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Radiochemical Studies of Neutron-Induced Fission of U^{235} and U^{238} and the Two-Mode Fission Hypothesis*

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It has been suggested that the effect of changing excitation energy on the shape of the fission product mass-yield curve from fission of a single nuclear species is due to the change in the relative amounts of two energy-independent modes of fission, each giving rise to its own characteristic mass-yield curve. It is shown here that this hypothesis predicts a linear relationship between the fission yields of any pair of fission products measured at a set of excitation energies. Linear relationships are also predicted between pairs of fission yields measured relative to the yield of some reference fission product. Fission product yields relative to the fission yield of Mo^{99} were measured for fission of U^{235} and of U^{238} with neutron beams of mean energies ranging from 2 to 10 Mev. The predicted linear relationships were observed in all cases. However results for fission yields from U^{235} with thermal neutrons do not fall on the corresponding observed lines. The two-mode fission hypothesis is a possible explanation for the linear relationships observed but does not explain all of the data.

I. INTRODUCTION

In past years many experiments have been performed in which the mass distributions of fission products have been measured for a number of different nuclides undergoing fission at various excitation energies. No completely satisfactory explanation has been found for the changes that occur in the mass distribution of fission products when the excitation energy is varied in a particular nuclide about to undergo fission. However recourse has frequently been made to the suggestion of Turkevich and Niday[1] that there are two fundamentally different modes by which fission may proceed. Both modes are possible, but one generally predominates at low excitation energies and the other at high energies, the relative proportions of the two modes changing with excitation energy. These modes lead to distinctly different mass distributions of products, the observed mass distribution being the resultant of the contributions of both modes.

The mass distributions attributed to the two modes are generally considered to be the two-humped "asymmetric" and the one-humped "symmetric" mass distributions. There has been some study on the competition between symmetric and asymmetric fission,[2] however these studies have considered principally the gross features of the mass distribution. Recently Ford[3] has shown that if in the two-mode fission hypothesis certain conditions are fulfilled, the mass-distribution curves can be broken down and analyzed in terms of two-dimensional vector subspaces. In Sec. II we show that under the same conditions imposed by Ford, a different method of analysis predicts certain straight-line relationships between fission yields of the different mass chains.

We also present in Sec. IV some new data on relative yields in the fission of U^{235} and U^{238} induced by neutrons in the few-Mev range. We feel that these data are in many respects better suited for the testing of the two-mode fission hypothesis than those previously available. These data have been examined to see how closely their behavior follows that to be expected from the two-mode fission hypothesis and the results of this analysis are presented in Sec. V.

II. DERIVATION OF STRAIGHT-LINE RELATIONSHIPS

The general nature of our postulated two modes of fission may be similar for various nuclides undergoing fission, but it is obvious from available fission-yield data that the details of the mass distributions characteristic of each of the two modes must differ for different nuclear species. Consequently in the development which follows we limit ourselves to consideration of fission occurring in a single nuclear species.

Our basic assumption is, therefore, that we are dealing with a single nuclear species undergoing fission and that there are two and only two modes for the fission process to follow. The two fission modes yield different mass distributions and the observed mass-yield curves are simply combinations of those mass distributions for the two modes in varying proportions. We also make the assumption (as did Ford) that a variation in excitation energy does not cause a change in the mass distributions of either of the two modes, but that the changes in the observed mass-yield curve are brought about by changes in the relative proportions of fissions proceeding by each of the two modes.

We can now proceed in the following manner. Let us refer to the product distribution of one fission mode as type A fission and to that of the other mode as type B fission. We shall consider here only those fission yields representing total cumulative yields for an entire mass chain. Suppose the subscript i refers to some arbitrary mass chain.

Let a_i = fission yield of mass chain i in type A fission, b_i = fission yield of mass chain i in type B fission, y_i = observed fission yield of mass chain i, f_A = fraction of total fissions resulting in type A fission, and $f_B = 1$

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[1] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951).

[2] For a summary of this work see I. Halpern, Ann. Rev. Nuclear Sci. **9**, 245 (1959).

[3] G. P. Ford, Phys. Rev. **118**, 1261 (1960).

$-f_A$ = fraction of total fissions resulting in type B fission. We can then write for the observed fission yield of i,

$$y_i=f_Aa_i+f_Bb_i=f_Aa_i+(1-f_A)b_i,$$
$$y_i=f_A(a_i-b_i)+b_i, \tag{1}$$

hence

$$f_A=(y_i-b_i)/(a_i-b_i).$$

Since the choice of mass chain i was arbitrary we can also write for any other mass chain j,

$$f_A=(y_j-b_j)/(a_j-b_j).$$

Therefore

$$(y_i-b_i)/(a_i-b_i)=(y_j-b_j)/(a_j-b_j).$$

Since a_i, b_i, a_j, and b_j are all independent of energy, the above expression represents a linear relationship between y_i and y_j that can be written

$$y_i=c_{ij}y_j+d_{ij}, \tag{2}$$

where c_{ij} and d_{ij} are independent of energy.

