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Nuclear-charge distribution near symmetry for thermal-neutron-induced fission of ^{235}U

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Available data concerning nuclear-charge distribution for thermal-neutron-induced fission of ^{235}U have been evaluated for the region near symmetry ($104 \leq A \leq 129$, $42 \leq Z \leq 50$). Evaluated data or averages are listed and are interpreted by the use of empirical Z_P and A'_P models. The parameters and functions derived for the two models are presented and are compared with each other and with the data. Both models represent most data satisfactorily, and they permit the calculation of more reliable estimates of unmeasured independent-fission-product yields near symmetry than has been possible in the past. The two models show the same major trends, so the physical phenomena deduced from the trends are not model dependent. The independent yields of fission products with Z values of 50 and 42 are much larger than those of products with Z values of 43 through 49. The yields for products with Z values of 49 and 43 are larger than those of products with Z values of 44 through 48. These yield differences indicate that the stable filled, or nearly filled, 50-proton shell in the heavy fragment causes the fission probability to increase rapidly with mass number in the range from 120 to 130 between improbable symmetric and probable asymmetric fission. The lack of even-odd-proton and -neutron effects near symmetry indicates that excitation energies are higher for ^{236}U nuclei undergoing symmetric fission than for those undergoing asymmetric fission.

I. INTRODUCTION

Few data concerning nuclear-charge distribution near symmetric mass and charge division have been available until recently, and most of the available data are for thermal-neutron-induced fission of ^{235}U. In this paper, data for products from thermal-neutron-induced fission of ^{235}U with $104 \leq A \leq 129$ and $42 \leq Z \leq 50$ are evaluated, and average or selected values are interpreted by the use of two empirical models. One, the conventional Z_P model,[1–5] describes the dispersion of fractional-independent yields for each A about Z_P, the most probable nuclear charge.[5] The other, the A'_P model,[1,3,4] describes the dispersion of independent yields for each pair of Z's ($Z_L+Z_H=Z_{\rm fis}$) about A'_{P_H}, the most-probable average heavy-precursor mass number. (See Ref. 6 for definition of symbols.) Derived functions for the two models are compared with the data, with each other, and where possible, with predictions of the scission-point theory.[7] Observations are made concerning the influence of the 50-proton shell and nucleon-pairing effects on fission yields near symmetry.

For products from thermal-neutron-induced fission of ^{235}U that contribute to the peaks of the mass-yield curve, functions from both the Z_P and the A'_P models are reasonably well known and have changed little in recent years.[1–3,8] For example, the two solid lines in Fig. 1 represent similar ΔZ functions,[1,8] ΔZ being the difference between Z_P and $Z_{\rm UCD}$, the Z for unchanged charge distribution (UCD) as defined in Ref. 6. The dashed lines in Fig. 1, representing several estimated ΔZ functions, illustrate how poorly the ΔZ function has been characterized below $A'_H=130$.

Radiochemical data reported in 1958 and 1962 suggested that Z_P remained close to $Z=50$ for several mass numbers just below $A=130$, possibly resulting in a sharp break in the ΔZ function near $A=130$.[9,10] The evidence was fragmentary, however, so a linear extrapolation of the ΔZ function from -0.45 at $A'=130$ to 0.0 at symmetry ($A'=118$) was proposed in 1969 for thermal-neutron-induced fission of ^{235}U (curve C, Fig. 1).[8] Similar extrapolations were made for other fission reactions,[11] and these have been used for estimating unmeasured independent yields in the Evaluated Nuclear Data Files, including ENDF/B-V.[12,13] Predictions from the scission-point theory[7] showed no sharp break near $A'=130$ and approxi-

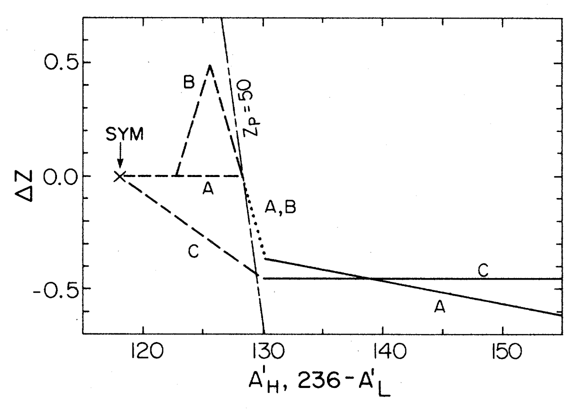

FIG. 1. Previous representations of ΔZ vs average heavy-fragment mass number, A'_H. (——), linear ΔZ functions for peak yields: A from Ref. 1 [$\Delta Z(A'=140)=-0.47$, $\partial\Delta Z/\partial A'=-0.01$], C from Ref. 8 [$\Delta Z(A'=140)=-0.45$, $\partial\Delta Z/\partial A'=0.00$]; $(\cdot\cdot\cdot\cdot)$, ΔZ function (A,B) near $Z_P=50$ from Refs. 1 and 3; (— — —), estimated ΔZ functions near symmetry: A from Ref. 1, B from Ref. 3, C from Ref. 8; ($\times$), symmetry ($A'_H=118$, $\Delta Z=0.0$).

mated the linear extrapolation (line C in Fig. 1) in approaching symmetry, as shown by the dashed line in Fig. 2, to be discussed.

During the 1970's, data[14,15] from the LOHENGRIN fission-product-recoil separator, as well as new radiochemical data,[16,17] showed that a sharp break in ΔZ near $A'=130$ did indeed occur, as illustrated in Fig. 1 by the dotted line (labeled A,B), which represents common portions of ΔZ functions from recent versions[1,3] of the Z_P model derived from these and other data. An earlier version[18] proposed that the ΔZ break occurred abruptly from -0.45 at $A=130$ to 0.00 at $A=129$. However, the behavior of the ΔZ function nearer symmetry was still unknown,[1-3,19] as were the behaviors of functions for the A'_P model near symmetry.

Recent yield measurements for indium[19,20] and technetium[14,15,21,22] fission products and for members of the $A=121$ decay series,[23] coupled with data[24] from earlier measurements for fission products near symmetry, now allow some inferences concerning the systematics of nuclear-charge distribution near symmetry for thermal-neutron-induced fission of ^{235}U. Some aspects of the systematics have been discussed previously,[19,23] but the systematics are treated more fully and quantitatively in this paper.

