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MASS YIELDS FROM FISSION BY NEUTRONS

BETWEEN THERMAL AND 14.7 MEV

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ABSTRACT

Radiochemically determined mass-yield curves are given for the fission of  $U^{235}$  and  $U^{238}$  by 14.7-Mev neutrons. Symmetric and to a less extent, very asymmetric modes of fission are more probable at that energy than in thermal fission. Yields of four fission products from the fission of  $U^{235}$  have been measured as a function of neutron energy in the range thermal to 14-Mev.

The yields of eleven masses have been measured from the fission of  $Np^{237}$  by degraded fission spectrum neutrons. The mass-yield curve is similar to that from the thermal fission of  $Pu^{239}$  with a ratio of peak to valley yields of approximately 175.

Relative yields of one peak fission product and four valley fission products have been determined under the following conditions: fission of  $U^{235}$  and  $Pu^{239}$  with thermal neutrons; fission of  $U^{235}$ ,  $Pu^{239}$  and  $U^{238}$ with fission spectrum neutrons; and fission of  $U^{235}$  and  $Pu^{239}$  with the intermediate neutron spectrum at the center of the Los Alamos Fast Reactor. Absolute yields of Mo<sup>89</sup> have been determined from the fission of  $U^{233}$ ,  $U^{235}$ , and  $Pu^{239}$  with thermal neutrons.





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## I. INTRODUCTION

Fission has been observed in a wide variety of nuclides over a large range of excitation energies. For thermal fission, symmetric splitting and very asymmetric splitting are both highly improbable. When fission yield is plotted against mass number (mass-yield curve), the familiar doublepeaked curve is obtained with a deep valley between the peaks. In general the mass-yield curves vary with fissioning nuclide and with excitation energy. This paper presents mass-yield curves for the fission of  $U^{235}$  and  $U^{238}$  with 14.7-Mev neutrons. Yields of some of the fission products that are indicative of the energy dependence of the yield curve have been measured for neutron energies between thermal and 14.7 Mev. In addition, representative fission yields for Np<sup>237</sup> and certain valley nuclide fission yields for  $U^{235}$ ,  $U^{238}$ , and  $Pu^{239}$  are given for fissions induced by various neutron spectra similar to those met in reactor technology. The method used was a radiochemical comparison of these yields with yields from the thermal fission of  $U^{235}$ .

In fission at relatively low excitation energies, the fission products are  $\beta^-$  unstable. The primary fission fragments for a given mass chain do not all have the same nuclear charge. For any one mass chain some nuclear charge will be formed with highest yield, and there will be appreciable contributions from charges different by one, smaller contributions from charges different by two, and so forth. The percent of fissions resulting in the direct formation of a nuclide is defined as its independent yield. The independent yield of a nuclide plus the independent yields of its predecessors is its cumulative yield. The sum of all independent yields of a given mass is the total chain yield. In order to measure a total chain yield by radiochemical methods, it is desirable to analyze for a late member of the chain. The last radioactive member is best, but practical considerations may rule against this choice.





II. GENERAL PROCEDURES

## A. Radiochemistry

In the radiochemical assays, an inactive carrier of the element for which an analysis was being made was added to an aliquot of the solution containing fission products. After steps to encourage exchange, the element was separated chemically from other elements present, weighed as some suitable compound to determine chemical yield, and then mounted for counting. Details of the chemical and counting procedures are given in a report by Jacob Kleinberg et al.<sup>1</sup> Methane flow beta proportional counters<sup>2,3</sup>,<sup>4</sup> were used for all determinations except that of  $Cs^{136}$ , which was counted on a sodium iodide scintillation counter. The decay of samples was followed to check radiochemical purity and to resolve mixed activities in those cases where more than one isotope of the same element contributed to the counting rate. Counting rates were corrected for chemical yield, decay, and self-absorption to obtain activities at the end of irradiation.

