined values of σ and Z_p are different from Crouch's values, but the essential characteristic is the same.

The first time we observed the relationship between $\sigma(A)$ and $Z_p(A)$ shown in Fig. 1 was thought to be the first time the effect had been noted. However, in a recent report by Lang et $al.^{28}$ on a very careful measurement of fission yields for ²³⁵U(n_{th} , f), these authors exhibit this same effect on their Fig. 13 as a function of light-mass kinetic energy. Values of σ and Z_p obtained from their Table 8 are very close to the Strittmatter results²⁷ shown in Fig. 1(b). Thus the modulation of σ with Z_p has been observed from three independent analyses of three different data sets, and may be considered to be confirmed. It is interesting that the amplitude of the modulation is larger for the Crouch evaluation¹³ than for the other two analyses, since the Crouch evaluation presumably accounted for the pairing effect using the constant K in Eq. (4) . It is also interesting, although we make no specific use of this observation, that the peaks of the modulation in Fig. $1(a)$ are mostly even-mass A, whereas the odd-mass A tend to have a less-pronounced effect. For analyses not using a constant K, as shown in Fig. 1(b), the odd-mass A values have larger amplitudes than in Fig. 1(a).

The particular data exhibited in Fig. ¹ are, of course, only for $n_{\text{th}} + {}^{235}\text{U}$; we assume, however, that the even-odd oscillatory character of σ as a function of Z_p also applies to the $n_{\text{th}} + {}^{229}\text{Th}$ fission-product data set. A sinusoidal functional dependence was assumed; specifica11y

$$
\sigma(Z_p) = 0.62 - 0.15 \cos(\pi Z_p) \tag{6}
$$

This functional relationship appears similar to one studied by Wahl.²⁰ Wahl utilized a least-squares analysis of all of the parameters of the Gaussian charge dispersion, including possible oscillations for σ , given, in his notation, by

$$
\sigma_Z(A) = \overline{\sigma}_Z - (\sigma_{\text{AMP}})\cos\{[Z_p(A) - 50]\pi\}
$$

FIG. 1. Charge dispersion parameter [σ in Eq. (1)] plotted as a function of most probable charge $[Z_p]$ in Eq. (1)l for analyses of fission product yields from thermalneutron fission of ^{235}U . The upper box (a) exhibits the data for these parameters deduced by Crouch (Ref. 13). The horizontal line at σ =0.61 represents the value of σ . used by Crouch for those A for which the independent yield data were insufficient for a two-parameter analysis. The lower box (b) exhibits the results deduced by Strittmatter (Ref. 27) for a completely different set of data. The horizontal line at σ = 0.625 represents only an approximate average of the points plotted in this box. Different symbols are used to delineate even and odd masses, so as to exhibit the lack of a definitive dependence of σ as a function of A. The lines in the upper box are included as eye guides only.