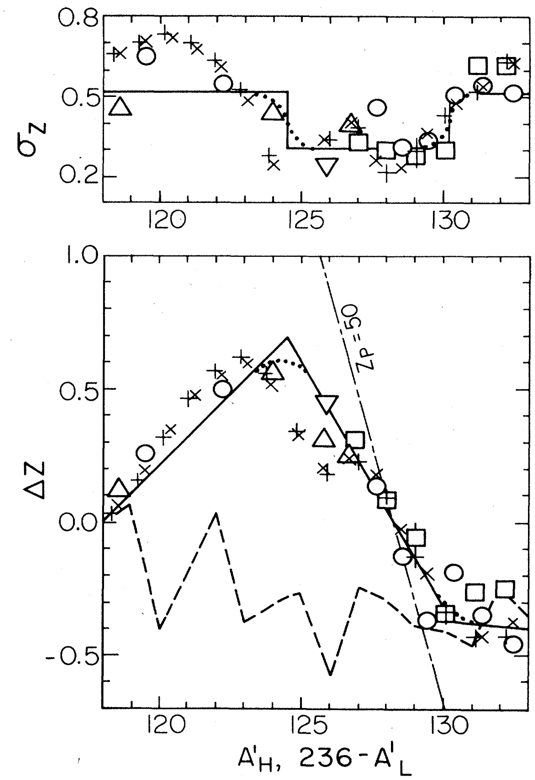

FIG. 2. New representations of ΔZ and σ_Z near symmetry versus the average heavy fragment mass number, A'_H. (○), heavy and (□), light fission-product values derived from three or more FI values; (△), heavy and (▽), light fission-product ΔZ and σ_Z values derived from one FI value, σ_Z or ΔZ being taken from the line in the other plot; (×), heavy and (+), light fission-product $\Delta \bar{Z}$ and $\sigma_{\bar{Z}}$ values calculated from the A'_P model ($\Delta \bar{Z}$ and $\sigma_{\bar{Z}}$ are defined in Ref. 45); (——), derived functions described in the text with parameters from Table III; (· · · ·), manually rounded functions; (— — —), ΔZ from the scission-point theoretical model (Ref. 7). Values for points with $A'_H > 130.2$, shown for continuity, were calculated from the current data file (Ref. 24). The calculated σ_Z value for ^{125}Sn and the calculated $\sigma_{\bar{Z}}$ values for $A=109$ and $A=124$ are not shown because dispersion widths are very small, and calculated values are <0.1 or indeterminate.

II. TREATMENT OF DATA

Measured yields of individual fission products were collected from the literature and placed in a computer file.[24] Independent (IN) and cumulative (CU) yields were divided by mass-number yields[12] (Y_A) to give fractional-independent (FI) and fractional-cumulative (FC) yields. Small corrections were applied to FC values to convert them to FI values.[25]

For each fission product the FI values were averaged with weights of $1/\sigma^2$, σ being the reported experimental error or the evaluator's error,[26] if assigned. For mass numbers with nearly complete sets of experimental FI values [$\sum$(FI)>0.9], experimental values and small estimated values[25] were normalized so that $\sum$(FI)$=1.00$.

The average FI values for fission products with $104 \leq A \leq 129$ and $42 \leq Z \leq 50$ are listed in Table I with uncertainties, which are the larger of the internal error $[1/\sum(1/\sigma^2)]^{1/2}$ or the external error (the internal error times the square root of the weighted variance). The IN values, used with the A'_P model, are also listed in Table I. They are products of FI and Y_A (Ref. 12) values, with appropriate propagation of errors. Reference numbers to the literature values used are listed in the fourth column of Table I. The $\bar{\nu}_P$ values,[19] used to estimate average precursor-fragment mass numbers, A', are listed in the fifth column of Table I. The last two columns contain calculated FI values to be discussed.

A number of yield values obtained by different methods of measurement are inconsistent. Possible sources of systematic error for several methods are discussed elsewhere.[19] The data sets with inconsistent values were analyzed separately because of difficulty in deciding which were the more reliable. The results of the separate analyses were compared to aid in deriving the unified nuclear-charge-distribution systematics presented here. Subsequently, some values were assigned evaluator's errors,[26] other values were normalized (footnote e to Table I), and limits and some values were excluded. Values, other than limits, that were not used for determining averages or model parameters are listed in Table II. The average values from Table I, where available, are shown in Table II for comparison, together with the calculated values from the Z_P and A'_P models to be discussed.

III. EMPIRICAL MODELS

Empirical models derived from experimental data correlate the systematics of nuclear-charge distribution in

TABLE I. Experimental and calculated fractional independent yields (FI), experimental independent yields (IN), and average numbers of neutrons ($\bar{\nu}_p$) emitted to form fission products.

Fission product	Experimental FI[a]	Experimental IN (%)[a]	References to sources of data used	$\bar{\nu}_p$	Calculated FI[b] Z_P model	Calculated FI[b] A'_P model
^{100}Mo	0.015 ±0.013	0.094 ±0.081	15,27,28	1.70	0.0033	0.015
^{101}Mo	0.036 ±0.011	0.186 ±0.058	14,15,27–29	1.76	0.026	0.067 (−2.8)
^{102}Mo	0.132 ±0.026	0.569 ±0.111	14,15,27,28	1.81	0.156	0.193 (−2.4)
^{103}Mo	0.312 ±0.034	0.946 ±0.103	14,15,27,28	1.86	0.377 (−1.9)	0.378 (−1.9)
^{104}Zr	0.066 ±0.013	0.126 ±0.026	14,15,27	1.91	1.3×10^{-4} (4.9)	0.017 (3.7)
^{104}Nb	0.308 ±0.022	0.591 ±0.043	14,15,27,29	1.91	0.337 (−1.3)	0.346 (−1.7)
^{104}Mo	0.624 ±0.034	1.196 ±0.068	14,15,27	1.91	0.660 (−1.1)	0.634
^{104}Tc	0.0018±0.0018	0.0034±0.0034	21,22	1.91	0.0025	0.0031
^{105}Nb	0.179 ±0.017	0.172 ±0.016	14,15,27,29	1.96	0.134 (2.7)	0.185
^{105}Mo	0.816 ±0.026	0.786 ±0.028	14,15,27	1.96	0.849 (−1.3)	0.798
^{105}Tc	0.003 ±0.007	0.003 ±0.007	21,22	1.96	0.017 (−2.1)	0.015 (−1.8)
^{106}Nb	0.039 ±0.017	0.016 ±0.007	14,15	2.01	0.037	0.058 (−1.2)
^{106}Mo	0.906 ±0.037	0.364 ±0.015	14,15	2.01	0.888	0.869
^{106}Tc	0.055 ±0.008	0.022 ±0.003	14,15,22	2.01	0.076 (−2.7)	0.072 (−2.3)
^{107}Nb	0.016 ±0.017	0.0024±0.0025	14,15	2.06	0.0066	0.015
^{107}Mo	0.798 ±0.040	0.117 ±0.007	14,15	2.06	0.766	0.683 (2.9)
^{107}Tc	0.183 ±0.040	0.027 ±0.006	14,15,22	2.06	0.227 (−1.1)	0.303 (−3.0)
^{108}Tc	0.470 ±0.170	0.024 ±0.009	22	2.11	0.474	0.734 (−1.55)
^{117}Ag	0.316 ±0.076	0.004 ±0.001	30	1.58	0.333	0.318
^{118}Pd	0.283 ±0.033	0.0030±0.0006	31	1.49	0.314	0.338 (−1.66)
^{118}Ag	0.550 ±0.100	0.0058±0.0014	31	1.49	0.606	0.487
^{118}Cd	0.157 ±0.105[c]	0.0017±0.0012		1.49	0.072	0.134
^{121}Ag	0.117 ±0.050	0.0015±0.0007	23	1.20	0.127	0.132
^{121}Cd	0.610 ±0.090	0.0079±0.0013	23	1.20	0.657	0.523
^{121}In	0.240 ±0.080	0.0031±0.0011	23	1.20	0.212	0.339 (−1.2)
^{121}Sn	0.030 ±0.040	0.0004±0.0005	23	1.20	0.0033	4.9×10^{-5}
^{123}Sn	0.124 ±0.055	0.0020±0.0009	16,19	1.01	0.161	0.0078 (2.1)
^{125}Sn	0.321 ±0.128[d]	0.0093±0.0037	19,20	0.83	0.510 (−1.5)	0.252
^{126}In	0.313 ±0.133[e]	0.0178±0.0077	20	0.73	0.269	0.367
^{127}In	0.274 ±0.097	0.0345±0.0123	19,20	0.64	0.114 (1.7)	0.113 (1.7)
^{127}Sn	0.661 ±0.098[c]	0.0833±0.0128		0.64	0.865 (−2.1)	0.842 (−1.8)
^{127}Sb	0.057 ±0.010	0.0072±0.0013	32	0.64	0.022 (−3.5)	0.045 (1.2)
^{128}In	0.061 ±0.031	0.0215±0.0108	19,20	0.54	0.036	0.030
^{128}Sn	0.888 ±0.038[c]	0.312 ±0.016		0.54	0.887	0.858