## B. Treatment of Data

Records of neutron intensity as a function of time were used to correct activities for decay during irradiation. The following treatment applies to all of the cases dealt with in this work. Consider the case of a two-member mass chain for which no chemical separations were made until less than 0.1/2 of the parent remained. Let  $k_1$  be the fraction of the yield produced as the parent, while  $k_2 = 1 - k_1$ , the fraction of the yield produced directly as the daughter. It can be shown that for the daughter, the correction for decay during irradiation differs from the case of the one-member chain only by division by the factor  $k_1 + (k_2 \lambda_1)/(\lambda_1 - \lambda_2)$  where  $\lambda_1$  and  $\lambda_2$  are the decay constants of the parent and daughter. For many fission product chains, corrections for decay during irradiation were made as if the nuclides were formed directly in fission since  $\lambda_1 \gg \lambda_2$  and  $k_1 + (k_2 \lambda_1)/(\lambda_1 - \lambda_2)$  is near unity. Where there are no known predecessors we have assumed this to be the case. The few cases requiring information on the values of  $k_1$  and  $k_2$  will be discussed later.

All yields that we report were determined by the comparison method except some absolute yields of  $Mo^{99}$ . To avoid absolute beta-counting we compare all other fission yields with the yields from thermal neutron fission of  $U^{235}$ . We measure the ratio of a fission product activity to the activity of the peak nuclide  $Mo^{99}$  which we have chosen as standard. If s denotes the standard and x denotes any other fission product, then

$$\frac{A_{x}}{A_{s}} = \frac{\epsilon_{x} \lambda_{x} y_{x}}{\epsilon_{s} \lambda_{s} y_{s}} .$$
 (1)





A is a counting rate, or activity, obtained as explained under Radiochemistry, and further corrected for decay during irradiation,  $\in$  is the fraction of the disintegrations that are counted, and y is the fission yield. If  $A_x/A_s$  is measured for  $U^{235}$  thermal neutron fission and also for fission under some other conditions (different fissioning nuclide or different excitation energy), then we define a quantity R as follows;

$$R = \frac{A_x^{\dagger}/A_s^{\dagger}}{A_x/A_s}, \qquad (2)$$

where the unprimed activities refer to thermal neutron fission of  $U^{235}$  and the primed activities to any other kind of fission. From equations (1) and (2) it follows that

$$y_{X}^{*} = y_{X} \frac{y_{S}^{*}}{y_{S}} R \qquad (3)$$

Equation (3) is used to calculate cumulative and independent yields. The chief disadvantage of this method is that the yields are no more accurate than the assumed thermal neutron mass-yield curve. The  $U^{295}$  yields,  $y_x$ , were selected from references 5 through 10 and are given in Table III. For valley yields of less than 1% we have assumed that the mass-yield curve is symmetrical about mass 117.

The yield of  $Mo^{99}$  from thermal fission, y<sub>s</sub>, and the yields at other energies for U<sup>235</sup> and U<sup>238</sup>, y'<sub>s</sub>, were determined by Terrell et al.<sup>5</sup> For fission by neutrons of less than fission-spectrum energy we have assumed that the  $Mo^{99}$  yield, y'<sub>s</sub>, has not changed from thermal fission of the same nuclide, and y'<sub>s</sub> for Np<sup>237</sup> is the same as for thermal U<sup>235</sup>. The thermal yield of  $Mo^{99}$  from Pu<sup>239</sup> is from this work.

### III. EXPERIMENTS AND RESULTS

## A. Absolute Yields of Mo<sup>99</sup>

Irradiations to determine the absolute cumulative yield of Mo<sup>99</sup> were made in the thermal column of the Los Alamos Homogeneous Reactor. A small sample of fissionable material was placed between two monitor foils of the same material in an argon-filled twin-chamber fission counter.<sup>11</sup> Fissions in the monitors were counted during irradiation so that the fissions occurring in the sample could be calculated.

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The fission pulse spectra, as recorded with a 100-channel pulse height analyzer,<sup>12</sup> were used to estimate the small number of fission pulses lost below the bias of the counters. Since the sample was separated from each monitor by a 0.017 cm aluminum mounting plate and the 0.0013 cm platinum backing of the foil, the flux in the sample was assumed to be the same as in the monitors.