	$Z_i - 2$	Z_I-1					
$Z =$			Z_I (even)	Z_I+1	Z_I+2	Z_1+3	$Z_I + 4$
				Independent yields ^a			
0.0 $Z_p = Z_I +$	0.08	9.69	80.5	9.69	0.08		
0.1	0.05	7.10	79.1	13.6	0.19		
0.2	0.04	5.57	75.1	18.8	0.53		
0.3	0.04	4.72	69.3	24.5	1.43		
0.4	0.06	4.26	62.6	29.7	3.37	0.01	
0.5	0.08	3.96	55.8	33.5	6.59	0.05	
0.6	0.10	3.68	49.4	35.7	10.9	0.17	
0.7	0.12	3.36	43.7	36.7	15.7	0.42	
0.8	0.12	2.96	38.5	37.0	$20.6\,$	0.82	0.02
0.9	0.10	2.47	33.9	37.0	25.2	1.35	0.04
1.0	0.07	1.92	29.5	36.9	29.5	1.92	0.08
$1.1\,$	0.04	1.35	25.2	37.0	33.9	2.47	0.10
$1.2\,$	0.02	0.82	20.6	37.0	38.5	2.96	0.12
1.3		0.42	15.7	36.7	43.7	3.36	0.12
1.4		0.17	10.9	35.7	49.4	3.68	0.10
1.5		0.05	6.59	33.5	55.8	3.96	0.08
1.6		0.01	3.37	29.7	62.6	4.26	0.06
$1.7\,$			1.44	24.5	69.3	4.72	0.04
1.8			0.53	18.8	75.1	5.57	0.04
1.9			0.19	13.6	79.1	7.10	0.05
2.0			0.08	9.69	80.5	9.69	0.08
				Cumulative yields			
0.0 $Z_p = Z_I +$	0.08	9.77	90.2	99.9	100.0		
0.1	0.05	7.15	86.2	99.8	100.0		
0.2	0.04	5.61	80.7	99.5	100.0		
0.3	0.04	4.77	74.1	98.6	100.0		
0.4	0.06	4.31	67.0	96.6	100.0		
0.5	0.08	4.04	59.9	93.4	100.0		
0.6	0.10	3.79	53.2	88.9	99.8	100.0	
0.7	0.12	3.48	47.1	83.9	99.6	100.0	
0.8	0.12	3.08	41.6	78.6	99.2	100.0	
0.9	0.10	2.58	36.5	73.5	98.6	100.0	
1.0	0.08	1.99	31.5	68.5	98.0	99.9	100.0
1.1	0.04	1.39	26.5	63.5	97.4	99.9	100.0
1.2	0.02	0.84	21.4	58.4	96.9	99.9	100.0
1.3		0.42	16.1	52.9	96.5	99.9	100.0
1.4		0.17	11.1	46.8	96.2	99.9	100.0
1.5		0.05	6.64	40.1	96.0	99.9	100.0
1.6		0.01	3.38	33.0	95.7	99.9	100.0
$1.7\,$			1.44	25.9	95.2	100.0	$100.0\,$
1.8			0.53	19.3	94.4	100.0	100.0
1.9			0.19	13.8	92.9	100.0	100.0
2.0			0.08	9.77	90.2	99.9	100.0

TABLE III. Fractional yields $(\%)$ for Z relative to Z_n.

This table is set up for the integer " Z_I " an even number. Thus if the actual Z_p lies, e.g., between 54.0 and 54.9, the Z_I of this table is set equal to 54, and one may determine fractions yields for Z between 52 and 58. If the actual Z_p lies, e.g., between 37.0 and 37.9, the Z_I of this table is set to 36 and one may determine fractional yields between 34 and 40.

The overall least-squares analysis for $^{235}U + n_{th}$ fission resulted in $\overline{\sigma}_Z \simeq 0.53$ and $\sigma_{AMP} \approx 0$ to within the calculated uncertainty, and he concluded that the oscillation could be neglected for ²³⁵U + n_{th} and three

other fission reactions he studied $(^{233}U + n_{th},)$ $Pu + n_{th}$, and ²⁵²Cf spontaneous fission).

However, the existing data for $^{229}Th + n_{th}$ fission definitely substantiate the oscillatory behavior of $\sigma(Z_n)$, at least for this fission reaction; indeed, one empirically cannot correlate the various data sets for 229 ^Th fission unless some nonconstant relationship $\sigma = f(Z_n)$ is adopted. We used the functional dependence given in Eq. (6) because of its simplicity; the constants 0.62 and -0.15 were determined by trial and error to give reasonable results as well as to provide the needed correlation among the various data sets.

Using Eq. (6) reduces to one parameter $Z_p(A)$ needed to analyze the data to determine charge distributions for each A. Calculations of $F^i(A,Z)$ from Eq. (2) were carried out for integer values of Z and for Z_p in steps of 0.1 charge units. After some trial and error, a value for $K=0.25$ was adopted, and then for each Z_p the normalization factor $N(A)$ was computed, Eqs. (4) and (5), resulting in a complete set of $F^{i}(Z, Z_{p})$, which are tabulated in Table III. Also tabulated are $F^{c}(Z, Z_{p})$, which are the sums of

the $F^{i}(z, Z_{p})$ for $z \leq Z$. Table III is set up for Z (of the fission product) and $Z_p(A)$ as functions of an integer Z_I . Z_I is chosen an even number for convenience (the tabular data repeat for $Z_1 + 2$).