TABLE I. (*Continued*).

Fission product	Experimental FI[a]	Experimental IN (%)[a]	References to sources of data used	$\overline{\nu_p}$	Calculated FI[b] Z_P model	A'_P model
^{128}Sb	0.049 ±0.022	0.0172±0.0078	17	0.54	0.076 (−1.3)	0.112 (−2.9)
^{129}In	0.042 ±0.022[e]	0.031 ±0.016	20	0.45	0.0085 (1.5)	0.0074 (1.6)
^{129}Sn	0.840 ±0.051[c]	0.628 ±0.054		0.45	0.791	0.723 (2.3)
^{129}Sb	0.114 ±0.046	0.085 ±0.035	17	0.45	0.201 (−1.9)	0.265 (−3.3)
^{130}In	0.0043±0.0020[e]	0.0078±0.0036	20	0.39	0.023 (−9.2)	0.0018 (1.3)
^{130}Sn	0.680 ±0.045[c]	1.226 ±0.085		0.39	0.645	0.553 (2.8)
^{131}In	0.0033±0.0016[e]	0.0096±0.0045	20	0.40	0.0044	3.5×10^{-4} (1.9)
^{131}Sn	0.293 ±0.028	0.864 ±0.082	17,33–36	0.40	0.307	0.311
^{132}In	$(9.7 \pm 5.4)10^{-4}$[e]	0.0042±0.0023	20	0.45	3.1×10^{-4} (1.2)	6.3×10^{-5} (1.7)
^{132}Sn	0.135 ±0.009	0.583 ±0.040	34,36,37	0.45	0.110 (2.7)	0.144

[a]Taken from a computer file containing an evaluated compilation of nuclear-charge-distribution data (Ref. 24).
[b]Values in parentheses are the following: (exp FI − calc FI)/(error exp FI) = AF; absolute values < 1.0 are not listed. Reduced-chi-square values, $[\sum(AF)^2/N]^{1/2}$, are 2.05 and 1.72 for the calculated Z_P- and A'_P-model values, respectively. $N = 44$, the number of observations.
[c]Derived by difference: $\mathrm{FI}(Z) = 1.0 - \sum_{i \neq Z} \mathrm{FI}(i)$.
[d]An evaluator's error of 0.005 was assigned to the mass spectrometric ^{125}In CU value (Ref. 20) of 0.022±0.002 to achieve a weight for the derived FC value approximately equal to that for the radiochemical FC value (Ref. 19), with which it was averaged.
[e]The average FI values for ^{127}In and ^{128}In are ~60% lower than values that would be derived from the mass-spectrometric CU values alone. Because ratios of mass-spectrometric yields should be more reliable than absolute yield values, yields for ^{126}In, ^{129}In, ^{130}In, ^{131}In, and ^{132}In were multiplied by 0.6±0.2, the uncertainty being estimated.

fission with parameters for simple mathematical functions.[1–4] The model parameters are useful for deducing shell and pairing effects and are more convenient for comparison with theory than are individual yields for many hundreds of fission products. The parameters and functions are also useful for estimating many hundreds of independent yields that have not been measured.[1,4] Estimated independent yields are useful for planning experiments and are needed for the complete yield sets used in nuclear-reactor design and evaluation.[13,43]

The dispersion in yields for both the Z_P and A'_P models is assumed to be Gaussian, but yields are modulated by proton- and neutron-pairing effects. The effects are applied by multiplication or division of Gaussian yields by $\overline{F_Z}$ and $\overline{F_N}$, the average even-odd-proton and -neutron factors (previously called[1–4,23] $\overline{EOZ}$ and $\overline{EON}$, respectively). The average Gaussian width parameters for the Z_P and A'_P models are represented by the symbols $\overline{\sigma_Z}$ and $\sigma_{A'}$, respectively; the values of these parameters are equal to the root-mean-square (σ_r) values for Gaussian dispersions corrected for grouping $[\sigma = (\sigma_r^2 - \frac{1}{12})^{1/2}]$.[44] It is convenient to compare Z_P and A'_P values with those for unchanged-charge division, $Z_{\mathrm{UCD}} = A'(\frac{92}{236})$ and $A'_{\mathrm{UCD}} = Z(\frac{236}{92})$; the differences,

$$\Delta Z = (Z_P - Z_{\mathrm{UCD}})_H = (Z_{\mathrm{UCD}} - Z_P)_L$$

and

$$\Delta A'_P = (A'_P - A'_{\mathrm{UCD}})_H = (A'_{\mathrm{UCD}} - A'_P)_L ,$$

are calculated and plotted. For both models the complementarity relationship, $A'_H + A'_L = A_{\mathrm{fis}}$, is used. The ΔZ and σ_Z functions for the Z_P model and the $\Delta A'_P$ and $\sigma_{A'}$ functions for the A'_P model are derived from data for both light and heavy products by the method of least squares for individual mass or atomic numbers and also globally for all mass or atomic numbers treated together. Complementary element yields, Y_{Z_L} and Y_{Z_H}, are required to be equal for the A'_P model ($Z_L + Z_H = Z_{\mathrm{fis}}$).