The linear relationships just derived hold for the absolute fission yields of mass chains. In attempting to determine the nature of the mass distribution resulting from fission, one frequently measures fission yields of several mass chains relative to the fission yield of some one standard mass chain. It is generally possible to measure such relative fission yields with greater ease and with greater precision than the corresponding absolute fission yields. Let us investigate therefore, what type of relationship is to be expected between the relative fission yields of different mass chains.

Let y_0 be the observed absolute fission yield of some mass chain chosen as an arbitrary "standard," relative to which all other fission yields will be measured. If y_i is the observed (absolute) fission yield of mass chain i, we may modify Eq. (2) by dropping the second subscripts to c and d when y_0 is used as the reference yield, and write

$$y_i=c_iy_0+d_i.$$

Rearranging we get

$$y_i/y_0=c_i+(d_i/y_0),$$

and finally

$$[(y_i/y_0)-c_i]/d_i=1/y_0. \tag{3}$$

Since i refers to any mass chain (except the one chosen as a standard) we may write a similar equation for some other mass chain j. Thus

$$\frac{(y_j/y_0)-c_j}{d_j}=\frac{1}{y_0}=\frac{(y_i/y_0)-c_i}{d_i},$$

and we may write

$$(y_i/y_0)=p_{ij}(y_j/y_0)+q_{ij}. \tag{4}$$

Since p_{ij} and q_{ij} are functions only of c_i, d_i, c_j, and d_j and are therefore independent of energy, Eq. (4) represents a straight-line relationship between the fission yields of the mass chains i and j relative to the standard mass chain.

We find it more convenient to deal with a quantity that is proportional to the relative fission yield and is usually known as an "R value."[4] The R value for mass chain i may be defined as follows:

$$R_i=\frac{(y_i/y_0)\text{ for given experiment}}{(y_i/y_0)\text{ for thermal neutrons on U}^{235}}.$$

The use of R values obviates the necessity of absolute counting, thereby making it possible to measure an R value more precisely than the fission yield itself. It should be noted that the denominator of the above expression is a fixed quantity, depending only on mass chain i and the choice of a reference standard. Therefore, the R value for mass chain i may be written

$$R_i=k_i(y_i/y_0).$$

We can now rewrite Eq. (4) as

$$R_i=\alpha_{ij}R_j+\beta_{ij}, \tag{5}$$

where α_{ij} and β_{ij} are constants. Thus we may use experimental R values to test whether or not there exist linear relationships between relative fission yields.

Tests of the two-mode fission hypothesis may be made by comparing the behavior of experimental mass-yield data with that predicted by the straight-line relationships just developed. There are a few things worthy of special note when seeking data with which to test the two-mode fission hypothesis. An adequate test of the predicted behavior requires that the mass yield data cover a sufficient range of values for both coordinates of the straight line. The greater the range in the excitation energies, the wider the range of values a particular mass yield will cover. However in our development we considered only fission occurring in a single nuclear species. In practice it is difficult to obtain large changes in a given mass yield over a range of excitation energies in which fission is produced by a single nuclide. Even at low excitation energies neutron emission will compete with fission in most nuclides. When the excitation energy is increased by a few Mev, fission can occur not only in the original compound nucleus, but also in the nucleus resulting after emission of a neutron. In many cases the mass distributions for each pair of basic fission modes for neighboring nuclides may be sufficiently similar so that no serious problem is encountered over reasonable ranges of excitation energies. However when contributions to the fission process from more than one nuclear species are energetically possible, they may well be the cause of deviations from predicted behavior. Although

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[4] W. E. Nervik, Phys. Rev. **119**, 1685 (1960).

ideal experimental conditions may be very difficult to attain, they can generally be most closely approximated by examining low-yield mass chains over a range of low-excitation energies, where these fission yields usually are changing most rapidly with small changes in energy.

One is not necessarily restricted to data from fission induced by monoenergetic particles. The observed mass distribution from fission produced by a spectrum of excitation energies can be considered a weighted average of the different mass distributions arising from fission by many single-excitation energies. If fission by a single-excitation energy produces a mass distribution which can be considered a linear combination of the mass distributions of the two hypothetical fission modes, then it follows that the mass distribution produced by a spectrum of excitation energies can also be considered a different linear combination of the mass distributions of these same two hypothetical fission modes. Fission produced by a spectrum of excitation energies can thus be treated as if it were the result of a single mean-excitation energy. Therefore, in an analysis of individual mass yields that does not involve the excitation energy explicitly (e.g., plotting the relative fission yield of one mass vs that of another mass), data obtained from bombardments carried out with projectiles of a mixed energy spectrum can be used together with data obtained from bombardments with monoenergetic particles.

