

Isotopic and Isotonic Yields in Nuclear Fission*

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The curves for isotopic and isotonic fission yields are calculated and shown to be the Weierstrass transforms of the isobaric yield curve. The well-known mass asymmetry of the fission is associated with asymmetry in charge and in neutron contents. There are "forbidden" zones at $44 < Z < 48$ and at $64 < N < 78$, where formation probabilities are extremely low. The distribution of the number of neutrons of the fission fragments is much narrower than what would be expected on the basis of the variation ranges for Z and N . The ambiguity existing about whether the fragments of a pair are meant to complement before or after prompt neutron boil-off is removed, and Glendenin's treatment is confirmed. There are indications which are in full agreement with the properties of Wahl's empirical Z_p function that the proton closed shell contribution (if any) to the fine structure observed in the isobaric yield curve may be larger than the contribution of the neutron closed shell. The isotopic yields of the known nuclides are significantly smaller than the yields defined by the full theoretical curves for $Z=38, 39, 40, 41, 52$, and 57 , indicating that new isotopes of Sr, Y, Zr, Nb, Te, and La are still to be identified as primary fission products from the thermal neutron fission of U^{235} .

INTRODUCTION

ANY hypothesis on charge distribution in nuclear fission defines two quantities: (1) The most probable atomic number (not necessarily an integer) Z_p for the product which has the highest yield among all products of a given mass chain A ; and (2) the fractional chain yield η for a nuclide defined as the yield of this nuclide divided by the total yield of the chain to which this nuclide belongs. The curve $\eta(Z, A)$ is called the charge-dispersion curve. This information, along with the experimental yield-mass curve $y(A)$, permits one to calculate the isotopic and isotonic fission yield curves, $\bar{y}(Z)$ and $\bar{y}(N)$, as follows:

$$\begin{aligned}\bar{y}(Z) &= \sum_A y(A) \cdot \eta(Z, A), \\ \bar{y}(N) &= \sum_A y(A) \cdot \eta(N, A),\end{aligned}\quad (1)$$

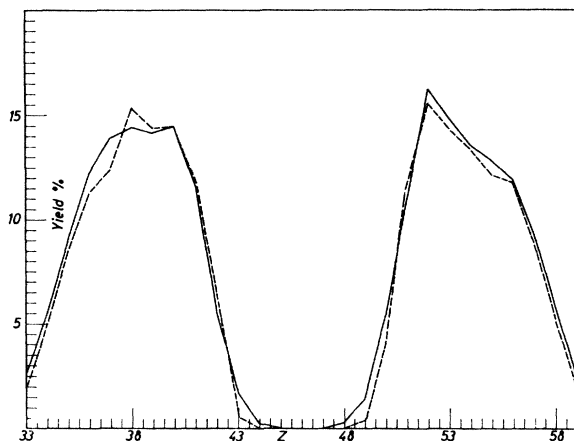


FIG. 1. Fission yields of isotopic fragments from $U^{235}(n_{th}, F)$, calculated by using the postulate of equal charge displacement (continuous line) and Wahl's empirical Z_p function (broken line).

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where

$$\begin{aligned}\eta(Z, A) &= f(Z - Z_p), & Z_p &= \varphi(A), \\ \eta(N, A) &= f(N - N_p), & N_p + Z_p &= A.\end{aligned}\quad (2)$$

The mathematical operation defined by the foregoing formulas is essentially a finite linear integral transformation of the function $y(A)$ into $\bar{y}(Z)$ or $\bar{y}(N)$, the kernel of the transform being $\eta(Z, A)$ or $\eta(N, A)$, respectively.