Monitor foils were prepared by electrodepositing the following approximate weights of fissionable material on a 2.54 cm diameter platinum disc: for  $U^{235}$ , 0.060 //g; for  $U^{233}$ , 0.035 //g; and for  $Pu^{239}$ , 0.100 //g. The samples for the  $U^{235}$  irradiation were approximately 6 mg of oxide on a 0.025 cm thick by 2.54 cm diameter nickel backing. Samples of  $U^{233}$  (1 mg) and  $Pu^{239}$  (10 mg) were prepared on 0.0025 cm thick nickel backings. A 0.0025 cm thick nickel foil was placed over the sample and crimped around the edge. After irradiation the entire package was dissolved for analysis. Mo<sup>99</sup> was separated and counted on a beta proportional counter which had been calibrated for absolute beta-counting. The relative amounts of fissionable isotope in the solution and on the monitor plates were determined by alpha counting for  $U^{233}$  and  $Pu^{239}$ , and by comparison fission counting for  $U^{235}$ .

The procedure used to calibrate the beta counter was to count freshly separated Mo<sup>99</sup> of high specific activity (40,000 disintegrations per minute in less than 10  $\mu$ g of solids) in a  $4\pi$ -geometry counter. The effect of the 6-hour Tc<sup>89</sup> daughter was subtracted by a least squares solution of the growth and decay curve. A second aliquot was processed and beta counted in the usual way to determine the ratio of counts to Mo<sup>99</sup> disintegrations. Although a period of two years elapsed between the calibration of the beta proportional counter and these absolute yield measurements, a UX reference standard showed negligible change during this time.

The results of our measurements agree reasonably well with those of other investigators as shown in Table I. Three measurements were made on  $U^{233}$ , six on  $U^{235}$ , and four on  $Pu^{239}$ . For each measurement four assays were made for Mo<sup>99</sup>. The errors quoted are larger than the standard deviations of the results (1%) to allow a reasonable margin for systematic errors which we are unable to estimate. If these yields were considered on a relative rather than an absolute basis, the errors would of course be smaller.







Absolute Cumulative Yields of  $Mo^{99}$  from Thermal Neutron Fission (in %)

<u> </u>	U <sup>233</sup> .	U <sup>235</sup>	Pu <sup>239</sup>
This paper NNES <sup>6</sup> Terrell <sup>5</sup> Reed <sup>7</sup> Steinberg <sup>13</sup>	4.96 ± 0.15 4.7 5.1	6.25 ± 0.19 6.2 6.14 ± 0.16 5.98 ± 0.18	6.02 ± 0.18 6.1

## B. Thermal Irradiations

Thermal activity ratios for use in the comparison method were determined from highly enriched samples of  $U^{235}$  irradiated in the thermal columns of the Los Alamos Fast Reactor and the Los Alamos Homogeneous Reactor. The cadmium ratios of the neutron spectra (activity of a bare indium detector divided by the activity of the same detector sheathed in cadmium) were 500 to 1.

Cumulative thermal yields for four valley nuclides from thermal fission of Pu<sup>239</sup> are included in Table V. From experience, the standard deviations on these analyses are approximately 2%.

## C. Fission Product Yields as a Function of Neutron Energy

R values were determined for  $Ag^{111}$ ,  $Cd^{115}$ ,  $Cs^{136}$ , and  $Ce^{143}$  from the fission of  $U^{235}$  with neutrons of known energy. Approximately 25-gram samples of enriched  $U^{235}$  were irradiated at the two Los Alamos Van de Graaff accelerators, using the p-t and d-d reactions as neutron sources. The spread in neutron energies for the 1.2- and 3.1-Mev irradiations was approximately 0.4 Mev; for the 5-Mev irradiation it was approximately 1 Mev. Mo<sup>99</sup>,  $Ag^{111}$ , and  $Cd^{115}$  were also determined from the fission of uranium or normal isotopic composition with 3.1-Mev neutrons. There was one irradiation at each energy, and only the molybdenum, cadmium, silver, and cerium analyses were made in duplicate. Fission yields,  $y_1^*$  were based on the Mo<sup>99</sup> yields,  $y_1^*$ , reported by Terrell. In neither case was it necessary to correct for the fissions in the other fissionable isotopes. The cumulative yields from these measurements are given in Table II. The errors of the R values used in equation (3) should be less than 5%.