III. EXPERIMENTAL

Fission at low-excitation energies is best effected by neutron bombardment because of the lack of a Coulomb barrier. Available sources of monoenergetic neutrons in this energy range do not have fluxes of high enough intensity to permit many radiochemical mass-yield determinations. However one can obtain neutron fluxes of mixed energies with sufficient intensities to permit a broad range of mass-yield determinations. It was pointed out that if linear relationships between individual mass yields do exist as predicted by the two-mode fission hypothesis, then these relationships hold regardless of the energy spectrum of the particles causing fission. Neutron fluxes with broad energy spectra can be produced by the Crocker Laboratory 60-inch cyclotron using charged-particle bombardment of various light elements. The intensities of these neutrons in the forward direction are much higher than can be obtained from monoenergetic sources. For these experiments we have used the following neutron spectra, listed by method of production, in order of increasing mean neutron energy: Be^9+12-Mev p^+; $\mathrm{Cu}^{63,65}+24$-Mev d^+; $\mathrm{Al}^{27}+48$-Mev He^{++}; Be^9+48-Mev He^{++}; Be^9+24-Mev d^+; and $\mathrm{Li}^{6,7}+24$-Mev d^+. No precise measurements were made of the mean energies of these neutron spectra, but roughly speaking they ranged from 2 or 3 Mev to about 10 Mev. In some cases bombardments were repeated using the same neutron source. No attempt was made to reproduce previous target positions so that the neutron fluxes and energy spectra probably differed somewhat. Data from these repeated bombardments should be similar but are not necessarily a true measure of reproducibility of the techniques. Each individual bombardment should provide a valid data point for testing linear relationships.

Targets were prepared from about 10 g of 0.001-in. uranium foil cut into about 25 pieces, each piece $\frac{5}{8}$ in. wide and 2 in. long. These pieces were stacked and wrapped with 0.001-in. 2*S* aluminum foil as a recoil catcher. One wrapped packet of enriched uranium (93% U^{235}) foils and one wrapped packet of natural uranium foils were stacked together and the whole wrapped again with 0.001-in. 2*S* aluminum foil to prevent external contamination. The whole target was then placed as close to the neutron source as possible and irradiated for about 5 hr, producing about 10^{15} fissions in each foil packet.

After irradiation the targets were stripped of the outer foil and separated. The inner aluminum wrapping foils were carefully removed and retained while the uranium foils were cautiously dissolved in hot concentrated nitric acid containing some hydrochloric acid. After the dissolution was complete, the aluminum foils were also dissolved in the proper target solutions. The solutions were cooled and diluted to known volume. Duplicate aliquots for each element were withdrawn and added to measured amounts of the proper carriers. Radiochemical purifications of samples were performed by standard methods[5–7] with some minor modifications.

In general the gamma radiations of the radioactive species were detected by a sodium iodide crystal, a photomultiplier tube, and an amplifier scaler. The lower discriminator of the amplifier scaler was set at 20 kev and the upper discriminator at 5 Mev. A beryllium absorber (1900 mg/cm^2) was placed between the sample and crystal to remove beta particles and to reduce effects of bremsstrahlung. Radiations from all nuclides were counted in this way except those from Sr^{89}, Pd^{109}, Pd^{112}, and Ag^{111}. In the last four cases beta particles were counted using a methane proportional counter, and self-scattering and self-absorption corrections were made. Bremsstrahlung from Y^{91} was counted on the gross gamma counter described above, since the amounts of Y^{91} were large enough to produce satisfactory counting rates.

The outputs of all scalers were attached to an IBM card-punching machine and data were automatically punched out as the individual count was completed. Data were taken over a period of three half-lives with at least four counts taken in any given half-life. This procedure was not practical, however, with $\mathrm{Cs}^{136,137}$.

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[5] M. Lindner, University of California Radiation Laboratory Report UCRL-4377 (unpublished).

[6] W. E. Nervik, J. Phys. Chem. **59**, 690 (1955).

[7] S. R. Gunn, H. G. Hicks, P. C. Stevenson, and H. B. Levy, Phys. Rev. **107**, 1642 (1957).

Here at least 20 counts were taken over a period of 4 weeks, the Cs^{136} activity allowed to decay to less than 1% of the Cs^{137} activity, and 5 more counts taken. When the data were complete the decay curves of the individual samples were analyzed by a least-squares method using the IBM 650 computer. The answers from the computer gave counting rates of each isotope corrected for decay (both during and after bombardment), chemical yield, aliquot, and self-scattering and self-absorption.