Recent work^{1,2} established that the charge-dispersion curve is the Gaussian:

$$\eta(Z, A) = (\pi C)^{-1/2} \exp[-(Z - Z_p)^2/C], \quad (3)$$

with $C=0.9$.² Therefore, by treating the mass number A as a continuous variable, one can write:

$$\bar{y}(Z) = \frac{1}{(\pi C)^{1/2}} \int_0^\infty y(A) \exp[-(Z - Z_p)^2/C] dA, \quad (4)$$

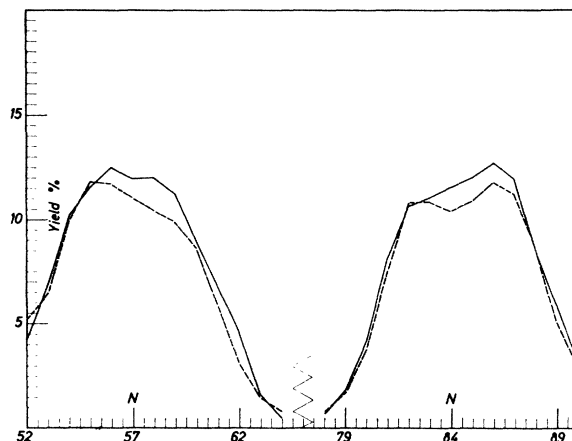


FIG. 2. Fission yields of isotonic fragments from $U^{235}(n_{th}, F)$, calculated by using the postulate of equal charge displacement (continuous line), and Wahl's empirical Z_p function (broken line).

¹ I. F. Croall, J. Inorg. Nucl. Chem. **16**, 358 (1961).

² A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1962).

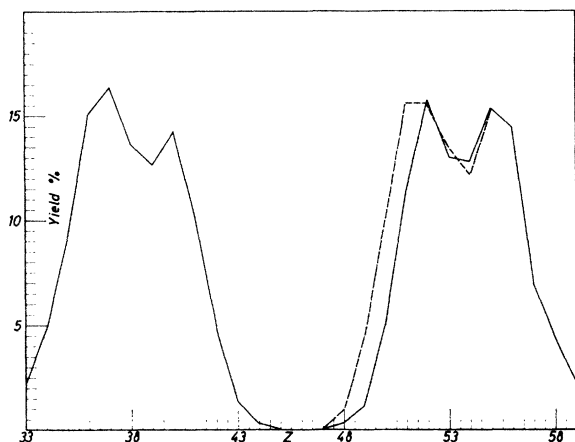


FIG. 3. Fission yields of isotopic fragments from $U^{235}(n_{th}, F)$, calculated by using Pappas' (continuous line), and Kennett and Thode's (broken line) discontinuous Z_p functions.

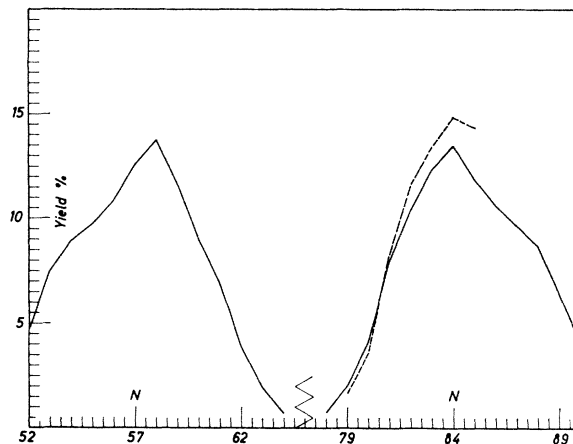


FIG. 4. Fission yields of isotonic fragments from $U^{235}(n_{th}, F)$ calculated by using Pappas' (continuous line), and Kennett and Thode's (broken line) discontinuous Z_p functions.

and

$$\bar{y}(N) = \frac{1}{(\pi C)^{1/2}} \int_0^{\infty} y(A) \exp[-(N - N_p)^2/C] dA. \quad (5)$$

The integral transformation defined by (4) and (5), whose kernel is the normalized Gaussian (3), is the Weierstrass transform. Therefore, the isotopic and isotonic fission yield curves are the Weierstrass transforms of the isobaric yield curve.

The integral forms given above are useful when an analytical expression for $y(A)$ is available. Fong's³ statistical theory of nuclear fission provides such an expression, but it is too complicated. However, since $y(A)$ is discontinuous and practically negligible below $A=80$ and above $A=160$, the finite summation formulas (1) and (2) may advantageously be used for calculating the transforms $\bar{y}(Z)$ and $\bar{y}(N)$.