Cumulative Fission Yields from  $U^{23}$  and  $U^{235}$  as a Function of Neutron Energy

		Cumulative Fission Yield, %				
	Neutron Energy	Ag <sup>111</sup>	Cd115	Cs136	Ce <sup>143</sup>	
U532	Thermal 1.2 Mey	0.0143 <sup>a</sup>	0.0104 <sup>a</sup> 0.019	0.0062 <sup>a</sup>	5•7 <sup>a</sup>	
	5 Mev 8.1 Mev	0.11 0.30	0.097 0.26	0.032	4.61	
	14.7 Mev	0.92	0.89	0.24	3.72	
U238	8.1 Mev	0.24	0.15			
	14.7 Mev	0.87	0.71	0.029		

a - from Col. 2, Table III.

## D. 14.7 Mev Neutron Fission of U<sup>235</sup> and U<sup>238</sup>

These irradiations were made at the Los Alamos Cockroft-Walton accelerator, using neutrons from the d-t reaction. The estimated average energy of the neutrons was 14.7 Mev, with a spread from 13.4 to 15.2 Mev. For  $U^{235}$ approximately 6-gram samples of highly enriched metal were placed as close as possible to the tritium target. Since the R values are independent of uncertainty in the thermal mass-yield curve, they are shown in Table III. Most of the errors shown are statistical, to indicate the precision of the results rather than to include any systematic error. They have been calculated from the standard deviation of the mean of the activity ratios by the usual rules for propagation of errors. For Sb<sup>126</sup>, <sup>129</sup> and Eu<sup>156</sup> the results are based on too few determinations to warrant this treatment.

The cumulative yields in Table III were calculated by equation (3). Terrell's yields for Mo<sup>99</sup> were used for  $y_{g}^{*}/y_{g}$ . Our yields, of course, reflect any errors in the thermal fission yields  $(y_{x})$  given in the table.

The R values, yields, and fractional chain yields of three shielded nuclides from  $U^{235}$  fission are shown in Table IV. These yields represent independent formation and may enable us to conclude something about the distribution of nuclear charge in fission.

Charge distribution enters the calculation of yields in two ways: first, in some of the R values, where  $k_1$  and  $k_2$  must be estimated for the decay correction; second, in calculation of total chain yields from cumulative yields. Since the charge distribution in fission changes rapidly by beta decay, it is not nearly as well known as the mass distribution. Glendenin, Coryell, and Edwards<sup>14</sup> introduced the hypothesis that for all mass numbers the charge distribution should be described by the same smooth symmetric curve. Applying this criterion to thermal fission yields, they rejected several theories and proposed the empirical hypothesis of equal charge displacement.