The R values of Sec. II can be rewritten as follows[4]:

$$R_i = \frac{(C_i/C_{\mathrm{Mo}^{99}}) \text{ for any type of fission}}{(C_i/C_{\mathrm{Mo}^{99}}) \text{ for } \mathrm{U}^{235}\text{, thermal neutron fission}},$$

where C_i is the corrected counting rate of nuclide i in its standard geometry and $C_{\mathrm{Mo}^{99}}$ is the corresponding value for Mo^{99}. The value of the denominator was measured for each nuclide in a series of calibration bombardments of U^{235} with thermal neutrons. In such a ratio the proportionality constant between counting rate (corrected for decay during bombardment) in a standard geometry and fission yield appears both in the numerator and the denominator. The constant will cancel, thus eliminating a measurement which can contain a large source of error.

The choice of fission-product nuclides was dictated by the desire to study a wide choice of fission products with principal emphasis placed upon nuclides having R values that would change by a large amount as the neutron energy increased. The behavior of such nuclides should provide the best test for the predicted linear behavior in the two-mode fission hypothesis. The fission products studied were Sr^{89}, Y^{91}, (Mo^{99}), Pd^{109}, Ag^{111}, Pd^{112}, Cd^{115}, Cs^{136}, Cs^{137}, Ba^{140}, Nd^{147}, Sm^{153}, Eu^{156}, and Tb^{161}.

The fission yields of all nuclides studied, except Cs^{136}, represent the total cumulative fission yield of that particular mass number. One measures only the independent yield of the shielded Cs^{136}, which was included in this study to compare the behavior of such a nuclide with the behavior of those representing the cumulative yield of an entire mass chain.

IV. RESULTS

The results of the experiments described in the previous section are presented in Tables I and II. Table I gives the R values for neutron-induced fission of U^{235}, and Table II gives the corresponding R values for neutron-induced fission of U^{238}. The R values for the nuclides listed represent the behavior of the total fission yield for that particular mass number with the sole exception of Cs^{136}, which is a shielded nuclide. In the case of Cd^{115}, R values were obtained from measurements on the 53-hr isomer. Some check measurements were made on the 43-day Cd^{115m} isomer and these indicated that over the energy range studied, the ratio of the two isomers remained essentially constant. Thus the R

TABLE I. R values for neutron-induced fission of U^{235}. (Standard deviations are ±5% unless otherwise given.)

Neutron source	Bbdt. No.	Sr^{89}	Y^{91}	Pd^{109}	Ag^{111}	Pd^{112}	Cd^{115}	Cs^{136}	Cs^{137}	Ba^{140}	Nd^{147}	Sm^{153}	Eu^{156}	Tb^{161}
p+Be	6	0.85	1.08	4.07	4.16	5.12	4.72	3.22	0.97	0.94	0.89	1.25	1.80	···
	11	0.83	1.03	4.09	4.34	5.47	4.69	2.91	0.87	0.94	0.90	1.20	1.72	6.59 ±0.46
	16	···	1.03 ±0.07	4.15	3.87	5.05	4.62	3.12	1.03	0.94	0.98	1.27	1.76	6.96
d+Cu	7	0.81	···	···	13.4 ±2.0	···	18.1	9.99	0.98	0.87	0.88	1.37	2.43	18.2
	14	···	···	14.9	17.6	23.4	22.9	···	···	1.02	···	···	···	···
α+Al	10	0.88	1.02	14.1	19.6	26.2	23.6	14.2	1.03	0.91	0.93	1.47	2.75	23.7
	12	0.77 ±0.07	1.00	14.0	17.4	24.6	22.2	13.3	1.03	0.92	0.92	1.42	2.69	24.0
α+Be	8	0.83	1.02	···	26.1 ±1.6	···	32.2	18.6	1.06	0.93	···	1.46	2.99	30.8
d+Be	5	0.84	1.06	21.4	26.5	34.1	33.6	18.5	1.01	0.86	0.88	1.48	3.00	32.1
d+Li	9	0.87	1.03	23.8	31.5	42.4	41.7	21.3	0.98	0.92	0.92	1.60	3.43	39.4
	13	0.91	1.10	23.1 ±2.3	33.1	42.3 ±4.2	38.9	22.9	1.10	0.99	0.96	1.62	3.47	39.2 ±2.7
	15	···	1.15 ±0.23	29.0 ±5.8	38.1 ±7.6	49.1 ±9.8	46.9 ±9.4	27.0 ±5.4	1.26 ±0.25	1.07 ±0.21	1.09 ±0.22	1.79 ±0.36	3.76 ±0.75	45.7 ±9.1

TABLE II. *R* values for neutron-induced fission of U^{238}. (Standard deviations are ±5% unless otherwise given.)