It is understood that the tabular values given in Table III are not expected to be the optimal representation of the behavior of the charge distribution for $n_{\text{th}} + {}^{229}\text{Th}$. They were used for the express purpose of determining charge distributions for many of the different A chains from the present data so as to determine chain yields from these data.

The next step, then, was to determine Z_p for as many chain yields as there are appropriate experimental data. There are two types of experimental yield data available for this task, and these are given in Table IV. The first type gives the results of analyzing gamma-ray-peak yield data as a function of decay time having well-defined growth-decay characteristics so that the fractional contribution of

^aData from I; Z_p adjusted for $F^c < 1.0$ of daughter.

^bIf 60% of Sr decays lead to population of Y^* .

'Data may be in error due to Xe gas loss.

Data from I except for 135 I (Table II) and 136 Xe.

 e Independent yield of 136 Cs from Gindler et al., Ref. 1.

Mass	Measured ^a	Inferred	$Z_p^{\ b}$	Mass	Measured ^a	Inferred	$Z_p^{\ b}$
≤ 76		0.05 ± 0.02		115	0.023 ± 0.003 ^c		
77	0.021 ± 0.003 ^c		(30.9)	116		0.020 ± 0.004	
78	0.32 ± 0.06^c		(31.4)	117	0.018 ± 0.002 ^e		
79		0.61 ± 0.08	(31.8)	118	0.017 ± 0.002 ^e		
80		1.15 ± 0.23	(32.2)	119		0.012 ± 0.002	
81		2.15 ± 0.22^d	(32.6)	120		0.010 ± 0.002	
82		4.08 \pm 0.41 ^d	(33.0)	121	0.009 ± 0.001 ^c		
83		5.83 ± 0.85 ^d	(33.4)	122		0.012 ± 0.002 ^d	
84	9.72 ± 0.56		34.1	123		0.013 ± 0.003	
85	9.20 ± 0.67		(34.4)	124		0.010 ± 0.003	
86	7.42 ± 0.74		34.7	125	0.007 ± 0.001 ^c		
87	6.64 \pm 0.27		(35.3)	126		0.007 ± 0.002	(48.7)
88	7.69 ± 0.47		35.6	127		0.021 ± 0.004	(49.1)
89	9.40 ± 0.72		36.1	128		0.061 ± 0.012	(49.5)
90	8.90 ± 1.41		(36.4)	129	0.084 ± 0.015 ^c		(50.0)
91	6.35 ± 0.19		36.9	130		0.23 ± 0.05	(50.4)
92	6.08 ± 0.47		(37.5)	131	0.609 ± 0.014		(50.8)
93	5.02 ± 0.32		38.1	132	1.25 ± 0.03		51.2
94	3.91 ± 0.39		38.2	133	3.08 ± 0.06		(51.6)
95	2.70 ± 0.07		38.5	134	6.02 \pm 0.19		52.2
96		1.13 ± 0.11^d	(38.8)	135	5.98 ± 0.15		52.7
97	0.714 ± 0.015		(39.3)	136		6.11 ± 0.19 ^f	52.9
98		0.20 ± 0.02^d	(39.7)	137		7.63 ± 0.25 ^f	(53.3)
99	$0.15 \pm 0.01^{\circ}$		(40.1)	138	8.58 ± 0.77		54.1
100		0.053 ± 0.011	(40.5)	139	8.38 ± 0.35		54.4
101		0.019 ± 0.004	(40.9)	140	8.05 ± 0.20		54.9
102		0.007 ± 0.002		141	7.37 ± 0.25		(55.1)
103	0.026 ± 0.004 c			142	7.58 ± 0.58		(55.4)
104		0.019 ± 0.004		143	8.36 ± 0.32		(55.8)
105	0.013 ± 0.002 ^c			144	9.39 $\pm 1.06^c$		(56.2)
106	0.012 ± 0.001 ^e			145	5.40 ± 0.79 ^g		(56.6)
107		0.009 ± 0.002		146	3.90 ± 0.26		(57.0)
108		0.010 ± 0.002		147	1.95 ± 0.13		(57.4)
109	0.012 ± 0.002 ^c			148		1.04 ± 0.21	(57.8)
110		0.017 ± 0.003		149	0.61 ± 0.07		(58.2)
111	0.023 ± 0.003 c			150		0.22 ± 0.05^d	(58.7)
112	0.020 ± 0.003 ^c			151		0.019 ± 0.004	(59.1)
113		0.016 ± 0.002		≥ 152		0.05 ± 0.02	
114		0.020 ± 0.004					
Sums	±2.5 84.3	±1.7 15.4			86.7 ±2.3	15.5 ±1.7	
Total	99.7 ± 3.0					102.2 ± 2.9	

TABLE V. Chain yields (%) for thermal-neutron fission of 229 Th.

