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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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ABSTRACT

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.

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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

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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,

$f_B = 1 - f_A$ = fraction of total fissions resulting in Type B fission.

We can then write for the observed fission yield of i,

$$y_i = f_A a_i + f_B b_i = f_A a_i + (1 - f_A) b_i . \quad (1)$$

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

Hence

$$f_A = \frac{y_i - b_i}{a_i - b_i} .$$

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

$$f_A = \frac{y_j - b_j}{a_j - b_j} .$$

Therefore

$$\frac{y_i - b_i}{a_i - b_i} = \frac{y_j - b_j}{a_j - b_j} .$$

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

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$$y_i = c_{ij} y_j + d_{ij} , \quad (2)$$

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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:

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

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

$$R_i = \alpha_{ij}R_j + \beta_{ij} , \qquad (5)$$

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

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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^+; $Cu^{63,65}$ + 24-Mev d^+; Al^{27} + 48-Mev He^{++}; Be^9 + 48-Mev He^{++}; Be^9 + 24-Mev d^+; and $Li^{6,7}$ + 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.

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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 $Cs^{136,137}$; 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]

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

where C_i is the corrected counting rate of nuclide i in its standard geometry and $C_{Mo^{99}}$ 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.

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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,

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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.)

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.

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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 $S = \sum (y_i - a - bx_i)^2 / (v_i^2 + b^2 u_i^2)$. 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, with 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.

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, $S = \sum (y_i - a - bx_i)^2 / (v_i^2 + b^2 u_i^2)$. The mean percentage deviation of the calculated and measured y values was computed for each line by the expression

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

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,

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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

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VII. ACKNOWLEDGMENTS

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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	---	---	---	---
α + 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
α + 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

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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}	$S=\sum \frac{(y_i-a-bx_i)^2}{v_i^2+b^2u_i^2}$
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

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

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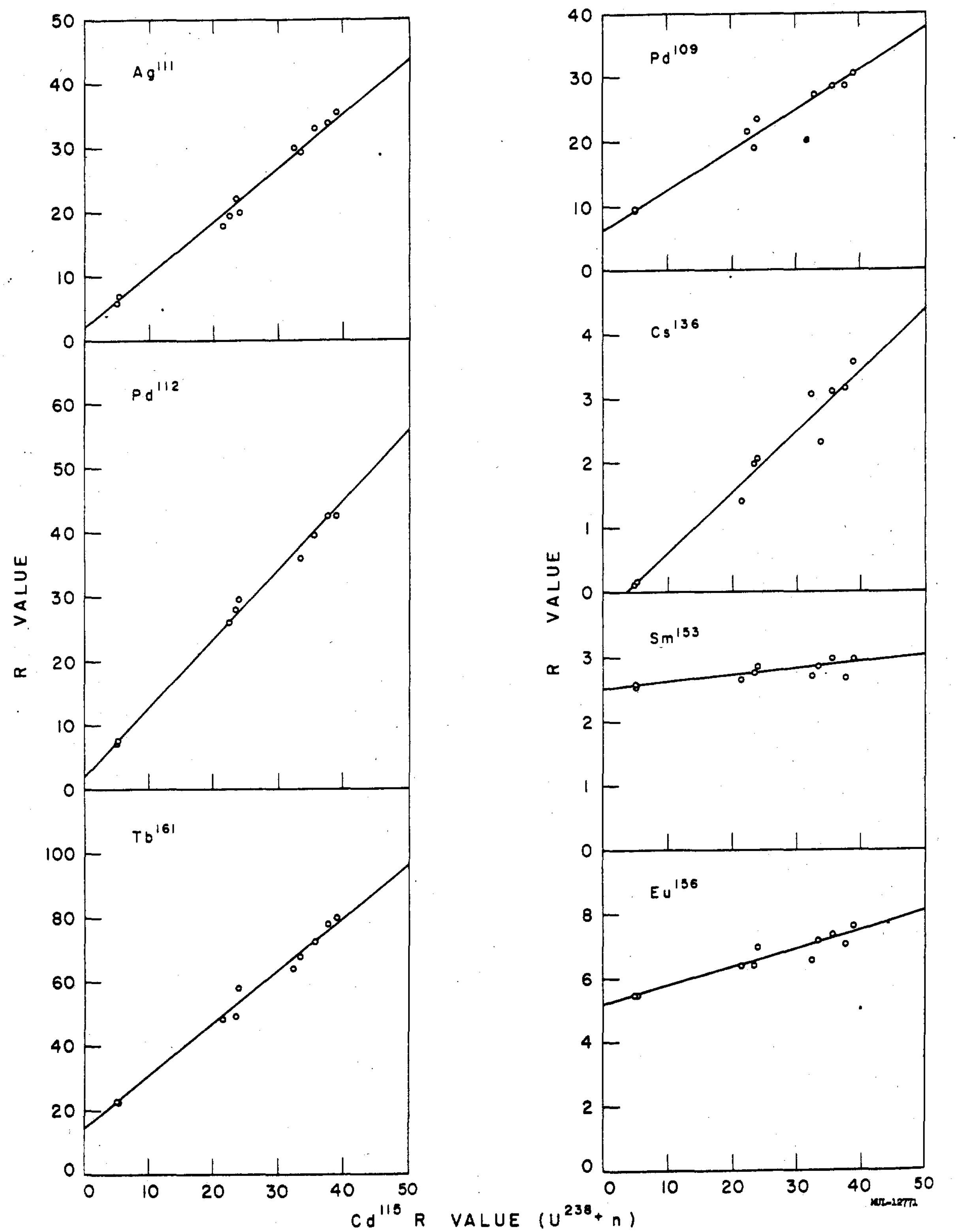

Fig. 2. R values of various species plotted vs Cd^{115} R values for neutron-induced fission of U^{238}.

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