Neutron source	Bbdt. No.	Sr^{89}	Y^{91}	Pd^{109}	Ag^{111}	Pd^{112}	Cd^{115}	Cs^{136}	Cs^{137}	Ba^{140}	Nd^{147}	Sm^{153}	Eu^{156}	Tb^{161}
$p+Be$	11	0.50	0.73	9.28	6.88	7.66	5.23	0.16±0.03	0.82	0.89	1.06	2.54	5.46	22.6±2.5
	16	...	0.75	9.63	5.88	7.28	5.05	0.12±0.03	0.94	0.88	1.11	2.57	5.47	22.7
$d+Cu$	7	0.52	0.76	...	18.0±1.8	...	21.4	1.40±0.10	0.94	0.84	1.02	2.64	6.37	48.3
	14	...	...	21.4	19.6	26.0	22.4	...	...	0.96	...	...	...	...
$\alpha+Al$	10	0.54	0.73	19.1	22.2	28.0	23.5	1.98±0.14	0.94	0.87	1.10	2.75	6.37	49.2±3.9
	12	0.54±0.05	0.81	23.4	20.0	29.7	24.1	2.06±0.14	1.02	0.95	1.14	2.86	6.95	58.1
$\alpha+Be$	8	0.54	0.74	...	30.0±1.8	...	32.5	3.07±0.21	0.92	0.87	0.95	2.70	6.52±0.39	64.2
$d+Be$	5	...	0.76	27.2	29.3	36.1	33.4	2.32±0.16	0.91	0.83	1.04	2.85	7.14	68.0
$d+Li$	9	0.56	0.72±0.06	28.5	33.8	42.6	37.7	3.15±0.22	0.91	0.87	1.08	2.67±0.24	7.06±0.64	78.2±7.0
	13	0.57	0.76	28.6±2.9	33.1	39.7±4.0	35.7	3.10±0.22	0.93	0.91	1.09	2.97	7.35	72.6
	15	...	0.75	30.5	35.6	42.6	39.0	3.57±0.25	0.99	0.93	1.14	2.97	7.59	80.0

value for the 53-hr Cd^{115} does represent the *R* value for the mass number 115.

The *R* values given include corrections for the fact that the "U^{235}" target contained a small percentage of U^{238} and the "U^{238}" target (normal uranium) contained a small percentage of U^{235}. (Contributions from other isotopes were negligible.) In order to make this correction it was assumed that both targets in a given run were exposed to identical neutron fluxes. This probably was not strictly true, but the corrections in all cases except one were less than 3% and the possible contribution to the overall error was negligible. The exceptional case was the measurement of the *R* values of Cs^{136} formed by fission of U^{238}, which were an order of magnitude lower than those arising from the fission of U^{235} with the same neutron spectra. Therefore the contribution of Cs^{136} from the fission of the U^{235} present in natural uranium was a significant portion of the total Cs^{136} formed in the bombardment. Because of the size of these corrections, we have assigned somewhat larger standard deviations to the final *R* values of Cs^{136} from U^{238} fission.

In those cases where more than one bombardment was made using the same neutron source, certain activity ratios gave evidence that the flux profile was not always identical in all bombardments with the same neutron source. This being so, it is possible that there may have been some variation in the energy spectra between different runs with the same neutron source. Because of this we have treated the results of each bombardment as individual pieces of data rather than averaging the results from repeated runs.

Experience with many similar radiochemical determinations led us to expect an overall reliability of 5% for the *R* values in Tables I and II. This was borne out on comparison of the results of "repeated" runs. As we have pointed out, there was evidence to indicate that "repeat" runs were not strictly identical with regard to flux profile and possibly energy spectrum. However if one treats the *R* values from "repeat" runs as duplicate measurements of the same quantity and performs an analysis of variance, the average overall standard deviation indeed comes out to be about 5%.

We decided to assign a minimum standard deviation of 5% to all *R* values. Where the discrepancy between duplicate samples or some other evidence indicated a lower reliability, such results were assigned correspondingly larger standard deviations. In Table I the *R* values for bombardment number 15 were assigned greater than normal errors because the behavior of such nuclides as Y^{91}, Cs^{137}, Ba^{140}, and Nd^{147} indicated a possible systematic error in the Mo^{99} data.

It should be noted that the reliability discussed here refers to random errors and does not include any effect due to a possible systematic error present throughout the entire series of experiments. However the likelihood of such a systematic error being present in any significant magnitude is felt to be very small.

V. ANALYSIS OF RESULTS AND TESTS OF THE TWO-MODE FISSION HYPOTHESIS

The data presented in Tables I and II can be tested to see if they are consistent with the two-mode fission hypothesis by choosing the *R* values of one species to be the values of the *x* coordinate and then plotting the corresponding *R* values of the other species as the *y* coordinates. Our treatment of the two-mode fission hypothesis in Sec. II predicts that we will obtain a straight line for each set of *R* values. To obtain the most suitable plots one should choose for the *x* coordinate the *R* values of a mass number whose relative fission yield undergoes a large change over the energy range studied. We chose Cd^{115} for this purpose because of the wide range and generally good reliability of its *R* values.