A. The Z_P model

Figure 2 shows the results of calculations with the Z_P model; the points are for individual mass numbers, and the solid lines represent simple straight-line functions with parameters derived from the global treatment. For $A'_H \geq 130.2$, the lines are for functions with parameters derived previously $[\Delta Z(A'=140) = -0.47$, $\partial\Delta Z/\partial A' = -0.010$, $\overline{\sigma_Z} = 0.52$, $\overline{F_Z} = 1.27$, $\overline{F_N} = 1.08]$.[1] Below

TABLE II. Experimental data not used for determination of model parameters.

Fission product	Experimental FI(FC)	Reference	Calculated FI(FC) Z_P model	A'_P model
^{102}Tc	0.005 ±0.003	15	6.1×10^{-4}	1.1×10^{-4}
^{103}Mo	(1.007 ±0.014)[a]	38	(0.9926)	(0.9994)
^{103}Tc	0.026 ±0.009	15		
	0.030 ±0.007	27	7.4×10^{-3}	5.9×10^{-4}
	0.025 ±0.012	28		
^{104}Tc	0.032 ±0.012	15		
	0.060 ±0.010	27	2.5×10^{-3}	3.1×10^{-3}
	0.030 ±0.023	14		
	[0.0018±0.0018][b]			
^{105}Zr	0.043 ±0.0060	27	7.3×10^{-6}	2.0×10^{-3}
^{105}Mo	(0.983 ±0.016)[a]	38	(0.983)	(0.985)
^{105}Tc	0.053 ±0.017	15		
	0.060 ±0.015	27	0.017	0.015
	0.033 ±0.023	14		
	[0.003 ±0.007][b]			
	(0.76 ±0.04)	39	(>0.9999)	(>0.9999)
^{106}Nb	0.100 ±0.017	27	0.036	0.058
	[0.039 ±0.017][b]			
^{106}Mo	0.695 ±0.043	27	0.888	0.869
	[0.907 ±0.037][b]			
	(1.065 +0.095 −0.087)[a]	38	(0.924)	(0.928)
^{106}Tc	0.200 ±0.040	27	0.076	0.072
	[0.055 ±0.008][b]			
^{106}Ru	0.004 ±0.045	27	1.6×10^{-6}	2.7×10^{-6}
^{107}Tc	0.145 ±0.026[c]	14	0.23	0.30
	[0.183 ±0.040][b]			
	(0.33 ±0.05)	39	(>0.9999)	(0.9998)
^{115}Pd	(0.89 ±0.01)	40	(0.984)	(0.960)
	(0.905 ±0.072)	41		
^{128}I	(4.0 ±0.9)10^{-5}	42	1.6×10^{-15}	6.4×10^{-8}

[a]Not used because only Tc present as Tc(VII) was separated from Mo for yield measurements (Ref. 21).
[b]Average FI (from Table I) calculated without the data listed immediately above.
[c]Superseded by a later value (Ref. 15) that was used.

$A'_H=130.2$, the ΔZ function was assumed to intersect the $Z_P=50$ line at $\Delta Z=0.0$ ($A'_H=128.3$), and three parameters were determined: $\Delta Z_{\max}$, $\Delta A'_Z$, and $\overline{\sigma_{50}}$. $\Delta Z_{\max}$ is the maximum value of ΔZ in the region near symmetry. $\Delta A'_Z$ [previously called BREAK (Ref. 2)] is the horizontal displacement of the steeply rising ΔZ function from the $Z=50$ line measured from the point of intersection ($A'_H=129.2$, $\Delta Z=-0.36$) of the $Z=50$ line and the extrapolation of the nearly flat ΔZ line from the region above $A'_H=130.2$. $\overline{\sigma_{50}}$ is the average value of the width parameter, $\overline{\sigma_Z}$, in the region near $Z_P=50$; this region was assumed to cover the A'_H range in which the ΔZ function increases sharply. It was also assumed that the ΔZ function decreases linearly from $\Delta Z_{\max}$ to 0.0 at symmetry ($A'_H=118$). The parameters determined are given in Table III.

The value of $\overline{\sigma_Z}$ near symmetry is the same, although less precisely determined, as the value of 0.52 ± 0.02 for

TABLE III. Z_P model parameters determined globally.

Parameter	Value determined	A'_H
ΔZ_{max}	0.70±0.10	124.5
$\Delta A'_Z$	1.00±0.20	129.2—130.2
$\overline{\sigma_{50}}$	0.31±0.02	124.5—130.2
$\overline{\sigma_Z}$	0.52±0.08	<124.5
$\overline{F_Z}$ [a]	0.96±0.10	<124.5
	0.98±0.04	124.5—130.2
$\overline{F_N}$ [a]	1.01±0.11	<124.5
	1.03±0.03	124.5—130.2

[a]Values of $\overline{F_Z}=1.00$ and $\overline{F_N}=1.00$ were used for calculation of other parameter values and for lines in Fig. 2.

asymmetric mass division.[1] The results from most calculations are for $\overline{\sigma_Z}=0.52$.

Calculations that included the average even-odd-proton or -neutron factors gave values close to unity, so these factors were taken to be 1.00 for A' between 105.8 and 130.2 ($104 \leq A \leq 129$).

B. The A'_P model

Figures 3 and 4 show the results of calculations with the A'_P model for $Z=50$ and 42 and for $Z=49$ and 43, respectively; the points are for individual mass numbers, and the lines represent Gaussian fits to the data. The parameters determined are listed in Table IV.

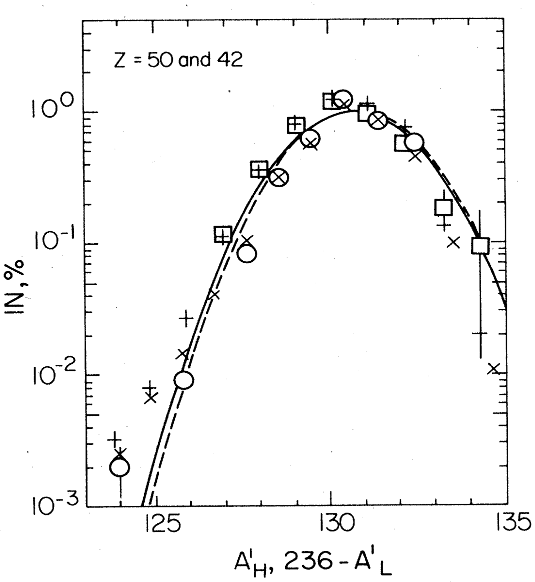

FIG. 3. Independent yields, IN, of nuclides with $Z=50$ (Sn) and 42 (Mo) vs the average heavy-fragment mass number, A'_H. (○), Sn and (□), Mo yields from Table I; (×), Sn and (+), Mo yields calculated from the Z_P model (Ref. 45); (——), Gaussian function derived by the element-pair method; (— — —), Gaussian function derived by the global method. Parameters for the functions are from Table IV.

