Fission Yields of Natural Uranium with Deuterons of 5, 10, and 13.6 Mev: Deuteron Capture and Competition with Stripping*†

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Radiochemical determinations of yields in the fission of natural uranium with 5-, 10-, and 13.6-Mev deuterons have been made for 35 mass chains from mass numbers 72 to 159. The total fission cross sections at the three energies were found to be 3.5, 86, and 430 mb, respectively, by integration of the mass-distribution curve. These cross sections include contributions from the fast-neutron background, which is negligible at the two higher energies but which constitutes nearly all of the observed fission at 5 Mev. Although in gross features the yield-mass curves resemble those for other fissile nuclei, some important differences are apparent. The highly asymmetric yields change nearly as rapidly with energy as those of the valley; the deuteron yield curves are the broadest that have been observed at these energies.

From analysis of yield measurements for nearly complementary masses it has been found that fewer neutrons are emitted in modes leading to highly asymmetric products as compared to the most probable modes. The average number of neutrons per fission is found to be 3.5, 5.0, and 5.0 with deuterons of 5, 10, and 13.6 Mev.

The measured valley-to-peak yield ratios at 10 and 13.6 Mev are smaller than would be expected from deuteron capture. It is concluded that at these energies a small but significant contribution is made by fission following a (d,p) or (d,n) stripping reaction. From known (d,p), (d,n), and fission cross sections, the proton-energy spectrum in (d,p) stripping on heavy nuclei, and estimated branching ratios for modes of decay of a heavy excited nucleus, it is calculated that the fraction of fission events following deuteron capture is 0.8 ± 0.2 with 13.6-Mev deuterons and 0.75 ± 0.25 with 20-Mev deuterons, the remainder following a (d,p) or (d,n) stripping reaction.

I. INTRODUCTION

THE study of yields in the deuteron-induced fission of uranium can be used as an indication of the mode of interaction of the deuteron with the target nucleus. The radiochemical determination of the mass distribution of fission products is capable of high resolution over a wide range of yields. The yield curves are useful in a variety of work on the fission process. The valley-to-peak (v/p) ratio can be related to the excitation energy of the fissioning nucleus, and information can be obtained about ν, the average number of neutrons emitted in a given fission mode, as a function of the mass ratio of the fission products. In a related paper,[1] data are presented on the distribution of nuclear charge in the deuteron fission of U^{238} and Th^{232}.

Until recently, the deuteron has been used infrequently as the bombarding particle in fission studies at low energies[2–5] (<25 Mev) although a number of high-energy experiments[6,7] have been reported. This is due in part to the complications that arise in interpreting results. Since an important interaction of a deuteron with a heavy target nucleus is stripping,[8] uncertain energy deposition in the residual nucleus preceding fission is possible. From fission-fragment ranges, however, Douthett and Templeton[2] in 1954 concluded that their results in the 18-Mev deuteron fission of natural uranium were consistent with fission following deuteron capture. In more recent work[3,4] in which mass distributions were measured in deuteron fission of U^{233}, Pu^{239}, and Pu^{240} the results are less readily understood from this point of view.

The fission asymmetry or v/p ratio has been interpreted[9–12] as a measure of the excitation energy of the

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fissioning nucleus and is largely independent of the exciting particle or the heavy fissionable mass. It was felt that if the ν/p ratio could be measured with deuterons of several energies, at the Coulomb barrier and below, and if cross sections for the (d,p), (d,n), and fission reactions were known or could be measured, an estimate could be made of the fraction of fission events that follow capture and stripping.

Another motivation for this work was to measure ν, the average number of neutrons emitted in a given fission mode, as a function of the fission mode. Smooth mass distributions are usually obtained by reflecting a measured yield about an axis of symmetry, which is a measure of ν. Tewes and James[13] and Schmitt and Sugarman[14] found that it was not possible to use a constant symmetry axis and still obtain a reasonable fit to all of their data. The results could be explained by assuming ν to be smaller for highly asymmetric fission modes than for more probable modes. In this work we have measured a number of yields of approximately complementary masses to obtain evidence of appreciable change in ν as a function of the mass ratio of products formed.

This paper gives details on the radiochemical yields determined by the first three authors at Clark University; the yields obtained by the fourth author at M.I.T. are taken from the following paper.[1]

II. EXPERIMENTAL PROCEDURE

A. Irradiations

All deuteron irradiations were carried out in the external beam of the Massachusetts Institute of Technology cyclotron. Uranium foil[15] 0.7 mil thick (27.8 mg/cm^2) of natural isotopic composition was the target material in all irradiations except two early experiments at 13.6 Mev in which natural UO_3($\sim$50 mg/cm^2) was used. The target was wrapped in an Al-foil envelope (8.5 mg/cm^2) and was secured to the water-cooled target holder by 13.7 mg/cm^2 of Dural foil. The deuteron beam, which has a maximum energy of 15.2 Mev inside the cyclotron, traversed a 10.3-mg/cm^2 Dural window and a few centimeters of He at 1 atmosphere before impinging on the target assembly. The calculated deuteron energy[16] on striking the U foil is 13.6 Mev and the foil is calculated to be 0.5 Mev thick. The UO_3 targets were of the order of 1.5 Mev thick. The beam energy was computed to be 9.9 and 5.2 Mev with 106 and 169 mg/cm^2 total Al and Dural absorber. Background runs were made with 308 mg/cm^2 Al and Dural absorber. The range of a 15.2-Mev deuteron in Al or Dural was taken to be 202 mg/cm^2. The U foil is calculated to be 0.8 Mev and 1.2 Mev thick at 9.9 and 5.2 Mev, respectively. Kafalas and Irvine[17] have measured energy straggling in Cr absorbers. Assuming that these data are equally applicable to the Al and Dural absorbers, we calculate the energy bands to be 13.6$\pm$0.6 Mev, ($9.9_{-0.7}{}^{+0.9}$) Mev, and ($5.2_{-1.1}{}^{+1.3}$) Mev. For convenience we refer to the latter two energies as 10 Mev and 5 Mev.