'Does not include uncertainty in absolute normalization assigned to present results.

^bData from Table IV. Values in parentheses were obtained from Eq. (8) for the light masses and Eq. (9) less 0.4 charge units for the heavy masses.

'Data taken from Gindler et al., Ref. 1.

dInterpolated reflected data.

'Data taken from Borisova et al., Ref. 4.

From renormalized xenon data of Harvey et al., Ref. 3.

⁸From ¹⁴⁵Ce data of I.

the parent can be ascertained. (See Refs. 9 and 10 for details.) From the experimental ratios in the penultimate column of Table IV, one may search the calculated F^c in Table III to determine a range of Z_p . For example, for $A=139$,

$$
C^{c}(Cs)/[C^{c}(Cs) + C^{i}(Ba)] = (71 \pm 18)\% .
$$

The parent, Cs, has odd Z, so one looks in the column $Z = Z_1 + 1$, and finds $F^c \sim 89\%$ for $(Z_I+0.6)$ and $F^c \sim 53\%$ for $(Z_I+1.3)$. Hence the applicable range of Z_p is 54.6–55.3. In this manner, ranges of Z_p are given for seven masses (For $A=130$, both the Sn parent and Sb daughter are isomers.) The second type of data represents direct determinations of F^c from measured cumulative yields and estimated chain yields. This is a circular process, since the chain yields are subject to adjustments after the ranges of Z_p have been deduced. In every case, however, the adjustments to $C(A)$ are small and have no effect (<0.1 units of charge) in the deduced range of Z_p .

Finally, to determine the $C(A)$ given in Table V, yield data for specific fission products given in Table II are corrected for F^c determined from Table III from the "best" value of Z_p deduced from the results just discussed in Table IV. Other required values of Z_p , given in parentheses in Table V, were obtained from the equations for Z_p given in the recent report of Waldo *et al.*²⁹ as discussed below. All values of $Z_p(A)$ used are also included in Table V. For most \vec{A} , the cumulative fission-product yield for the fission product given in Table I is essentially the chain yield $C(A)$, since $F^c \sim 100\%$ for these fission products. For a few A, the required $F^c < 100\%$, and so $C(A)$ is larger than the yield obtained for the largest Z nuclide of that A . The largest such correction is for $A=90$, where $F^c(Kr)=0.67\pm0.07$ for $Z_n = 36.4 \pm 0.1$. The 10% uncertainty in F^c is quadratically included in the given $\Delta C(A = 90)$.

In the above fashion, $C(A)$ were determined for $A=84-95, 97, 131-135, 138-143, 145$ (from ¹⁴⁵Ce yield in I), 146, 147, and 149, and are given in the second column of Table V. These $C(A)$ are shown graphically in Fig. 2, where the heavy mass yields are plotted as a "reflection" of the light mass yields, are plotted as a Terrection of the fight mass yields
i.e., for $A_H = 230 - A_L - \overline{v}$, where $\overline{v} = 2.14$ neutron per fission.¹⁷ Also given in the second column of Table V are $C(A)$ deduced from data reported by Gindler et al.¹ for $A = 77, 78, 99, 103, 105, 109, 111,$ 112, 115, 118, 121, 125, 129, and 144, and from data reported by Borisova et al.⁴ for $A = 106$ and 118. In the third and seventh columns of Table V are data inferred from interpolation or from renormalized data of Harvey et $a\overline{l}$ ³. From the latter reference³ are $C(A)$ from data for Xe isotopes for $A = 136$ and 137. For the interpolated data, interpolations were first