## TABLE III

<del></del>		U <sup>235</sup>			U <sup>238</sup>	** <del>**</del> ********************************
Nuclide	Thermal yield, %	R	Cumulative yield, %	Estimated total yield, %	R	Cumulative yield, %
2.4-hr Br <sup>8</sup>	<sup>3</sup> 0.62	2.5 ± 0.2	1.30	1.30		······································
54-day Sr <sup>8</sup>	<del>9</del> 4.78	1.09 ± 0.01	4.38	4.38	0.69 ± 0.01	3.0
9.7-hr Sr <sup>9</sup>	1 5.07	0.99 ± 0.02	4.19	4.19		
17-hr Zr <sup>9</sup>	7 5.50	1.05 ± 0.01	4.87	4.96	0.94 ± 0.01	4.8
67-hr Mo <sup>9</sup>	9 6.14		5.17	5.17		5.7
40-day Rul	<sup>03</sup> 2.85	1.38 ± 0.01	3.31	3.31		:
36-hr Rh <sup>1</sup>	0.92	$2.41 \pm 0.14$	1.86	1.86	3.98 ± 0.08	3.4
1-year Rul	06 0.345	4.86 ± 0.04	1.41	1.41		
13.4-hr Pd <sup>10</sup>	0.024	51.0 ± 0.4	1.03	1.03		
7.5-day Ag1	0.0143	76.3 ± 0.8	0,92	0.92	65.8 ± 0.9	0.87
21-hr Pd <sup>1</sup>	0.0127	94.7 ± 0.8	1.01	1.05		••••
53-hr Cd <sup>1</sup>	15 0.0115	101.4 ± 1.5	0.89	0.89	74 = 3	0.71
27-hr Sn <sup>1</sup>	<sup>21</sup> 0.0115	96 <b>. ±</b> 3.	0.93	0.93	69 <b>±</b> 2	0.73
10-day Sn <sup>1</sup>	25 0.024	74.6 ± 1.4	1.51	1.55	. 36 ± 1	0.83
9-hr Sb <sup>13</sup>	26 0.046	<u>3</u> 8• ≠2	1.47	1.47	·	
95-hr Sb <sup>14</sup>	<sup>27</sup> 0.110	20.6 + 0.3	1.9	1.9	14.1 ± 0.4	1.43
4.2-hr Sb <sup>12</sup>	29 0.92	2.7 ¥ 0.3	2.1	2.4		
8-day I <sup>13</sup>	2.97	1.62 ± 0.03	4.05	4.05		
77-hr Te <sup>10</sup>	<sup>32</sup> 4.45	1.13 ± 0.03	4.23	4.91	1.14 ± 0.05	4.7
21-hr I <sup>133</sup>	<sup>3</sup> 6.62	·	-	5.5 a		
52-min 1134	4 7.81			5.8 a		
6.7-hr 113	5 6.4			5.5 a		
85-min Bala	<sup>39</sup> 6.5	0.89 ± 0.04	4.87	4.97		
12.8-day Ba	6.3	0.86 ± 0.02	4.58	5.05	0.79 ± 0.02	4.6
33-hr Cel	5.7	0.77 ± 0.01	3.72	3.72		
280-day Cel	4 4.90	0.69 ± 0.02	2.75	3.11	0.75 ± 0.03	3.4
15-day Eul!	56 0.027	4.6 ± 0.2	0.11	0.11	8.1 = 0.3	0.22

## Fission Yields from 14.7-Mev Neutron Induced Fission

a - from reference 17

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Yields of Shielded Nuclides from 14.7-Mev Neutron Fission of U<sup>235</sup> and U<sup>235</sup>

		US32			U238	<u></u>
Nuclide	R	Yield, %	Fraction of chain	R	Yield, %	Fraction of chain
35.9-hr Br <sup>82</sup> 23-hr Nb <sup>96</sup> 13-day Cs <sup>136</sup>	$\begin{array}{r} 44 + 2 \\ 6 + 1 \\ 46 \cdot 0 + 0 \cdot 5 \end{array}$	0.0015 0.0029 0.24	0.0016 0.00059 0.045	5.1 +	0.2 0.029	0.0052

Pappas <sup>15</sup> has proposed that equal charge displacement applies before neutron emission. To calculate an independent yield by his method, given A and Z, first calculate

 $Z_{p} = Z_{A+n} - 1/2(Z_{236-A-n} + Z_{A+n}$   $Z_{A+n} \text{ and } Z_{236-A-n} \text{ are from the lines} \qquad 1.0$ of maximum stability given by Coryell.<sup>16</sup>  $Z_{A+n}$  and  $Z_{236-A-n}$  are often called the "most stable charge," and  $Z_{p}$  "the most probable charge." Next, read the fraction of the chain from the empirical curve of fractional yields plotted against chain position  $(Z - Z_{p})$ , Fig. 1. For thermal fission Pappas assumed n = 1, i.e., that one neutron was emitted per fragment.

This hypothesis has been extended in the present study to 14.7-Mev fission. It was found that the best fit of the data was obtained with n = 3 for the heavy peak nuclides, and n = 2 for light ones. The evidence is shown in Fig. 1, where the fractional chain yields of the shielded nuclides are compared with Pappas' curve. We have also shown the data on fractional chain yields of various Te and I nuclides measured by Wahl.<sup>17</sup> Nb<sup>96</sup> does not fit the curve for either thermal or 14.7-Mev fission



Fig. 1. Variation of yield with nuclear charge for 14.7-Mev neutron fission of  $U^{235}$ ,  $\forall$ , upper limit. The curve is from Ref. 15. Iodine and tellurium isotopes are from Ref. 17.

for either thermal or 14.7-Mev fission. It is interesting, however, that the equal charge displacement would predict the measured R value.