Irradiations were for 10 to 60 min at beam intensities of about 20 microamperes. In most runs the target area was smaller than the beam area. In these cases only relative yields of fission products were determined. For the cross-section measurements, the target area was substantially larger than the beam area and accurately positioned to capture the entire beam. The beam intensity was monitored by a thermocouple that had been calibrated against a Faraday cup.[18]

B. Chemical Separations

After irradiation, the target foil and Al envelope were dissolved in 4M HCl and conc. HNO_3. Dilute H_2O_2 was added to convert all the U in solution to the hexavalent state. The solution was diluted with HCl to make a stock solution from which aliquots were withdrawn for various chemical operations. The chemical methods are briefly described in the Appendix.

C. Counting

All counting was done on a set of three intercalibrated thin-wall, cylindrical, flow Geiger counters, $\frac{3}{4}$ in. in diameter and 5 in. long, of a type that has been described.[19] The counting gas was 98.7% He and 1.3% isobutane at one atmosphere. For high counting rates the usual 2 in. of Pb shielding was used. Anticoincidence shielding in addition to massive Fe shielding was used to reduce background for samples of very low activity. Background rates were about 35 counts per minute inside 2 in. of Pb and about 2 counts/min with anticoincidence inside 6 in. of Fe. The plateau slopes of the counters were about 2% per 100 volts over a 200-volt region.

Samples that had been obtained on thin filter paper ($\sim$8 mg/cm^2) were mounted on half-cylinders of Lucite (420 mg/cm^2) that fit snugly around the counters in close cylindrical geometry. All counting corrections applied were those developed especially[20,21] for these conditions. Nuclides were identified within a given chemical fraction by half-period and absorption characteristics. In no case were there significant deviations from literature values.[22]

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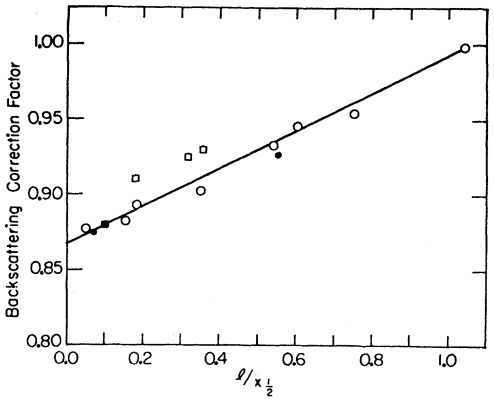

FIG. 1. The factor by which an observed activity must be multiplied in order to remove the backscattering contribution from thick Lucite is plotted as a function of the reduced sample thickness, $l/x_{\frac{1}{2}}$, where l is the sample thickness in mg/cm^2 and $x_{\frac{1}{2}}$ is the absorption half-thickness in the same units. o Sr^{89} (E_β=1.463 Mev), ● As^{77} (E_β=0.700 Mev), □ Pr^{143} (E_β=0.932 Mev), ■ Br^{83} (E_β=0.940 Mev).

Suttle and Libby[20] have shown that in cylindrical geometry with thick samples, truly exponential absorption of β particles occurs, and Libby[21] has found that the half-thickness in mg/cm^2 of Al($x_{\frac{1}{2}}^{Al}$) for a given β particle is related to the maximum β energy E_β in Mev by the relation:

$$x_{\frac{1}{2}}^{Al}=38E_\beta^{\frac{3}{2}}. \quad (1)$$

In other absorbers of weight-average atomic weight M, the absorption half-thickness in mg/cm^2, $x_{\frac{1}{2}}^{M}$, is given[21] as

$$x_{\frac{1}{2}}^{M}=x_{\frac{1}{2}}^{Al}\left(\frac{127}{100+M}\right). \quad (2)$$

Libby's formulas[21] apply to samples in which the thickness is greater than one absorption half-thickness. Under these conditions the contribution from backscattered radiation is small. For samples thinner than this, backscattered β particles contribute appreciably and an experimental absorption curve for a single β will show curvature due to the softer scattered component. We have measured as a function of sample thickness the contribution of backscattered β rays in Sr^{89} and some other monoenergetic β emitters by measuring the deviation from linearity in the absorption curve. From these measurements a backscattering correction factor can be obtained. Figure 1 shows the correction factor as a function of reduced sample thickness $l/x_{\frac{1}{2}}$ (sample thickness divided by absorption half-thickness). Counting rates were corrected for this effect assuming that the same correction was applicable to all β energies. This problem arose since many chemical fractions contained several β components due to different nuclides or complex decay schemes and it was not convenient to make the sample thick with respect to all energies. While the Libby method[20,21] was developed for simple decay schemes (one β only), it has been used successfully also for complex decay schemes[23] in which the abundances were known.