Accordingly, we have calculated the isotopic and isotonic yield curves by making use of various postulates⁴⁻⁶ of charge distribution as well as Wahl's^{2,7} empirical Z_p function, in the case of the thermal neutron fission of U^{235} . Some of the calculated curves are shown in Figs. 1 to 4. The tabulated values for $y(A)$ reported by Steinberg and Glendenin⁸ are used throughout.

RESULTS AND DISCUSSION

The curves thus obtained lead to the following observations:

³ P. Fong, Phys. Rev. **102**, 434 (1956).

⁴ L. E. Glendenin, C. D. Coryell, and R. R. Edwards, in *Radiochemical Studies: The Fission Products*, edited by Coryell and Sugarman, (McGraw-Hill Book Company, Inc., New York, 1951), p. 489.

⁵ A. C. Pappas, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 7, p. 19.

⁶ T. J. Kennett and H. G. Thode, Phys. Rev. **103**, 323 (1956).

⁷ A. C. Wahl, J. Inorg. Nucl. Chem. **6**, 263 (1958).

⁸ E. P. Steinberg and L. E. Glendenin, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 7, p. 3.

(1) Both $\bar{y}(Z)$ and $\bar{y}(N)$ are "double-humped" as is the original $y(A)$ curve, signifying that the asymmetry of fission with respect to mass is associated with asymmetries in nuclear charge and in neutron contents.

(2) The $\bar{y}(Z)$ and $\bar{y}(N)$ curves must satisfy some strict conservation conditions, and, therefore, provide useful tests for the charge-distribution postulate used to relate Z_p to A . These conditions are:

(a) The sum of the abscissa of any two points of equal ordinates situated on two complementary branches of the curves must equal 92 for the $\bar{y}(Z)$ and 141.5 [see statement (9)] for the $\bar{y}(N)$ curve, in the case of the thermal neutron fission of U^{235} . (The light side of the light peak is complementary to the heavy side of the heavy peak, and the heavy side of the light peak is complementary to the light side of the heavy peak).

(b) The area under the $\bar{y}(Z)$ curve, as well as that under the $\bar{y}(N)$ curve must equal the area under the original $y(A)$ curve. This is a requirement of the conservation of the number of fragments.

(3) The characteristic parameters of the $\bar{y}(Z)$ and $\bar{y}(N)$ curves are *additive* with respect to the corresponding parameters of the $y(A)$ curve (see Table I).

(4) The peak widths of $\bar{y}(Z)$ and $\bar{y}(N)$ differ only slightly, whereas the trough width for $\bar{y}(N)$ is more than twice that for $\bar{y}(Z)$ (refer again to Table I).

(5) There are "forbidden" zones on the isotopic and isotonic yield curves at $44 < Z < 48$ and at $64 < N < 78$, respectively, where the formation probabilities are extremely low; the maximum total yields amount to less than 0.5% in these zones. It is interesting to note that Wahl *et al.*² conclude indirectly that $42 < Z < 50$ is a forbidden range; however, they do not specify such a range for neutron contents.

(6) The much larger extension of the forbidden zone for neutron is to be correlated *both* with the wider variation range of the number of neutrons and with the

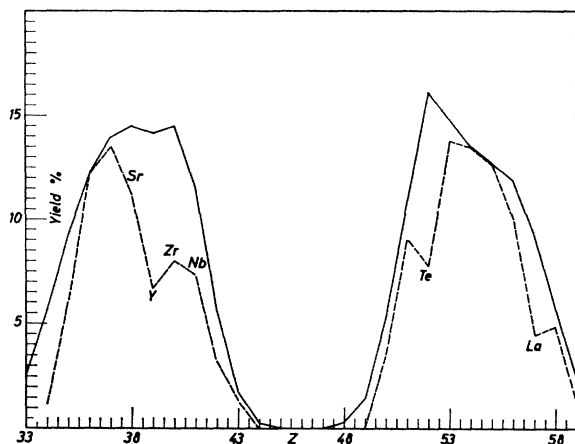


FIG. 5. A comparative plot of the full theoretical isotopic yield curve (continuous line) and the curve calculated by taking into account exclusively the known nuclides (broken line), indicates that new isotopes of Sr, Y, Zr, Nb, Te, and La are still to be identified.