Using the independent yields from this charge distribution, we return to the calculation of R for those cases where the nuclides had a parent of appreciable half life. For  $Rh^{105}$  and  $Sb^{126}$  the two values for the decay



correction factor for thermal and  $1k_{1}$ ? Here dission differ by less than 1%. For Br<sup>83</sup>, isomerism of the predecessors provents the use of the charge distribution curve in estimating  $k_{1}$  and  $k_{2}$ . Wherefore, the true R value may be 2.5 ± 0.5. The error given in Table III does not include this effect. Fractional chain yields measured by Wahl were used to calculate the decay correction for  $I^{131}$ , and the error includes Wahl's errors for  $k_{1}$  and  $k_{2}$ .

Total chain yields for 14.7-Mev fission of  $U^{235}$ , calculated from the cumulative yields and the equal charge displacement hypothesis, are shown in the fifth column of Table III.

The R values and yields from 14.7-Mev fission of  $U^{238}$  are shown in the last two columns of Table III. These were determined by duplicate analyses of two normal uranium irradiations. In each case the error we quote is greater than the spread of results and is estimated from experience. For  $Cs^{136}$  it was necessary to correct the activity ratio for the contribution from  $U^{235}$  fission. Since the yield of only one shielded nuclide ( $Cs^{136}$ , Table IV) is not necessarily indicative of the charge distribution, no total chain yields have been calculated for 14.7-Mev fission of  $U^{238}$ .

## E. U<sup>235</sup>, U<sup>238</sup>, and Pu<sup>239</sup> Fission Induced by Degraded Fission Spectrum Neutrons

In one series of experiments  $Pu^{239}$  and uranium metal highly enriched in  $U^{235}$  were irradiated in Port 5W, the most energetic spectrum available in the Los Alamos Fast Reactor. Activity ratios relative to  $Mo^{99}$  were measured for  $Pd^{109}$ ,  $Ag^{111}$ ,  $Pd^{112}$ , and  $Cd^{115}$ . In a second series of experiments we attempted to use fission spectrum neutrons. The same nuclides were determined for  $U^{235}$ ,  $U^{235}$ , and  $Pu^{239}$  irradiated in a highly enriched  $U^{235}$  metal capsule. The walls of the capsule were sufficiently thick (0.397 cm) to remove all of the incident thermal neutrons. There was, however, a small contribution from fission neutrons that had been scattered and somewhat degraded in the graphite stringer and the aluminum metal cooling block surrounding the capsule. From experience the errors of the activity ratios should be less than 3%. The yields given in Table V have been calculated using equation (3). For  $U^{235}$  and  $Pu^{239}$  we have assumed that the yield of  $Mo^{99}$ . y', is the same as thermal fission. For  $U^{236}$  we have used the data of Terrell<sup>5</sup> et al.,<sup>5</sup> to estimate the  $Mo^{99}$  yields as 6.32%.

#### TABLE V

Yields of Valley Elements from Fission by Various Neutron Spectra

		Fission	Yield,%	
Type of Fission	Pd109	Ag <sup>111</sup>	Pd112	Cd115
$U^{235}$ , thermal neutrons $U^{235}$ , degraded fission spectrum $U^{235}$ , capsule neutrons	0.024 0.053 0.115	0.0143 <sup>8</sup> 0.022 0.053	0.0127ª 0.020 0.052	0.0104ª 0.017 0.045
U <sup>238</sup> , capsule neutrons	0.252	0.059	0.058	0.036
$Pu^{239}$ , thermal neutrons $Pu^{239}$ , degraded fission spectrum $Pu^{239}$ , capsule neutrons	1.56 1.48 1.60	0.200 0.237	0.120 0.127 0.177	0.034 0.045 0.075

a - from Col. 2, Table III





## F. Np<sup>237</sup> Fission Yields

A sample of an oxide of  $Np^{237}$  was irradiated by degraded fission spectrum neutrons. R. N. Olcott of this Laboratory estimated that the average energy of the neutrons causing fission was 1.1 Mev. There was only one irradiation, and the number of fissions was insufficient to allow separate aliquots for each element. After appropriate exchange steps, barium, strontium, cerium, and europium were determined from one aliquot; tin, antimony, and tellurium, from another; and silver and cadmium from a third. Molybdenum and zirconium were determined from separate aliquots. All analyses were made in duplicate. The cumulative yields shown in Table VI were calculated by equation (3) assuming the Mo<sup>89</sup> yield,  $y_s^i$ , was the same as in U<sup>235</sup> thermal neutron fission. No total chain yields were calculated, since we have no information for the charge distribution.