Corrections for external absorption by counter wall (2.70 mg/cm^2), air (0.5 mg/cm^2), and sample cover (0.9 mg/cm^2 of Mylar), as well as self-absorption in the sample, were made using the Libby method. Experimental half-thicknesses were used when they had been measured; otherwise Eqs. (1) and (2) were used to calculate a half-thickness.

Observed activities were corrected also for saturation, counter dead-time (0.18% per thousand counts/min), and geometry factor. Total fission yields were then computed for the mass number by applying corrections for parent half-period and for estimated fractional chain yield[1] of the species measured. Finally, formation cross sections were calculated from known beam intensities and the number of U atoms in the target.

III. RESULTS

The formation cross sections for each mass chain measured are listed in Table I and the distribution for 35 mass points is shown in Fig. 2. By integrating each mass distribution, we obtain the total fission cross section to be 3.5, 86, and 430 mb at 5, 10, and 13.6 Mev, respectively.

Each datum in Table I is the mean of at least two determinations (three or four in the 13.6-Mev case) except in the case of those shown without error. The error indicated is simply the mean deviation of replicate experiments and is not intended to represent the errors of the experiment. On an absolute basis

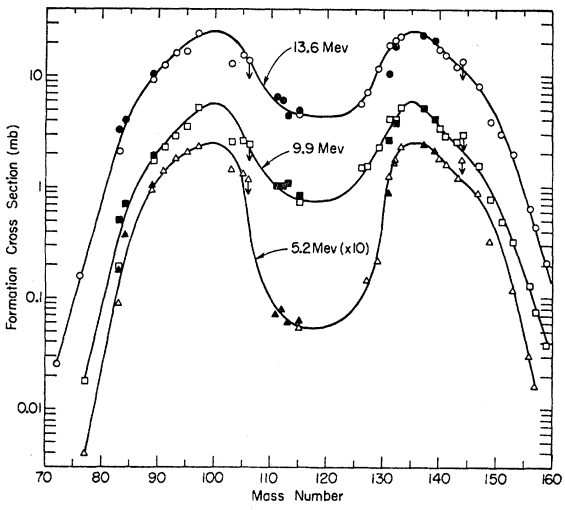

FIG. 2. Yield-mass distribution in deuteron-induced fission of natural U with deuterons of 5, 10, and 13.6 Mev. Open symbols are data obtained at Clark University; closed symbols are M.I.T. data.[1] The yields of Ru^{106} and Ce^{144} are considered to be only upper limits. The yield of Ru^{103} has been neglected in drawing the smooth curve. The 129-chain yield has been taken as twice the Sb^{129} yield (see Table I, footnote *m*).

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TABLE I. Chain yields of products formed in the fission of natural uranium with deuterons of 5, 10, and 13.6 Mev.

Mass No.	Nuclide isolated and measured[a]	5 Mev (μb)	10 Mev (mb)	13.6 Mev (mb)	D_∞(bkgd)/D_∞(5Mev)
72	49-hr Zn	...	...	0.026±0.005	
77	38-hr As[b]	0.4	0.017	0.16±0.04	
83	2.40-hr Br[c]	9	0.19±0.04	2.2±0.1	
89	53-day Sr	94±4	1.76±0.06	9.3±0.4	0.84
91	58-day Y	143±7	2.3±0.1	12.2±0.8	0.93
93	10-hr Y	180	2.9	16.4±0.8	
95	65-day Zr[d]	205±15	3.5±0.2	16.8±0.7	
97	17-hr Zr →72-min Nb[e]	233±16	5.2±0.2	24.1±0.2	
103	43-day Ru	141±2	2.6	13±2	
105	4.5-hr Ru[f]	134±3	2.6	15.7±1.5	
106	1-yr Ru →30-sec Rh[g]	120±20[l]	2.4[l]	14.0±0.7[l]	
115	54-hr Cd	4.8±0.5	0.73±0.03	4.2±0.2	1.0
	43-day Cd	0.4±0.2[l]	0.046±0.004	0.30±0.02	1.0
126	9-hr Sb	...	1.2±0.1	3.9	
	28-day Sb[h]	...	0.35±0.02	1.7	
127	93-hr Sb →9.3-hr Te[e]	15±1	1.56±0.06	7.3	
129	4.6-hr Sb →72-min Te[e]	11[m]	1.15±0.10[m]	5.9[m]	
131	8.14-day I	128±1	4.2±0.1	19±2	
132	2.4-hr I[i]	172±3	4.2±0.2	20.6±1.1	
133	20.5-hr I	237±9	5.3±0.2	23	
140	12.80-day Ba	181±9	3.4±0.2	17.5±0.9	0.89
141	33.1-day Ce	160±16	2.9±0.1	15.6±1.6	0.83
143	13.7-day Pr[j]	120±7	2.6±0.1	12.1±0.3	0.91
144	280-day Ce →17-min Pr[k]	183±4[l]	3.0±0.3[l]	14±2[l]	0.90
147	11.3-day Nd	89±4	1.6±0.1	8.7±0.3	
149	54-hr Pm	33±2	0.8	3.8±0.6	
151	27.5-hr Pm	...	0.5	3.0±0.2	
153	47-hr Sm	12.1±0.1	0.33±0.01	2.0±0.1	0.95
156	15.4-day Eu	3.0±0.5	0.132±0.002	0.66±0.08	
157	15.4-hr Eu	1.6	0.076±0.002	0.45±0.06	
159	18.0-hr Gd	...	0.039±0.001	0.21±0.01	
	Total fission cross section	3500 μb	86 mb	430mb	Mean ratio: bkgd/5 Mev 0.92