It would be meaningless with respect to the test to plot data for a nuclide whose *R* values remain essentially unchanged since these would obviously fall on a nearly horizontal line. Only those species whose *R* values show a considerable change are significant, and the *R* values for these species have been plotted as *y* coordinates against the corresponding *R* values for Cd^{115}.

The data plotted in this way are presented in Figs. 1 and 2. The straight lines shown are those fitted to the data in a manner described below. It is clear that for all these species, including the shielded nuclide Cs^{136}, the data are quite consistent with the straight-line behavior predicted by the two-mode fission hypothesis. In the energy range covered there appears to be no significant tendency toward curvature, even in the case of Cs^{136} in U^{238} fission where the data are most scattered. These plots include all data for these nuclides obtained in this set of experiments. (No pertinent data from Tables I and II were thrown out because of large deviation from the line.)

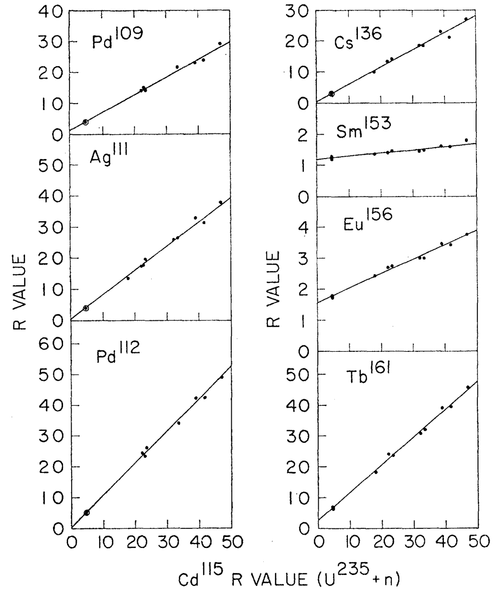

FIG. 1. *R* values of various species plotted vs Cd^{115} *R* values for neutron-induced fission of U^{235}.

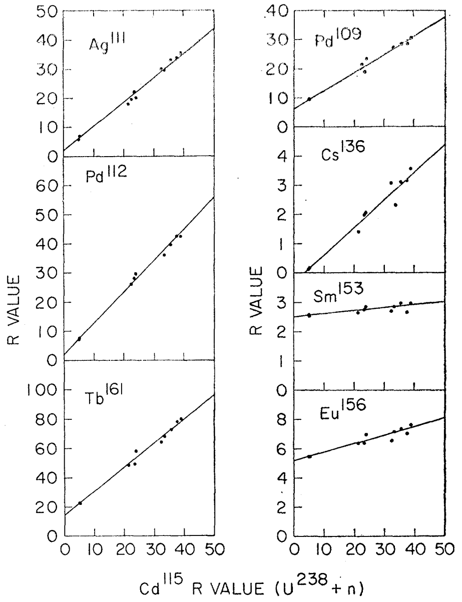

FIG. 2. *R* values of various species plotted vs Cd^{115} *R* values for neutron-induced fission of U^{238}.

The excitation energies produced by the neutron fluxes used are sufficiently high for us to expect that the mass yields represent significant contributions from more than one nuclear species undergoing fission. However the mass distributions of the two basic modes of the neighboring parent nuclides appear to be similar enough so as not to perturb the linear behavior.

The *R* values for thermal neutron fission of U^{235} should also fall on the corresponding lines in Fig. 1. Since an *R* value is simply the relative fission yield of a species normalized to the relative fission yield of the same species in thermal-neutron fission of U^{235}, the *R* values for all species in thermal-neutron fission of U^{235} by definition must be equal to 1. Therefore the lines in the above plots representing behavior of *R* values in U^{235} fission should all pass through the point (1,1). Such does not appear to be the case for all the lines in U^{235} fission. Since this question is of considerable significance, some additional analysis of the straight lines is warranted.

We wished to fit a straight line of the form $y=a+bx$ to each set of data (where y is the *R* value of some species and x is the *R* value of Cd^{115}), using a least-squares method to obtain values of a and b for each case. We can then examine how well these lines fit the experimental points and to what extent the lines may deviate from the point (1,1). The point (1,1) was ***not*** used in obtaining the least-squares line.