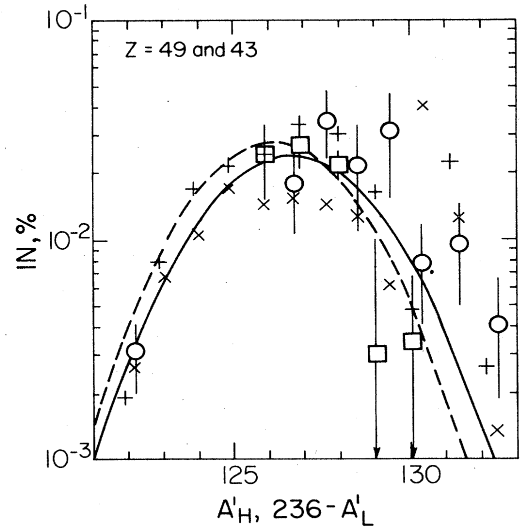

FIG. 4. Independent yields, IN, of nuclides with $Z=49$ (In) and 43 (Tc) vs the average heavy-fragment mass number, A'_H. (○), In and (□), Tc yields from Table I; (×), In and (+) Tc yields calculated from the Z_P model (Ref. 45); (——), Gaussian function derived by the element-pair method; (— — —), Gaussian function derived by the global method. Parameters for the functions are from Table IV.

There were too few data to treat elements with Z values from 44 through 48 as described above, so the six data available were treated together. A linear $\Delta A'_P$ function was assumed, which equaled 0.0 at $Z=46$ (symmetry). Y_Z values were estimated from the average mass-number yield (Y_A) in the valley: $(0.011\%)(\frac{236}{92})=0.028\%$. Results of the calculations are summarized in Table IV and in Fig. 5. The derived linear $\Delta A'_P$ function for $46 \leq Z_H \leq 48$ is shown in Fig. 6.

Calculations that included determination of $\overline{F_N}$ for all three groups of elements gave values close to unity, as shown in Table IV. Therefore, a value of $\overline{F_N}=1.00$ was used for most calculations.

The global treatment for the A'_P model[1,3] was designed to treat data for all Z's together, and the data set can include both IN values and experimental Y_A values, which are the sums of all IN values for each A. The experimental Y_A values were taken from Rider's compilation;[12] only values with errors <20% of the value were used because many of the other values were interpolations.[46] A minimum error of 5% of a value was assigned so that Y_A values with relatively small errors were not weighted orders of magnitude more than IN values, which are more directly related to nuclear-charge distribution.

The results of the global A'_P calculations are given in brackets in Table IV. The parameter values from the global treatment are similar to those determined for element pairs or groups because parameters for the same functions were determined in both treatments for the Z range from 42 through 50. The differences are due to inclusion of Y_A values in the data set for the global treatment.

TABLE IV. A_P' model parameters. Unbracketed values are from treatment of data for element pairs or groups. Values in brackets are from the global treatment.[a]

Parameter	Values determined for $Z_H=$ 50	49	46–48
Y_Z,%	4.07±0.45	0.14±0.02	(0.028)[b]
	[4.13±0.35]	[0.15±0.03]	[0.027±0.006][c]
$\Delta A_P'$	2.45±0.20	0.95±0.35	−1.10 ±0.35[d]
	[2.54±0.15]	[0.40±0.50]	[−1.20 ±0.80][d]
$\sigma_{A'}$	1.63±0.13	2.23±0.25	1.22 ±0.15
	[1.59±0.09]	[2.08±0.40]	[1.38 ±0.01][e]
$\overline{F_N}$[f]	1.01±0.09	1.01±0.15	0.88 ±0.08
	[1.00][b]	[1.00][b]	[1.00][b]

[a]Equations and parameters used for $Z_H > 50$ are given below:

$Y(51 \le Z_H \le 59)$ were determined individually .

$$Y(Z_H > 59) = 50(F_Z)[\mathrm{erf}(VY) - \mathrm{erf}(WY)] ,$$

$$VY = \frac{54.2 - Z_H + 0.5}{1.88\sqrt{2}}, \quad WY = \frac{54.2 - Z_H - 0.5}{1.88\sqrt{2}} ,$$

$F_Z = 1.27$ for even Z; $F_Z = 1/1.27$ for odd Z ,

$$\Delta A_P'(Z_H > 50) = 1.163 + 0.529\{\ln[Y(Z_H+1)] - \ln[Y(Z_H-1)]\} ,$$

$\overline{\sigma_{A'}} = 1.38$, $\overline{F_N} = 1.065$

[b]Assumed value, see the text.

[c]Average Y_Z for $44 \le Z \le 48$; the values determined globally are the following: 0.029±0.010 %, 0.025±0.012 %, and 0.029±0.011 % for $Z=46$, 45 and 47, and 44 and 48, respectively.

[d]For $Z_H = 48$.

[e]$\overline{\sigma_{A'}}$ determined for all Z's except 42, 43, 49, and 50.

[f]$\overline{F_N} = 1.065 \pm 0.022$ for all Z's except 42–50.

IV. DISCUSSION

The FI values calculated from the global Z_P and A_P' models are listed in the last two columns of Table I for comparison with the experimental FI values. On the whole, agreement is satisfactory; most calculated FI values are within two standard deviations of experimental values. Only the Z_P-model FI value for ^{130}In disagrees with experiment by more than five times the experimental error. Reduced chi-square for Z_P-model values is 2.05, and it is 1.72 for A_P' model values; the difference is due mainly to the ^{130}In discrepancy, which will be discussed.

The functions and parameters derived from the Z_P and A_P' empirical models are compared to the data, to each other,[45] and where possible, to the scission-point theoretical model in Figs. 2–8.