[a] Observed half-lives are given. See reference 22 for literature values.
[b] Yield of As^{77} calculated assuming branching ratio of 12-hr Ge^{77} to 59-sec Ge^{77} is 0.52 as given by N. Sugarman, Phys. Rev. **89**, 570 (1953).
[c] Yield of Br^{83} calculated under the assumption (reference 1) that 45% of the 83 chain passes through 26-min Se^{83}.
[d] Counted through 10.1 mg/cm² of Al to minimize Nb^{95} contribution.
[e] Counted equilibrium mixture.
[f] Counted through 26.2 mg/cm² of Al to absorb conversion electrons of Rh^{105m}.
[g] Counted equilibrium mixture through 26.2 mg/cm² of Al to absorb radiations of Ru^{103} and Ru^{106}.
[h] Mass assignment not certain, not observed in thermal fission but found in 14-Mev neutron fission of U^{235}, J. Barnes and M. Freedman, Phys. Rev. **84**, 365 (1952); sum of the yields of the 9-hr and 28-day species plotted as point for mass 126 in Fig. 2.
[i] Measured I^{132} in equilibrium with 77-hr Te^{132}; yield is that of parent.
[j] Measured Pr^{143} in separated cerium fraction in which 33-hr Ce^{143} had decayed; yield is that of parent.
[k] Counted equilibrium mixture through 70 mg/cm² of Al to absorb radiations of Ce^{141} and Ce^{144}.
[l] Upper limit, contribution of long-lived impurities possible.
[m] According to Pappas (reference 52), in thermal-neutron fission of U^{235} the mass-129 chain yield should be taken as twice the Sb^{129} yield. The yields in this table are for Sb^{129}; in Fig. 2 the chain yield has been plotted.

the cross sections for producing each nuclide are probably no better than ±25%. The largest fraction of this error is attributed to uncertainties in the beam intensity. The relative yields are believed to be good to ±10% except in the case of certain nuclides obtained with very low counting rates (Zn^{72}, As^{77}, Ru^{103}, Ru^{106}, Cd^{115m}, Ce^{144}) and those for which chemical yield data were uncertain (Pm^{149}, Pm^{151}). Thus, the absolute fission cross sections obtained by integration of the smooth curves are probably not better than ±30%.

The results of the background runs (more absorber than the range of the deuterons) are also given in Table I. In column 6 are given the ratios of saturation activities D_∞ for the nuclides measured in both the background and 5-Mev irradiations. Presumably the fission observed in the background runs was due to fast neutrons. The constancy of the ratios averaging 0.92 indicates that nearly all the observed fission at 5 Mev can be attributed to fast neutrons. The true deuteron-fission cross section at 5 Mev is probably less than 0.5 mb.

From a consideration of the magnitudes of the fission cross sections, it is clear that neutron fission could not have contributed significantly to the observations made at 10 or 13.6 Mev. If the mean fast-neutron fission cross section of U^{238} is taken to be 0.5 barn,[24] it is calculated from the saturation activities of the background run that the neutron intensity on the entire target with 308 mg/cm² Al absorber was 2.3×10^{13}/min or about 3×10^{-3} of the deuteron intensity. The assumption has been made that the percent fission yields of the various nuclides observed in the background irradiation are the same as those in the 5-Mev irradiations.

The smooth-curve mass distributions in Fig. 2 are based on directly measured experimental points. No assumption was made about the symmetry of either peak or about a symmetry line between the peaks. Some general observations can be made in regard to the distributions shown.

1. The general shape of the curves in the neighborhood of the peaks is the same at all energies.

2. In all three mass distributions it appears that the heavy peak is somewhat narrower than the light peak although the difference is probably within experimental error. The same effect, due to a low shoulder on the heavy side of the heavy peak, has been observed[25] in the thermal-neutron fission of Pu^{239} and a similar effect has been reported by Petruska *et al.*[26] for U^{235} fission.

3. The curves at 10 Mev and 13.6 Mev are closely superposable except for an increased relative yield for the lightest nuclides (As^{77},Br^{83}) at 13.6 Mev. The close similarity is not understood.

4. The wings of the curves spread much more than those, for example, for the thermal fission[27] of U^{235}. Far out on the wings the logarithmic values of the yields are linear with mass number. The limiting slopes $d\log y/dA$ for the linear region as calculated from Fig. 2 and from other work in the literature are given in

[24] D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).
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[26] Petruska, Thode, and Tomlinson, Can. J. Phys. **33**, 693 (1955).
[27] E. P. Steinberg and L. E. Glendenin, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 7, p. 3, paper No. P/615.

TABLE II. Limiting slopes $d\log y/dA$ on light and heavy wings.