## TABLE VI

Nuclide	R	Fission Yield, %
Sr <sup>89</sup> Zr <sup>97</sup> Mo <sup>99</sup> Ag <sup>111</sup> Cd <sup>115</sup> Sn <sup>125</sup> Sb <sup>127</sup> Te <sup>132</sup> Ba <sup>140</sup> Ce <sup>144</sup> Eu <sup>156</sup>	0.27 1.04 5.3 3.5 4.7 3.0 1.15 0.80 0.75 8.3	1.3 5.7 6.14 0.076 0.036 0.11 0.34 5.1 5.0 3.7 0.23

## Np<sup>237</sup> Cumulative Fission Yields<sup>2</sup>

a - average energy causing fission estimated to be 1.1 Mev

b - assumed value

#### IV. DISCUSSION

The yields for  $Cd^{115}$ ,  $Sn^{125}$ ,  $Br^{83}$ , and  $Pd^{112}$  for 14.7-Mev fission of  $U^{235}$  given in Table III and Fig. 2 deserve special comment. Cumulative yields for  $Cd^{115}$  and  $Sn^{125}$  are based on the assumption that the ratio of independent yields of isomeric pairs is the same as the ratio of their yields from the decay of predecessors, which is presumed not to change. For the mass 115 chain, the charge distribution curve predicts negligible independent formation at Cd, and Wahl and Bonner<sup>18</sup> have found an equal R value for both isomers for 14.7-Mev fission. Biller<sup>19</sup> has observed that the higher spin state is favored when isomers are formed independently in fission. For the mass 125 chain, analyses were made for the 10-day  $Sn^{125}$ , which is believed to have the higher spin state. Since for the mass 125 chain the charge distribution





hypothesis predicts fractional chain vields at Sn of 0.08 for thermal and 0.29 for 14.7-Mev fission, a preference of higher spin states for independent formation implies a higher of evalue for the 10-day isomer than for the 9.5-minute isomer. The yield calculated assuming equal R values is too high, in accordance with Biller's observation and the charge distribution hypothesis. For the mass 83 chain, similar considerations applied to the 67-second and 25-minute Se predecessors of Br<sup>83</sup> would, through the decay correction, make the R value too high.

We have no explanation for the trend, increasing with energy, for  $Pd^{112}$  to be above the smooth curve. It is also above the curve for capsule fission of  $U^{238}$  (Fig. 3). For the photofission of  $U^{238}$  Richter and Coryell<sup>20</sup> obtained a similar result, whereas Schmitt and Sugarman,<sup>21</sup> who analyzed for Ag<sup>112</sup> rather than  $Pd^{112}$ , did not. Turkevich and Niday<sup>22</sup> assayed for  $Pd^{112}$  in the pile neutron fission of  $Th^{232}$  and obtained a value that could be interpreted as being above their smooth curve.

10.0



Fig. 2. U<sup>235</sup> neutron fission yields. The dashed curve is for thermal neutrons.

Fig. 3. U<sup>238</sup> fission yields with fission spectrum neutrons (- - -Ref. 24), ▲ capsule neutrons, □8.1-Mev neutrons, ● 14.7-Mev neutrons.

The mass-yield curves for 14.7-Mev fission of  $U^{235}$  and  $U^{238}$  are shown in Figs. 2 and 3. In both instances symmetric and very asymmetric fissions are more probable than for thermal or low-energy fission. It is also apparent that the heavy peaks have shifted toward lower mass number, whereas the positions of the light peaks appear unchanged.