It can be seen from Fig. 2 that the assumed straight-line ΔZ and σ_Z functions (solid lines) represent values derived for individual mass numbers (points) reasonably well. Also, corresponding functions, $\Delta \overline{Z}$ and $\sigma_{\overline{Z}}$, derived from the A_P' model ($\times$ and $+$ symbols) show similar trends. ($\Delta \overline{Z}$ and $\sigma_{\overline{Z}}$ are defined in Ref. 45.) There is a sharp rise in ΔZ and in $\Delta \overline{Z}$ below $A_H' = 130$ to positive values, and they remain positive to symmetry ($A' = 118$). The σ_Z and $\sigma_{\overline{Z}}$ values are significantly smaller in the A' range from ~124 through ~130 than they are above and below this range. The ΔZ function predicted by the scission-point theory[7] (dashed line in Fig. 2) is much lower than the data

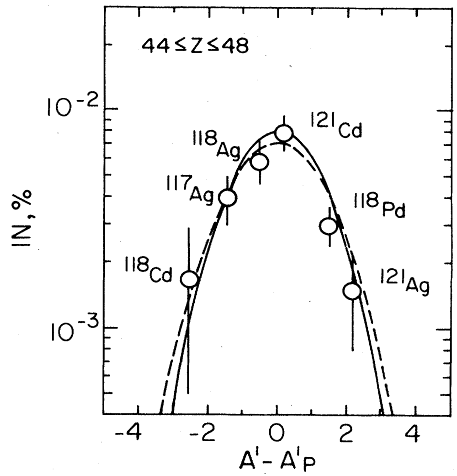

FIG. 5. Independent yields, IN, of nuclides with $Z=46$ (Pd), 47 (Ag), and 48 (Cd) vs $A' - A_P'$, A_P' being calculated from the $\Delta A_P'$ line in Fig. 6. (○), yields from Table I; (——), Gaussian function with $Y_Z = 0.027\%$ and $\sigma_{A'} = 1.22$; (— — —), Gaussian function with $Y_Z = 0.027\%$ and $\sigma_{A'} = 1.38$.

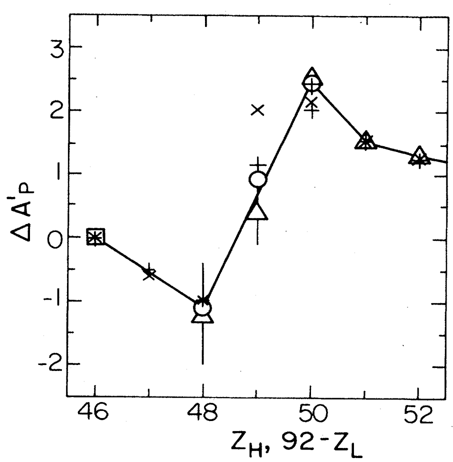

FIG. 6. $\Delta A_P'$ as a function of Z_H. (○), $\Delta A_P'$ derived by the element-pair method; (△), $\Delta A_P'$ derived by the global method; (×), $\Delta A_H'$ and (+), $\Delta A_L'$ calculated from the global Z_P model (Ref. 45); (□), $\Delta A_P' = 0.0$ (assumed); (——), straight-line function: $\Delta A_P' = 0.0$ at $Z=46$, $\Delta A_P' = -1.1$ at $Z=48$, and $\Delta A_P' = 2.5$ at $Z=50$. The values for points at $Z_H = 51$ and 52, shown for continuity, were derived by the global method from the current data file (Ref. 24).

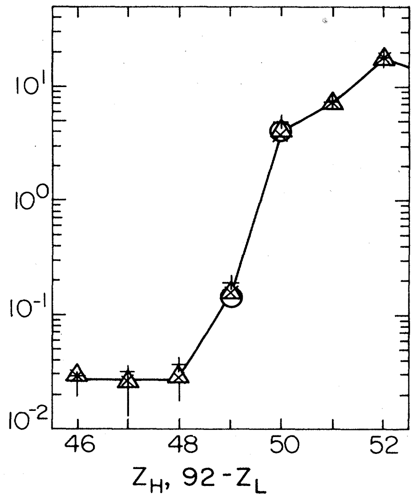

FIG. 7. Element yield, Y_Z, as a function of Z_H. (○), Y_Z derived by the element-pair method; (△), Y_Z derived by the global method; (×), Y_{Z_H} and (+), Y_{Z_L} calculated from the global Z_P model (Ref. 45); (——), straight-line function: $Y_Z=0.027\%$ for $Z_H=46$ to 48, $Y_Z=0.145\%$ for $Z_H=49$, and $Y_Z=4.10\%$ for $Z_H=50$. The values for points at $Z_H=51$ and 52, shown for continuity, were derived by the global method from the current data file (Ref. 24).

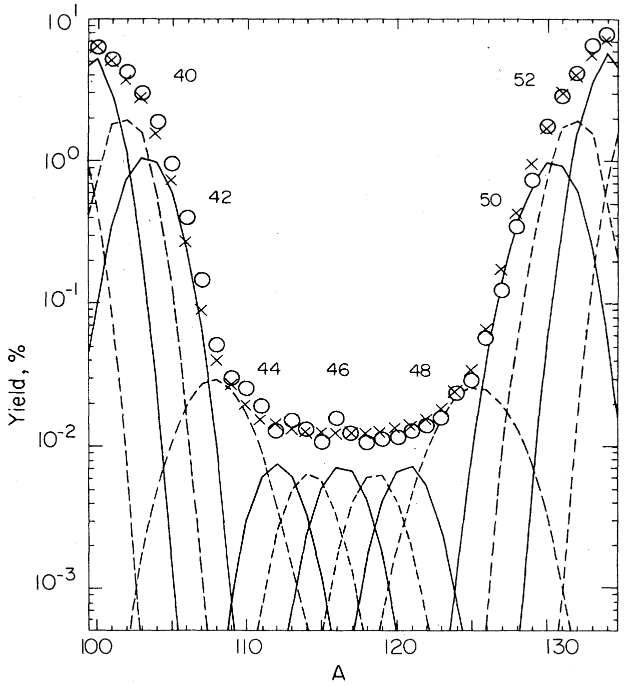

FIG. 8. Mass-number yields, Y_A, as a function of A. (○), experimental yields (Ref. 12); (×), calculated yields from the global A'_P model (Ref. 45); (——), IN for even Z elements (Z is given); (— — —), IN for odd Z elements. IN values are calculated from the global A'_P model.

points and the empirical ΔZ and $\Delta \overline{Z}$ functions, as pointed out previously.[1,19]

The sharp breaks in the ΔZ and $\overline{\sigma_Z}$ functions, shown as solid lines in Fig. 2, result from the simple straight-line functions assumed to illustrate general trends, and the sharp breaks contribute to some discrepancies between experimental and calculated values. When the functions were smoothed to the locations indicated by dotted lines in Fig. 2, the reduced-chi-square value was lowered from 2.05 to 1.84, and the ^{130}In discrepancy were reduced from 9 to 4 times the experimental error. However, smoothing caused the differences between experimental and calculated values for ^{104}Nb and ^{104}Tc to be increased from ~ 1 to 4 times the experimental error.