FIG. 2. Measured mass yields deduced from the present analysis for thermal-neutron fission of 229 Th. The heavy-element yield is plotted as a reflection of the light element, shifted to account for \sim 2.2 neutrons per fission observed for 229 Th fission.

made of "reflected" data, that is, $C(A) \sim C(230 - A - \bar{v})$. Thus $C(96) \sim C(131.86)$, and $C(131.86)$ was determined by interpolating between the values of $C(A)$ as a power of A for $A=131$ and 132 [i.e., linear interpolation of the log of $C(A)$ versus the log of A. Uncertainties $\Delta C(A)$ assigned to these data reflect $\Delta C(A)$ assigned to the "reflected" vields. The remaining $C(A)$ in columns 3 and 7 were determined by interpolation between $C(A_1)$ where $A_1 < A$ and $C(A_2)$ where $A_2 > A$; these interpolated $C(A)$ have assigned uncertainties of $\pm 20\%$ of $C(A)$.

The last two rows of Table V indicate partial and total sums of yields. The uncertainties to these sums reflect partial correlations among the individual $C(A)$, but not an overall normalization uncertainty associated with the determination of the number of fissions, n_f . The uncertainty in n_f comes primarily from the uncertainties in the fission cross sections' for ²²⁹Th fission, and is $\sim 13\%$.⁷ This is the overall uncertainty in normalization quoted in I. The "total" sums given in Table V suggest that the uncertainty in overall normalization should be less than \sim 13%. A difficulty in determining a better value is the fact that 16% of the light- and heavy-mass yields are inferred (from Table V) and one may question whether the $\sim 10\%$ uncertainties assigned to the inferred sums are realistic. On the other hand, it seems unlikely that either inferred summed

contribution is in error by a factor of 2, and therefore the measured sums should be valid to better than $\pm 10\%$. We suggest an overall normalization uncertainty of $\pm 7\%$ as a best judgement compromise between $\pm 3\%$ indicated in the last row of Table V and the $\pm 13\%$ assigned to Δn_f .

A check on the $C(A)$ given in Table IV was performed by determining the first moments of the light- and heavy-mass distributions. From these data $M_L = 87.8$ and $M_H = 139.9$. The estimated number of neutrons per fission is given by $230-M_L-M_H=2.3$, a value close to the adopted value¹⁷ of 2.14 \pm 0.04.

Elemental yields $C(Z)$ were computed from

$$
C(Z) \sum_{A} F^{i}(Z, A) C(A) , \qquad (7)
$$

where the $F^{i}(Z, A)$ were taken from Table III for values of $Z_n(A)$ given in Table V. To do so required determining all of the $Z_p(A)$ needed, and as mentioned above, for Z_p not obtained from the analysis summarized in Table III we adopted the formulae of Waldo et al.

$$
Z_p(A < 116) = 0.4153A - 1.19
$$

+ 0.167(236 - 92A_F/Z_F), (8)

$$
Z_p(A > 116) = 0.4153A - 3.43
$$

+ 0.243(236 - 92A_F/Z_F) , (9)

where $A_F = 230$ and $Z_F = 90$ for the present data set. Then $C(Z_L)$ were compared with $C(Z_H = 90 - Z_L)$, and the comparisons were not as satisfactory as desired. Much of the disagreement was resolved by a uniform subtraction of 0.4 charge units from $Z_p(A > 116)$ of Eq. (9), and these modified Z_p are those tabulated in the last column of Table V. The results for the $C(Z)$ computations are given in Table

VI, including data of Mariolopoulos et $al⁵$ for light elements for comparison. The character of the even-odd effect in $C(Z)$ reported by Mariolopoulos et $al⁵$ is well defined in the present results; there are, however, moderate quantitative differences. The tabular data indicate that the present $C(Z_L)$ compare reasonably well with the $C(Z_H)$. It is likely that further "adjustments" to the calculated Z_p [of Eqs. (8) and (9)] would provide even better agreement not only of the present $C(Z_L)$ with $C(Z_H)$ but also the present $C(Z_L)$ with the prior $C(Z_L)$. It is also quite likely that adjustments to the constants of Eq. (6) and/or to the constant K of Eq. (4) would further improve the desired agreements among the computed $C(Z)$. During the course of this study we tried several different "constants" in these last two mentioned equations, and observed only small changes in computed $C(Z)$ values. Most important, however, was that using different constants did not alter at all the determinations of $C(A)$ given in Table V.