Type of fission	Slopes $d\log y/dA$ Light wing	Heavy wing	Reference
U^{238} (5-Mev d,F)	+0.26	−0.22	This work
U^{238} (10-Mev d,F)	+0.20	−0.15	This work
U^{238} (13.6-Mev d,F)	+0.18	−0.15	This work
U^{235} (thermal n,F)	+0.53	−0.43	27
Th^{232} (6–11 Mev n,F)	+0.32	···	10
Th^{232} (8.0-Mev p,F)	+0.30	···	13
Th^{232} (9.3-Mev p,F)	+0.28	···	13
Th^{232} (13.3-Mev p,F)	+0.29	···	13
Th^{232} (17.8-Mev p,F)	+0.28	···	13
Th^{232} (21.1-Mev p,F)	+0.25	···	13
U^{238} (22-Mev X,F)[a]	+0.17	···	11
U^{238} (48-Mev X,F)[a]	+0.21	···	14
Pu^{239} (thermal n,F)	+0.29	···	25

[a] Bremsstrahlung fission (X,F).

Table II. The 13.6-Mev deuteron curve is the broadest one (lowest absolute slope) for which yield data are available out so far ($A<83$, $A>153$) with the possible exception of the photofission of U^{238} with 22-Mev bremsstrahlung.[11] The slope computed from data reported[14] for 48 Mev, however, is higher than this.

5. The v/p ratio is a sensitive indicator of excitation energy of the fissioning nucleus (see Sec. B under Discussion below). The wings appear to be almost as sensitive to energy change. In Fig. 3 the ratio of formation cross sections 13.6 Mev to 5 Mev is plotted logarithmically as a function of mass number. The solid curve is the ratio of smooth curves taken from Fig. 2. The open circles represent points calculated from the Clark experimental data of Table I. Closed circles are similar points from M.I.T. data.[1] Ratios measured at both Clark and M.I.T. are indicated by open circles when agreement is good. The detailed shape of the curve is not known with certainty. The ratio for both the very light and very heavy masses is large although none is as much as one-half the ratio at 115. In the very asymmetric regions ($A<83$, $A>153$) the ratio curve is linear with A, increasing with increasing asymmetry. The limiting outer slopes $d\log(\sigma_{13.6}/\sigma_5)/dA$ for light masses (−0.065) and heavy masses (+0.063) have the same magnitude within experimental error, implying that light- and heavy-mass yields change in the same way as the deuteron energy is changed. The comparison is not altogether valid since the fissioning nuclei with 5- and 13.6-Mev deuterons are not the same (239 and 240, respectively; see Sec. B, Discussion). However, on the average 1.5±0.7 fewer neutrons are emitted at the lower energy (see Sec. A, Discussion) which should compensate for the difference in mass. There are insufficient data available in the literature to compare these results with those for other fissile nuclei or other bombarding particles.

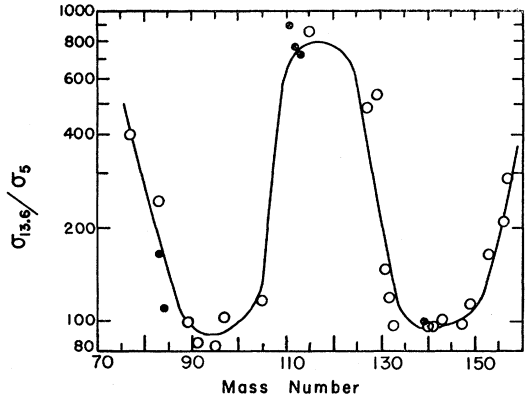

FIG. 3. Ratio of formation cross sections, 13.6 Mev to 5 Mev, as a function of mass number. The solid curve is based on the smooth curves of Fig. 2. Open circles are calculated from the data of Table I. Closed circles are similar values for M.I.T. data.[1] Ratios for masses measured at both Clark and M.I.T. are indicated by open circles when agreement is good.

IV. DISCUSSION

A. Average Number of Neutrons Emitted as a Function of Mass Ratio of the Products

Enough data have been obtained for a reasonably precise determination of ν, the average number of neutrons emitted in a given fission mode. Determinations can be made by adding the masses of complementary products (i.e., those of the same yield) and subtracting the sum from the mass of the fissioning nucleus. The ν values obtained are given in Table III. The error in ν has been estimated in the following way: If an error in the yield of a light product is assigned, this can be converted to an error in mass by assuming the smooth curves of Fig. 2 to be the best fit to the data. The error in the mass of the heavy complementary product is obtained by determining the range of heavy masses corresponding to the range in yield of the light mass. The error in ν is taken to be the root-mean-square error in the masses of light and heavy products. The error in relative yields has been assumed to be ±10% for masses 89 and 105 and ±20% for masses 72, 77, and 83. In general either the yield of the complementary heavy mass was measured or that of an adjacent mass. In only three cases, noted in the table, were ν values obtained by extrapolation. It has been assumed that the mass of the fissioning nucleus was 239, 240, and 240 at 5, 10, and 13.6 Mev. From the discussion in B

TABLE III. Number of neutrons emitted, ν, in a given fission mode[a] for deuteron energies of 5, 10, and 13.6 Mev.