There is a spread on the A' axis of ~ 0.5 units between Mo and Sn independent yields plotted in Fig. 3, an indication that the $\overline{\nu_p}$ values used might be too large. The Gaussian functions representing the data are essentially the same whether derived by the element-pair or global treatments, and the representation of data is satisfactory considering the Mo-Sn data spread. The values calculated from the Z_P model (× and + symbols) show the same general characteristics as the A'_P functions and represent individual data points better, reproducing approximately the spread between the Mo and Sn data. However, this better agreement results from the larger calculated yield for Mo (4.6%) than for Sn (3.7%), a violation of charge conservation. Correction to the average Y_Z of 4.15%, essentially the value derived from the A'_P model, moves Mo points (+) down and Sn points (×) up toward the calculated Gaussian curves.

There is considerable scatter among the Tc and In independent yields plotted in Fig. 4, and the dispersion in A' is larger than for other element pairs, as shown by the $\sigma_{A'}$ values in Table IV. The scatter, and possibly some of the dispersion, could be due to the use of data from a variety of sources. For example, the higher data points—for ^{127}In, ^{129}In, ^{131}In, and ^{132}In—all include yields measured mass spectrometrically,[20] and the last three points would be higher if there had been no normalization (Table I, footnote e).

However, much of the large dispersion in A' for Tc and In yields is probably real for the following reasons. The nuclear-charge dispersion for $A=121$ is normal and is consistent with other data for near-symmetric mass division, so there is no reason to suspect the ^{121}In yield. Although the yields[20] for ^{129}In, ^{130}In, ^{131}In, and ^{132}In are higher than the calculated curve and the very small experimental yields determined[21,22] for ^{104}Tc and ^{105}Tc, which are approximately complementary to the indium isotopes, there is independent evidence that ^{129}In, ^{130}In, ^{131}In, and ^{132}In are formed in fission in small but appreciable yield. Delayed-neutron activities[47,48] and/or gamma-ray measurements[49–51] have been reported for these nuclides formed from thermal-neutron-induced fission of ^{235}U. Therefore, it is probable that the mass dispersion for Tc and In is quite broad covering the product mass-number range from $A_H=121$ through $A_H=132$ ($122 \le A'_H \le 133$).

The representation of the data by a Gaussian curve (line in Fig. 4) is reasonably satisfactory considering the data scatter. The dashed curve from the global treatment is somewhat less satisfactory being shifted to the left (smaller $\Delta A_P'$, Table IV) because of inclusion of mass-yield data. The IN values derived from the Z_P model ($\times$ and $+$ symbols) represent the data moderately well except for the large calculated values near $A_H'=130$. The Z_P model calculated IN value for ^{130}In is five times larger than the normalized experimental value plotted or 3.5 times larger than the reported value.[20] The Z_P-model calculation generally gives larger Tc yields than In yields, except near $A_H'=130$.

The large dispersion of the Tc and In yields for products with A_H from 121 through 132, the mass-number range where mass yields increase by more than two orders of magnitude, is probably associated with this large yield change. Nuclides with the lower A_H, higher A_L, are near symmetry, and their independent yields contribute significantly to the small yields for these mass numbers. The higher A_H, lower A_L, nuclides are in the asymmetric range, where In and Tc independent yields make only very small contributions to the much higher yields for these mass numbers.

Figure 5 shows the IN data for Pd, Ag, and Cd fission products with the Gaussian curves derived from the data. It can be seen that both curves, one with $\sigma_{A'}=1.22$ and one with $\sigma_{A'}=1.38$ (the average for most Z's), represent the data satisfactorily. The A_P' values were derived from the $\Delta A_P'$ function shown as a line for $46 \leq Z_H \leq 48$ in Fig. 6.

Derived $\Delta A_P'$ values are plotted in Fig. 6. The maximum $\Delta A_P'$ value of 2.5 at $Z=50$ has been well established,[1,3,19] and evidence for a sharp drop in the function below $Z_H=50$ has been presented.[19] However, the nature of the function has been unknown between $Z_H=49$ and $Z=46$ (symmetry, where $\Delta A_P'$ is expected to be zero). Although uncertainties are large, negative values of $\Delta A_P'$ for $Z=48$ result from both the element-group and global treatments. Also, Z_P-model calculations give negative values for $\Delta A'$ at $Z=48$ and 47 consistent with the $\Delta A_P'$ function. The dip in $\Delta A_P'$ to negative values is related to the positive ΔZ values (Fig. 2), but the relationship is not simple because of variation of Y_A with A. The straight line from $Z_H=48$ to $Z=50$ fits the derived $\Delta A_P'$ values for $Z_H=49$ within the uncertainties given in Table IV. The value of the $\Delta A_P'$ line at $Z_H=49$ is 0.7, the same as the value derived from radiochemical data.[19]

Figure 7 shows a plot of element yields vs Z_H. It can be seen that yields for valley elements, Z_H from 46 through 48, are small and nearly constant, as was assumed for the element-group calculations. There is a sharp increase in element yield at $Z_H=49$ and $Z_H=50$, as observed previously.[1,3,19] The line connecting average Y_Z values is steepest between $Z_H=49$ and $Z_H=50$. Element yields derived from Z_P-model calculations show the same trends, but yields of complementary light and heavy elements are not equal, a violation of charge conservation.

Mass-number yields derived from the A_P' model are compared to experimental yields[12] near symmetry in Fig. 8. The Y_A function (shown as $\times$) derived from the global treatment represents the general trends in the data (shown as $\circ$) quite well, but it does not accurately reproduce abrupt changes in yields, such as those that occur in the steep rises from the valley to the peaks and near $A=116$.

The independent yields of isotopes of elements near symmetry calculated by the global A_P' method are shown as lines in Fig. 8. Because the calculated yields are plotted against A, not A', the curves are not Gaussian but consist of straight-line segments connecting IN values. As illustrated previously,[1,3] the initial small rise in Y_A in going from the valley to the peaks is due to increasing yields of nuclides with Z values of 49 and 43, and the steepest rise in Y_A is due to increasing yields of nuclides with Z values of 50 and 42. Calculations with either the Z_P or A_P' model give FI values between 0.6 and 0.9 for Sn isotopes with A from 126 through 129, and similar values are obtained for Mo isotopes with A from 104 through 107. Experimental FI values, where known, are also between 0.6 and 0.9 in these mass-number ranges.

V. CONCLUSIONS

This paper presents evidence that there is a strong preference for formation of nuclides with Z values of 50 and 42 in the complementary mass-number ranges $126 < A_H' < 131$ and $105 < A_L' < 110$. This preference can be associated with the stability of the 50-proton shell, which influences yields in the mass-number region between symmetry and the mass-yield peaks, where stabilities of the 82-neutron shell and distorted shells in the light fragment are important.[7,52] The evidence consists of the steep rise in ΔZ below $A_H'=130$, the small value of $\overline{\sigma_{50}}$ (Fig. 2), the large slope of the Y_Z vs Z function between $Z=49$ and $Z=50$, and the large Y_Z value for $Z=50$ and 42 compared to those for intermediate Z's (Fig. 7).