As far as we could ascertain the literature contains no complete treatment of the problem of performing a least-squares fit of a straight line to points for which both the x and y values are subject to errors that vary from point to point. Problems arise both in the assigning of proper weights to the points and in the solving of the normal equations. Deming[8] offers a method based on statistical adjustment of data. However we preferred to use a procedure based on suggestions made to us by Kaplan[9] of this laboratory. Both methods are based on the assumption that errors in x and y are independent. This is not strictly true in our case since an error in the determination of Mo^{99} will affect all R values in a like fashion. Under normal conditions total activities (counting rates) are determined with approximately the same percentage precision regardless of the activity level, provided the counting rate is above a certain minimum. In the least-squares method used, the ignoring of the correlation between the x and y errors due to the dependence of both R values on the Mo^{99} activity would have the effect of multiplying the weight assigned to each point by a factor which is approximately constant. Any constant factor would drop out when the normal equations were derived. Also overall errors in x and y are small compared to the total range covered. We therefore conclude that neglecting the correlation between the x and y errors in this case would have only a very slight effect on the values of the parameters a and b.

The normal equations to be solved for a and b were obtained by minimizing with respect to both a and b the weighted sum of the squares of the deviations from the proposed line. The weight of each point is chosen to be the reciprocal of the variance of that particular deviation, taking into account errors in both x and y. If the weights are assigned in this fashion, then regardless of whether we choose to minimize deviations in the y direction, deviations in the x direction, or perpendicular distances from the point to the line, the expression for the weighted sum of squares becomes $S=\sum(y_i-a-bx_i)^2/(v_i^2+b^2u_i^2)$. In this expression u_i is the standard deviation of x_i, and v_i is the standard deviation of y_i. Because of the way in which the parameter b appears in the sum S, we could not solve the normal equations directly for a and b. A method of successive approximations was used using an IBM 650 computer. The values of a and b determined for each of the lines shown in Figs. 1 and 2 are given in Tables III and IV.

TABLE III. Results of least-squares fit of straight lines to R values of various species plotted vs the R values for Cd^{115} in fast-neutron-induced fission of U^{235}.

Nuclide	Intercept a	Slope b	Number of data points	Mean % deviation, y_{calc} and y_{meas}	$S=\sum\dfrac{(y_i-a-bx_i)^2}{v_i^2+b^2u_i^2}$
Pd^{109}	1.477	0.5617	10	3.6%	2.15
Ag^{111}	0.453	0.7809	12	5.0%	3.74
Pd^{112}	0.336	1.0431	10	3.9%	2.42
Cs^{136}	0.507	0.5498	11	5.4%	5.20
Sm^{153}	1.194	0.00976	11	3.6%	2.29
Eu^{156}	1.549	0.04700	11	2.9%	2.47
Tb^{161}	2.650	0.8952	10	3.7%	2.15

TABLE IV. Results of least-squares fit of straight lines to R values of various species plotted vs the R values for Cd^{115} in fast-neutron-induced fission of U^{238}.

Nuclide	Intercept a	Slope b	Number of data points	Mean % deviation, y_{calc} and y_{meas}	$S=\sum\dfrac{(y_i-a-bx_i)^2}{v_i^2+b^2u_i^2}$
Pd^{109}	6.255	0.6246	9	5.8%	6.33
Ag^{111}	2.019	0.8272	11	6.4%	7.02
Pd^{112}	1.962	1.0754	9	3.5%	1.81
Cs^{136}	−0.348	0.0938	10	12.2%	11.85
Sm^{153}	2.497	0.01029	10	4.1%	3.49
Eu^{156}	5.170	0.05743	10	4.0%	3.66
Tb^{161}	14.340	1.6317	10	4.4%	3.23

Also given in Tables III and IV are some statistics which can serve as rough measures of how well the calculated lines $y=a+bx$ fit the experimental data. Listed are the number of experimental points for each line, the mean percentage deviation of the calculated and measured values of y, and the value for the minimized sum of squares, $S=\sum(y_i-a-bx_i)^2/(v_i^2+b^2u_i^2)$. The mean percentage deviation of the calculated and measured y values was computed for each line by the expression

$$100\%\times\left[\frac{1}{n-2}\sum\left(\frac{y_{\mathrm{meas}}-y_{\mathrm{calc}}}{y_{\mathrm{meas}}}\right)^2\right]^{\frac{1}{2}},$$

where n is the number of experimental points. This "average" deviation can be compared to the standard deviations given for the experimental data. The values for the minimized sum of squares S appear to be too low, despite the fact that no data points were excluded. This can be attributed, in part at least, to our ignoring the correlation between the x and y errors. We have no accurate measure of the correlation, but if we make the assumption that the activities of all nuclides (including Mo^{99}) are determined with the same percentage accuracy, the effect of ignoring this error correlation would cause S to be low by roughly a factor of 2. If these lines represent a normal fit to the experimental data, then the value of the total sum of squares divided by the total number of degrees of freedom should be on the order of 1. When we consider all the cases in Tables III and IV together, this ratio has the value of 0.498. When we attempt to correct this number for the effect of ignoring correlation between errors by multiplying by 2, the result is very close to 1. That this corrected value is so close to 1 is undoubtedly a coincidence, but it does indicate that these lines represent a "good fit" to the experimental data.