Fission mode (designated by mass of light fragment)	ν 5 Mev	10 Mev	13.6 Mev
72	···	···	2.9±0.8[b]
77	2.0±0.6[b]	2.2±0.7[b]	3.3±0.7
83	3.1±0.8	4.0±1.5	5.2±0.9
89	4.1±1.0	5.0±0.7	5.2±1.0
105	3.0±0.5	5.0±0.7	4.7±0.8

[a] The fissioning nucleus has been taken to be mass 239, 240, and 240 for deuteron energies of 5, 10, and 13.6 Mev, respectively.
[b] Obtained by extrapolation of smooth mass-distribution curve.

and C below, it is clear that at least part of the fission observed at 13.6 Mev (and hence at 10 Mev) follows a stripping reaction. The average mass of the fissioning nucleus then must be less than 240 at the two higher energies so that the corresponding ν values are somewhat too large as given in Table III. The magnitude of the systematic error is difficult to assess but it is probably about 0.2.

The trend is that neutron emission is less probable when highly asymmetric products are formed (as has been suggested[13,14] from experimental data for other types of fission).[28] This indicates that less energy (deformation and internal excitation) is available for neutron boil-off in far asymmetric fission as has been predicted in the statistical theory of fission.[29] Another important test lies in the determination of ν in the region of symmetric fission; this remains to be investigated. Since ν is relatively constant in the region of high yields, $\bar{\nu}$ (the average number of neutrons per fission) is taken to be the average of the ν values for masses 89 and 105, namely 3.5, 5.0, and 5.0 for the three energies, each probably reliable to ± 0.5 neutron. The present experiments cannot distinguish between prefission neutron emission and the neutrons that accompany fission, nor is it possible to determine the individual contributions of light and heavy fragments to ν. It is of course necessary to estimate $\bar{\nu}$ in order to calculate the correction for fractional chain yield.[1] If the valley-to-peak ratio and curve A of Fig. 4 are taken as a measure of excitation energy, $\bar{\nu}$ increases by 1.5 ± 0.7 units with a 7.5-Mev increase in excitation energy. This is consistent with the $d\bar{\nu}/dE$ value of 0.15 calculated by Leachman[30] for low-energy neutron fission.

B. Fission Asymmetry, v/p Ratios

There is substantial empirical evidence [3,9–12] that the valley-to-peak (v/p) yield ratio in asymmetric fission is a measure primarily of the excitation energy of the fissioning nucleus and is fairly independent of the heavy fissile nucleus involved. In Fig. 4 the v/p ratios that have been reported in the literature are plotted as a function of the excitation energy of the fissioning nucleus assuming the bombarding particle is captured in each case. The excitation energies were computed from mass data.[31] No single line can be drawn through all the points in the region of excitation greater than 15 Mev. In this region the data seem to fall into three groups. Group A includes data for which the fissioning nuclei are on the neutron-rich side of the valley of stability. Group B includes fissioning nuclei which are near stability or are neutron-deficient. Group C includes data for the proton-induced fission of U^{235} and U^{238} which seem to constitute a separate class. The coverage of published data is fairly complete. The dashed lines A, B, and C are arbitrary lines that with few exceptions represent the three groups within experimental error (usually $\pm 20\%$ in yield and ± 0.5 Mev in energy).

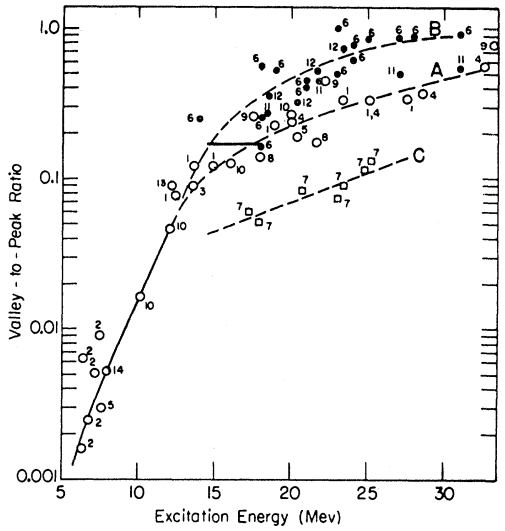

FIG. 4. Valley-to-peak ratio is plotted against excitation energy in Mev as calculated from nuclear masses[31] assuming capture of the fission-inducing particle. ○ fissioning nuclei on the neutron-rich side of the valley of stability; ● fissioning nuclei which are near stability or are neutron-deficient; □ proton-induced fission of U^{235} and U^{238}. The dashed lines, A, B, and C are arbitrary lines that with few exceptions represent the three groups within experimental error (usually $\pm 20\%$ in yield and ± 0.5 Mev in energy). The numbers refer to the following work: 1, reference 13; 2, reference 27; 3, reference 10; 4, A. S. Newton, Phys. Rev. **75**, 17 (1949); 5, R. W. Spence, U. S. Atomic Energy Commission Report AECU-649, 1949 (unpublished); 6, reference 3; 7, reference 12; 8, this work; 9, reference 38; 10, reference 11; 11, reference 33; 12, reference 4; 13, H. C. Richter and C. D. Coryell, Phys. Rev. **95**, 1550 (1954); 14, J. R. Cuninghame, J. Inorg. Nuc. Chem. **4**, 1 (1957).