For the  $U^{235}$  curve, the sum of masses on opposite sides of the valley and with the same yield is about four less than the mass number of the compound nucleus  $U^{236}$ . The same sum for the vings is 4.5 to 4.7 less than 236. This method has been used to estimate neutron emission associated with certain regions of fission yield curves.<sup>23</sup> Although the exact significance of this procedure is not clear, we wish to compare it with the n from charge distribution, equation (4), where we attribute three neutrons to the heavy peak and two to the light peak. Extra neutron emission from the heavy fragment is a possible explanation for the shift of the heavy peaks toward lighter mass. The apparent insensitivity of the light peak to energy change is not consistent with this point; however, it should be pointed out that some shift in the light peak could be obscured by the rising valley, and that our choice of n = 2 for the light peak is based on only one independent yield, that of Br<sup>82</sup>.

It has been noted that the heavy peak is insensitive to fissioning nuclide for low excitation energies.<sup>24</sup> A comparison of peak yields from 14.7-Mev fission in Table V shows that the heavy peak yields are nearly the same for  $U^{235}$  and  $U^{238}$ , whereas the light peak reflects the difference in the fissioning nuclides as in lower energy fission.

Wahl has found that the fine structure around mass 134 is less in 14.7-Mev fission of  $U^{235}$  than in thermal fission. We have insufficient data to draw any conclusion about the complementary spike around mass 100. There appears to be some fine structure near mass 90.

Newton<sup>25</sup> has made yield measurements for 37.5-Mev alpha-particle induced fission of  $Th^{232}$ . The compound nucleus, with 33-Mev excitation, has the same mass number and excitation as would result from 28-Mev neutrons on U<sup>235</sup>. Newton's results are qualitatively in the direction that our results indicate for 28-Mev neutron fission of U<sup>235</sup>. However, it should be pointed out that a fission-yield curve is an average over several states of angular momentum, and that there is no reason to believe that their relative abundances will necessarily be the same for alpha-particle irradiations of  $Th^{232}$  as for neutron irradiations of U<sup>235</sup>.

Yields of the valley nuclides  $Ag^{111}$  and  $Cd^{115}$ , as well as the shielded nuclide  $Cs^{136}$ , rise somewhat less than exponentially with neutron energy for  $U^{235}$  fission (Fig. 4). There is no evidence that the change of these yields with neutron energy is not smooth. There are not enough points, however, to determine if there is a break in the curve after the compound nucleus has sufficient energy to emit a neutron before fissioning. For  $U^{238}$ , the yields for near-symmetric fission are less than those from  $U^{235}$  for the same neutron energy, corresponding to the lower neutron binding energy.

The yields of two peak nuclides from  $U^{235}$  fission are shown as a function of neutron energy in Fig. 5. The yield of Mo<sup>99</sup>, a light peak nuclide, does not decrease with increasing energy until the valley yields become appreciable, as is to be expected from the normalization requirement. The yield of Ce<sup>143</sup>, on the other hand, decreases much more rapidly than that of Mo<sup>99</sup>, indicating that the shift of the heavy peak to lower masses begins at relatively low energies. Schmitt and Sugarman<sup>21</sup> found decreasing yields for Ba<sup>140</sup> from the photofission of U<sup>238</sup> as the maximum gamma-ray energy was increased from 7 to





21 Mev. This also may be interpreted.as a shift of the heavy peak.

The fission product yields from the fission of Np<sup>237</sup> with degraded fission-spectrum neutrons are compared with the mass-yield curves from thermal fission of  $Pu^{239}$  (Ref. 6) and fission spectrum fission of  $U^{238}$ (Ref. 24) in Fig. 6. The approximate energies of excitation for the three compound nuclei are 6.5, 6.4, and 7.6 Mev, respectively. The masses and charges are nearly the same, and fission yields are quite similar. The heavy peak from Np<sup>237</sup> fission seems to be broader at the base than the other two.

The yields of the valley nuclides from fission of  $U^{235}$  (Fig. 2),  $U^{238}$  (Fig. 3), and  $Pu^{239}$  (Fig. 7) by various degraded neutron spectra represent the products of fission yields by relative fission cross sections averaged over the spectra. These yields show the beginning of the rise in the valley with increasing energy.

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