The strong preference for formation of nuclides with $Z=50$ and 42 is associated with $Z=50$, not $Z=42$, because the light-element Z's at which abrupt Y_Z changes occur depend on the Z of the fissioning nucleus and are complementary to $Z_H=50$ and 49.[1] Evidence to support this interpretation is afforded by measured independent and element yields for other fission reactions. For thermal-neutron-induced fission of ^{239}Pu, independent yields of Tc isotopes are large,[21,22,53] and the abrupt drop in Y_{Z_L} occurs between $Z=44$ and $Z=45$.[53,54] For spontaneous fission of ^{252}Cf, the abrupt drop in Y_{Z_L} occurs between $Z=48$ and $Z=49$.[54] Also, the similarity in position on the A axis of the light side of the heavy mass-yield peak for many fission reactions suggest that $Z=50$ plays an important role in determining the mass numbers at which asymmetric fission becomes probable.[55] The A_P' model has been used with mass-yield data[12] to interpret this concept more quantitatively.[1]

The discrepancy between the empirical and theoretical ΔZ functions shown in Fig. 2 could be due[23] to the use of a small distance (1.4 fm) between nascent fragments in the scission-point theory.[7] A larger distance would raise the theoretical ΔZ function because the nascent fragments would be less distorted, allowing the stable spherical 50-proton shell to influence the charge distribution more strongly[23,52] than was assumed.[7]

Explanation of the observed lack of even-odd-proton and -neutron effects requires consideration of two charge- and mass-number regions. In the mass-number region where Z_{P_H} is near 50, there is actually a very large even-odd-proton effect due to the strong preference for formation of nuclides with $Z=50$ and 42. If σ_Z is held constant at 0.52, the average value for other mass numbers,[1] a least-squares calculation gives a value of $\overline{F_Z}=1.5$ for the $124.5 \leq A'_H < 130.2$ region, a value considerably larger than $\overline{F_Z}=1.27$, the average value for most mass numbers.[1] Thus, use of $\overline{F_Z}=1.5$ and $\sigma_{50}=0.52$ is another way to represent the 50-proton-shell effect in this mass-number range, but use of the Z_P-model parameters, $\sigma_{50}=0.31$ and $\overline{F_Z}=1.00$, gives a better representation of the data.

It has been observed that excitation of ^{236}U from the interaction of ^{235}U with neutrons of several MeV greatly reduces the average even-odd-proton effect,[2,56] and it is reasonable to assume that the inverse is true. Therefore, the lack of even-odd-proton and -neutron effects in the $118.0 < A'_H < 124.5$ region (Table III) and for Z_H from 46 through 48 (Table IV) indicates that excitation energies are higher for ^{236}U nuclei undergoing symmetric fission than for those undergoing asymmetric fission.

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[6]$\Delta Z=(Z_P-Z_{\rm UCD})_H=(Z_{\rm UCD}-Z_P)_L$, $Z_{\rm UCD}$ being the Z for unchanged-charge distribution $[Z_{\rm UCD}=A'(Z_{\rm fis}/A_{\rm fis})]$. The subscripts H, L, and fis refer to heavy and light products and to the fissioning nuclide, respectively, and A' is the average precursor-fragment mass number $[A'=A+\overline{\nu_p}(A)]$. The $\overline{\nu_p}$ function, the average number of neutrons emitted to form fission products for each A vs A, is shown and described in Ref. 19.

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[25]FC values were converted to FI values by applying the corrections (C_F) listed below. For FC<0.5, FI(Z)=FC(Z)$-C_F$; for FC$\geq$0.5, FI($Z+1$)=1.0$-$FC(Z)$-C_F$. C_F was determined from a Gaussian charge-dispersion curve with $\sigma_Z=0.55$; the error in C_F, which is twice C_F, covers possible variation of σ_Z values up to 0.75 (Ref. 2). If $F=\{$FC(Z) or 1.0$-$FC(Z)$\}$ is <0.01, $F_C=0.0\pm0.1F$; if $F=0.01$ to 0.1, $C_F=0.001\pm0.002$; if $F=0.1$ to 0.2, $C_F=0.003\pm0.006$; if $F=0.2$ to 0.3, $F_C=0.007\pm0.014$; if $F=0.3$ to 0.4, $F_C=0.015\pm0.030$; if $F=0.4$ to 0.5, $F_C=0.025\pm0.050$.

[26]Although many FI values from measurements with the LOHENGRIN and HIAWATHA fission-product-recoil separators are in agreement with FI values from radiochemical and mass-spectrometric measurements, very small values are not (Ref. 3). Therefore, some of the yields were assigned evaluator's errors, which are larger than reported experimental errors. The larger of ±0.02 or 5% of a value was assigned to FI values from measurements which allowed summation over most kinetic energies and ionic-charge states (Refs. 15 and 27). Evaluator's errors of ±0.03 or 7% of a value were assigned to FI values from measurements only at or near average kinetic energies and ionic-charge states (Refs. 14 and 28).

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$$\Delta\overline{Z}=(\overline{Z}-Z_{\mathrm{UCD}})_H=(Z_{\mathrm{UCD}}-\overline{Z})_L\,,\quad \overline{Z}=\sum(Z)(\mathrm{FI})\,,$$

$$\sigma_{\overline{Z}}=(\sigma_r^2-\tfrac{1}{12})^{1/2},\quad \sigma_r^2=\sum(Z-\overline{Z})^2(\mathrm{FI})\,.$$

If the dispersion of derived FI values for each A were Gaussian, $\Delta\overline{Z}$ and $\sigma_{\overline{Z}}$ would be equivalent to the Z_P-model ΔZ and σ_Z parameters. Values of $\mathrm{IN}=(\mathrm{FI})(Y_A)$ derived from the Z_P model, FI being calculated and Y_A being experimental (Ref. 12), can be used to determine $\Delta\overline{A'}$, $\sigma_{\overline{A'}}$, and Y_Z:

$$\Delta\overline{A'}=(\overline{A'}-A'_{\mathrm{UCD}})_H=(A'_{\mathrm{UCD}}-\overline{A'})_L\,,$$

$$\overline{A'}=(1/Y_Z)\sum(A')(\mathrm{IN}),\quad Y_Z=\sum(\mathrm{IN})\,,$$

$$\sigma_{\overline{A'}}=(\sigma_r^2-\tfrac{1}{12})^{1/2}\,,$$

$$\sigma_r^2=(1/Y_Z)\sum(A'-\overline{A'})^2(\mathrm{IN})\,.$$

If the dispersion of derived IN values were Gaussian, $\Delta\overline{A'}$ and $\sigma_{\overline{A'}}$ would be equivalent to the A'_P-model $\Delta A'_P$ and $\sigma_{A'}$ parameters.

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