The grouping can perhaps be explained by prefission events. Compare two compound nuclei excited to the same energy (~15–30 Mev), one of which is neutron-rich and the other neutron-deficient. If neutron emission can compete more strongly with fission[32,33] in the neutron-rich nucleus, it is possible that fission occurs from a state of lower average excitation energy for a neutron-rich nucleus as compared to that of a neutron-deficient nucleus. This would lead to smaller v/p ratios for neutron-rich nuclei of the same apparent excitation energy.

Taking curves A and B to represent extreme ranges and our v/p ratios of 0.021, 0.14, and 0.17 with deuterons of 5, 10, and 13.6 Mev, we find the expected excitation energies of the fissioning nuclei to be 10, 14–16, and 14.5–18 Mev, respectively. The range for the 13.6-Mev

[28] Experimental data obtained by neutron counting from spontaneous fission of Cf^{252} [Hicks, Isle, Pyle, Choppin, and Harvey, Phys. Rev. **105**, 1507 (1957)] do not show this trend, but ν is small in this case and the effect would be difficult to observe.

[29] P. Fong, Phys. Rev. **102**, 434 (1956); **89**, 332 (1953).

[30] R. B. Leachman, Phys. Rev. **101**, 1005 (1956).

[31] Glass, Thompson, and Seaborg, J. Inorg. Nuc. Chem. **1**, 1 (1955).

[32] E. J. Winhold and I. Halpern, Phys. Rev. **103**, 990 (1956).

[33] Glass, Carr, Cobble, and Seaborg, Phys. Rev. **104**, 434 (1956).

case is shown as a horizontal line in Fig. 4. Mass calculations[31] show that deuteron capture at 10 and 13.6 Mev leads to excitation energies of 18 and 21.6 Mev. The discrepancy can be explained if it is assumed that some of the fission events followed a (d,p) or (d,n) stripping reaction.

In a study of the energy spectrum of the protons emitted in the stripping reaction of 14-Mev deuterons on natural U and other heavy elements, Aschenbrenner[34] found that, on the average, the excitation energy left in the residual nucleus was about 10 Mev. The v/p ratio with 10-Mev excitation energy from Fig. 4 is about 0.021. If 20% of the fissions at 13.6 Mev occurred with 10-Mev excitation and 80% with 21.6-Mev excitation, the v/p ratio would be $0.2(0.021)+0.8(0.25)$, which within experimental error is the same ratio we observe at 13.6 Mev.

On the other hand, it is clear that the contribution of fission following stripping could not be large. It might be argued that the fission we observe follows a stripping reaction that leaves the residual nucleus excited to, say, 14 Mev. Aschenbrenner's results[34] indicate that in heavy nuclei the number of low-energy protons emitted, which would correspond to large excitation of the residual nucleus, is relatively small.[35] The magnitude of the observed fission cross section at 13.6 Mev (430 mb) seems very much too large to have followed only a small fraction of stripping reactions. We conclude that at 13.6 Mev, fission predominantly follows deuteron capture, although some contribution from fission following stripping is necessary to account for our v/p ratios. In an accompanying paper[1] substantially the same conclusion is drawn independently from charge-distribution data. In Sec. *C* below, some quantitative estimates are made of the contributions of capture and stripping preceding fission.

Although this argument is based on proton spectra, the same notions should be applicable to neutron spectra and fission following a (d,n) reaction.

Because of the general similarity of the mass distributions and the v/p ratios, we feel that 10-Mev and 13.6-Mev fission are nearly the same although the part played by stripping may be somewhat different in the two cases. It is of interest to note that the calculated[36] formation cross sections of the compound nucleus Np^{240} are 100 and 670 mb with 10- and 13.6-Mev deuterons respectively ($r_0=1.5\times10^{-13}$ cm), in about the same ratio as the total fission cross sections of Table I.

With 5-Mev deuterons the calculated[36] formation cross section is less than 10^{-3} mb. Our results indicate that nearly all the fission we observe can be attributed to neutron background. The v/p ratio is consistent with fission induced by neutrons of about 5 Mev. As previously stated the contribution of fission following (d,p) stripping must be small.

Jones *et al.*[12] have argued that if the logarithm of the v/p ratios is plotted against $(E^*-5)^{-\frac{1}{2}}$, where E^* is the excitation energy (in Mev) of the fissioning nucleus, a straight line results. Since the quantity $(E^*-5)^{-\frac{1}{2}}$ has the dimensions of a reciprocal temperature, it is suggested that the slope of the line is a measure of the difference in energy required for symmetric *versus* asymmetric fission. When v/p ratios recently obtained in this work and elsewhere are plotted on such a diagram, we find that no straight line can be fitted. In fact the data shown in Fig. 4 are more consistent with a straight line when the abscissa is taken to be $(E^*)^{-\frac{1}{2}}$ (as in U^{238} photofission[11]) although there are large random deviations.

C. Analysis of Cross Sections for Deuteron Reactions

As indicated in the discussion above, the magnitude of the v/p ratio strongly suggests that most of the fission events follow the formation of a compound nucleus in which the whole deuteron is captured. However, it is well known that deuteron bombardment often results in excited nuclei in which only the proton or neutron has been captured, and the other particle is rejected by the residual nucleus[8] (stripping). Experimental studies of proton spectra and proton angular distributions from the (d,p) reaction on heavy nuclei (forward peak in angular distribution of low-energy protons) show that some interactions of the stripping type result in highly excited nuclei.[34] It is the purpose of this section to make a quantitative estimate of the contribution to fission from the various possible excited nuclei.