property stated in (4). In fact, the extension of the neutron forbidden zone is too large to be explained as being due exclusively to the trivial fact that the variation range for N is wider than that for Z . If it were so, one would expect the N -forbidden zone to be only $(141.5)/(92)=1.54$ times, and not $(19.5)/(8.5)=2.3$ times wider than the corresponding Z zone. From Table I, the over-all widths at half-height are found to be $(2)(7.2)+(8.5)=22.9$ and $(2)(8.3)+(19.5)=36.1$ for $\bar{y}(Z)$ and $\bar{y}(N)$, respectively, and their ratio $(36.1)/(22.9)=1.58$ is in excellent agreement with 1.54, i.e., with the value to be expected if the effect were simply a matter of variation range. The fact that the over-all width is divided between the trough and the two peaks in this way:

Light group peak	Trough	Heavy group peak
8.3	19.5	8.3

and not in this way:

$(1.58)(7.2)=11.3$ $(1.58)(8.5)=13.4$ $(1.58)(7.2)=11.3$
 signifies that the neutron number distribution of the fission fragments of the light and heavy group is *relatively narrower* than the corresponding proton distribution and, as a result, the N -forbidden zone is (about 45%) larger than what would normally be expected by comparison with the Z -forbidden zone. This is an interesting property of the fission process which, as far as we know, has not been pointed out before.

(7) No one of the charge distribution postulates introduces any isotopic or isotonic yield anomalies at $Z=50$ or at $N=82$. Pappas⁵ discontinuous Z_p produces a pronounced structure on the $\bar{y}(Z)$ curve, but the maxima do not occur at shell closures.

(8) Discontinuities in Z_p produce discontinuities on

TABLE I. Some characteristic parameters of the isotopic, isotonic, and isobaric yield curves, calculated by using Wahl's empirical Z_p function. Note that the parameters for $\bar{y}(Z)$ and $\bar{y}(N)$ are additive with respect to $y(A)$.

Curve	Average ^a Z , N , or A number		Sum	Peak width	Trough width
	Light group	Heavy group		at half-height	at half-depth
$\bar{y}(Z)$	38.15	53.95	92.10	7.2	8.5
$\bar{y}(N)$	56.69	84.78	141.47	8.3	19.5
Sum	94.84	138.73	233.57	15.5	28.0
$y(A)$ ^b	95	139	234	15	28

^a Weighted average.

^b Values taken from reference 8.

the $\bar{y}(Z)$ curve alone; the $\bar{y}(N)$ curve is always smooth, implying that the proton closed-shell contribution (if any⁹) to the fine structure should be larger than the neutron closed shell contribution. This conclusion is in agreement with the fact that Wahl's⁷ empirical Z_p line tends to approach and remain close to the 50-proton shell edge, but there is no pronounced tendency for it to remain close to the 82 (or 50) neutron shell edge.

(9) As Coryell¹⁰ states, there is ambiguity about whether the fission fragments of a pair are meant to complement each other before or after prompt neutron boil-off. Now, this ambiguity seems to be removed. In fact, we found that a difference of 2.5 in the total mass of the complementary fragments causes the $\bar{y}(Z)$ and $\bar{y}(N)$ curves to shift in opposite directions in such a way that, in accordance with Glendenin *et al.*⁴ and not with Pappas,⁵ the masses of a pair must be considered as complementing mutually *after* and not before prompt neutron boil-off; otherwise, the proton and neutron conservation conditions are both clearly violated.

(10) The isotopic yield curve drawn by taking into consideration exclusively the *known* nuclides, neglecting the calculated yields for all those nuclides that are not indicated on the Chart of Nuclides,¹¹ is irregular and indicates that new isotopes of Sr, Y, Zr, Nb, Te, and La are still to be identified as primary products of the thermal neutron fission of U^{235} . (Fig. 5).

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⁹ H. Farrar and R. H. Tomlinson, *Can. J. Phys.* **40**, 943 (1962).

¹⁰ C. D. Coryell, M. Kaplan, and R. D. Fink, *Can. J. Chem.* **39**, 646 (1961).

¹¹ *Chart of Nuclides*, prepared by the Institute of Radiochemistry, Nuclear Research Center, Karlsruhe, Germany. (Literature revised to July 1961.)