CONCLUDING REMARKS

The *primary* data of this experiment are the gamma-ray yields given in the second column of Table I. Using literature values for gamma-ray branching ratios and radionuclide half-lives, observed gamma-ray yields were ascribed to decay of specific fission products, and cumulative fissionproduct yields were determined for these fission products. The major weakness of this method is the need to rely on the nuclear data, especially the gamma-ray branching ratios, any one of which may be incorrect by more than the assigned uncertainty. Future evaluations of the $n_{\text{th}}+{}^{229} \text{Th}$ fission yields should seek and use current values for these ratios, and we encourage adjustments to deduced fission-

	Present results ^a	Previous results ^b			
Heavy element	Yield $(\%)$	Light element	Yield $(\%)$	Light element	Yield $(\%)$
58 Ce	4.1 ± 0.4	32 Ge	4.5 ± 0.6	vG c	$5.2 + 0.4$
57 La	7.1 ± 0.9	33As	6.2 ± 1.0	33As	4.3 ± 0.4
56 Ba	25.6 ± 2.6	34 Se	22.6 ± 2.0	34Se	27.7 ± 0.7
55 _S	14.8 ± 1.1	35Br	13.3 ± 1.2	35Br	12.8 ± 0.8
54Xe	24.2 ± 1.9	36 Kr	24.9 ± 2.2	36 Kr	22.6 ± 0.6
$_{53}$ I	9.9 ± 0.8	37Rb	9.3 ± 1.1	37Rb	12.3 ± 0.7
52 Te	13.0 ± 0.9	₃₈ Sr	14.3 ± 1.2	38Sr	10.0 ± 0.5
51Sb	2.4 ± 0.8	39Y	3.4 ± 0.3	39Y	3.2 ± 0.6
50 Sn	$0.9 + 0.1$	$_{40}Zr$	$1.0 + 0.1$	$_{40}Zr$	2.0 ± 0.6

TABLE VI. Yields of complementary elements for thermal-neutron fission of 229 Th.

^aIncludes 7% absolute normalization uncertainty.

 b Mariolopoulos *et al.*, Ref. 5.</sup>

product yields as dictated by improved nuclear data.

From the deduced fission-product yields in Table II combined with those for short-lived fission products in I, supplemented by data obtained by other $g_{\text{r} \text{c} \text{u} \text{r}}^{1-\frac{1}{2}}$ for those fission products not unambiguously observed in the present series of experiments, we deduced a complete set of mass yields $C(A)$ given in Table V. This task involved determining a clearly nonunique set of independent yields given in Table III as well as a clearly nonunique set of mostprobable charges Z_p given in Table V. We do not claim that the set of $Z_p(A)$, or the $Fⁱ[Z, Z_p(A)]$ of Table III, are optimal; it seems likely, however, that the overall series of computations to obtain these variables are generally valid, and that the strong functional dependence of $\sigma = \sigma(Z_p)$ as exhibited in Fig. ¹ is a real effect and can be utilized as done in this paper.

The mass distribution as shown in Fig. 2 seems internally consistent except for $A=93$, 94, and/or 134, 135. The $C(A=94)$ depends upon the cumula-

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tive fission yield for $94Y$ for which we used a smaller gamma-ray branching ratio¹⁹ than given in the evaluated literature.¹⁸ Gindler *et al.*¹ obtained $C^{c}(94Y)=(4.5\pm0.4)\%$, a value somewhat larger than ours, but not sufficiently so to resolve the observed difference. The mass yields for $A=84-90$ appear to be in quite good agreement with the complementary mass yields for $A = 138 - 144$, especially considering the number of variables and calculations required to obtain these $C(A)$. Indeed, the $C(A)$ and $Z_n(A)$ values given in Table V combined with independent yield values given in Table III can be used to compute any independent yield for $n_{\text{th}} + {}^{229}\text{Th}$ fission, perhaps more reliably than using the present Evaluated Nuclear Data File evaluation, 6 based as it was only on the data in Refs. ²—4.

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