The choice of Mo^{99} as a standard relative to which the other fission yields are measured is arbitrary, as is the choice of the R values of Cd^{115} for the x coordinates in the graphs. Although these choices appeared to be most

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[8] W. E. Deming, *Statistical Adjustment of Data* (John Wiley & Sons, Inc., New York, 1938), 1st ed.

[9] Edward L. Kaplan (private communication).

suitable, other nuclides could have been used for these purposes. The authors have tried several of these other possibilities without noting anything that would alter the conclusions given here.

We conclude that the data presented in this paper for fission induced in U^{235} and U^{238} by fast neutrons are consistent with the behavior predicted by the two-mode fission hypothesis. In Sec. II we made the assumption that the mass distributions of the two fission modes did not change with increase in excitation energy of the nucleus undergoing fission, but said nothing concerning charge distribution. The R values for the shielded nuclide Cs^{136} represent the relative independent yield of a given charge and mass, whereas the R values for all the other nuclides represent relative total-mass-chain yields. It is interesting to note that the Cs^{136} data from both U^{235} and U^{238} fission also appear to follow a linear behavior, although the fit is not quite as good as for the other nuclides. This linear behavior suggests that the charge distributions of the two modes also remain relatively unchanged with increasing excitation energy. Comparison of the rapid rise of the Cs^{136} R values with the relatively constant Cs^{137} R values further suggests that the charge distributions for each of the two basic modes differ quite markedly.

As was pointed out previously, if the data from thermal neutron fission were to be consistent with the two-mode interpretation of the fast-neutron fission data, then the lines for the fast-neutron fission of U^{235} should all pass through the point (1,1). Using the parameters of the lines obtained above we can calculate the R values for the various other species when the R value for Cd^{115} is equal to 1. Table V lists these calculated values for neutron-induced fission in U^{235}. With the exception of Cs^{136}, the deviation of the various lines from the point (1,1) is far greater than could be reasonably expected from statistical considerations alone.

It is quite probable that the fission process is much more complex than the picture set forth in Sec. II. The discrepancy between the thermal-neutron data and the fast-neutron data in U^{235} fission would seem to bear this out.

It does not seem likely that this discrepancy is entirely explained by the fact that thermal neutron fission represents fission from a single nuclide, whereas the fast-neutron fission-yield data represents fission from more than one nuclide. If contributions from a second species undergoing fission were such as to cause the lines to deviate from the thermal neutron point, they would also be most likely to cause a noticeable curvature in the lines. However over certain ranges of excitation energies, the two-mode fission hypothesis may serve as a very good approximation, and this manner of plotting fission-yield results may be a useful means of correlating data. In any case, any hypothesis proposed as an alternative to that of the two modes of fission will have to be able to explain the linear behavior of the data shown here.

TABLE V. Values of R_i for $R(Cd^{115})=1$ (thermal fission) calculated from the lines fitted to the data for fast neutron fission of U^{235}.

Nuclide	Pd^{109}	Ag^{111}	Pd^{112}	Cs^{136}	Sm^{153}	Eu^{156}	Tb^{161}
R_i	2.04	1.23	1.38	1.06	1.20	1.60	3.55

The authors will publish shortly some R value data for the fission of the U^{236} compound nucleus at somewhat higher excitation energies than those studied here. These excitation energies were attained by bombarding Th^{232} with helium ions. Although the data do deviate from linear behavior at higher excitation energies, they do not contradict conclusions drawn here.

VI. SUMMARY

It was shown that under certain conditions the two-mode fission hypothesis predicts certain straight-line relationships between yields of the various fission products. New data have been presented on relative yields from fission induced in U^{235} and U^{238} with fast neutrons, and it has been shown that these data follow the straight-line relationships predicted on the basis of the two-mode fission hypothesis. The shielded nuclide Cs^{136} appears to behave in the same fashion as the nuclides that represent total cumulative chains. This would suggest that if the two-mode fission hypothesis were valid, the respective charge distributions of the two fission modes as well as their mass distributions remain unchanged with changing excitation energy.

However it is pointed out that consistency with the two-mode fission hypothesis would require that the data from thermal-neutron fission of U^{235} fall on the same lines obtained from the fast-neutron fission of U^{235}. This is shown to be definitely *not* the case. Before one can accept the two-mode fission hypothesis, this discrepancy must be resolved. The authors wish to suggest however, that the straight-line relationships observed here appear to be more than just a coincidence, and that any alternative model will have to account for this behavior.

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