Three different excited nuclei can be formed from a target T: (1) $[T+n]^*$ resulting from (d,p) stripping, (2) $[T+p]^*$ resulting from (d,n) stripping, and (3) $[T+d]^*$ resulting from deuteron capture. The excited nuclei are assumed to have the properties of the compound-nucleus model, that is, the energy is distributed throughout the system and there is no memory of the mode of formation. Experimental cross sections for deuteron reactions and the spectra of protons and neutrons resulting from stripping processes furnish enough information to calculate cross sections for various modes of excited-nucleus formation and the probability of resulting decay processes. The existence of direct interactions such as (d,t) and (d,He^3) does not invalidate this treatment provided that few fission events follow these processes.

Let σ_1, σ_2, and σ_3 be the cross sections for excited-nucleus formation by (d,p) and (d,n) stripping and d capture, respectively; ${}^iq_n$, ${}^iq_{2n}$, etc. be the probability that the ith excited nucleus emits one neutron, two neutrons, etc., possibly followed by γ emission; ${}^iq_\gamma$ be the probability that only γ emission occurs; and ${}^iq_F$

[34] F. A. Aschenbrenner, Phys. Rev. **98**, 657 (1955).

[35] For example, the cross section (reference 34) for total proton production [(d,p) reactions and electric breakup] of Pb^{208} bombarded with 14.8-Mev deuterons is 363 mb. Less than 10% of these stripping processes leave the residual nucleus with more than 14-Mev excitation energy.

[36] M. M. Shapiro, Phys. Rev. **90**, 171 (1953).

be the probability that fission occurs whether accompanied by the emission of particles or not. Measured reaction cross sections will be denoted as follows: $\sigma_{(d,F)}$, the fission cross section; $\sigma_{(d,n)}$, the (d,n) cross section, etc. Restricting the incident deuteron energy E_d to less than 25 Mev and considering only fissile nuclei, we obtain the following relations:

$$\sigma_{(d,F)}=\sigma_1({}^1q_F)+\sigma_2({}^2q_F)+\sigma_3({}^3q_F), \quad (1)$$

$$\sigma_{(d,n)}=\sigma_2({}^2q_\gamma)+\sigma_3({}^3q_n), \quad (2)$$

$$\sigma_{(d,2n)}=\sigma_2({}^2q_n)+\sigma_3({}^3q_{2n}), \quad (3)$$

$$\sigma_{(d,3n)}=\sigma_2({}^2q_{2n})+\sigma_3({}^3q_{3n}), \quad (4)$$

$$\sigma_{(d,p)}=\sigma_1({}^1q_\gamma). \quad (5)$$

Theoretically[37] the probability of charged-particle evaporation (${}^iq_\alpha$, ${}^iq_p$, etc). from high-Z nuclides is expected to be extremely low, as borne out by the magnitudes of $(d,\alpha\, xn)$ and $(\alpha,\, p\, xn)$ cross sections.[3,33,38] These probabilities can therefore be considered to be negligible. Thus, we may normalize:

$${}^1q_\gamma+{}^1q_n+{}^1q_{2n}+{}^1q_F=1, \quad (6)$$

$${}^2q_\gamma+{}^2q_n+{}^2q_{2n}+{}^2q_F=1, \quad (7)$$

$${}^3q_n+{}^3q_{2n}+{}^3q_{3n}+{}^3q_F=1. \quad (8)$$

When the energy of the protons E_p is less than $E_d-2.23$ Mev, the residual nucleus is excited above the threshold for neutron emission and this or fission is very likely to occur; otherwise only γ emission can occur. For Eqs. (9) and (10) this principle is used to give q_γ.

$${}^1q_\gamma=\frac{\sigma_{(d,p)>}}{\sigma_{(d,p)}}\equiv\frac{\sigma_{(d,p)}(E_p>E_d-2.23)}{\sigma_{(d,p)}}, \quad (9)$$

TABLE IV. Summary of cross sections and decay probabilities used in the calculations for the U^{238} target.

Cross section (mb)	13.6 Mev	20 Mev	Reference
$\sigma_{(d,F)}$	430±150	900±300	This work, 7
$\sigma_{(d,n)}$	22±10	15±5	3, 4, 39
$\sigma_{(d,2n)}$	55±5	40±5	a
$\sigma_{(d,3n)}$	0	15±8	3, 4, 39
$\sigma_{(d,p)}$	180±30	100±20	39
Decay probability			
${}^1q_\gamma$	0.40±0.10	0.20±0.10	34
${}^1q_{2n}$	0	0	b
${}^2q_{2n}$	0	~0	b
${}^3q_F$	0.95±0.03	0.98±0.02	3, 33, 38
${}^3q_n$	0.008±0.007	0.003±0.003	3, 33, 38
${}^3q_{2n}$	0.05±0.04	0.01±0.01	3, 33, 38
${}^3q_{3n}$	0	0.01±0.01	3, 33, 38

[a] W. W. Crane and G. M. Iddings, University of California Radiation Laboratory, Livermore (unpublished data).
[b] Estimated.

[37] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 373.
[38] S. E. Ritsema, University of California Radiation Laboratory Report UCRL-3266, 1956 (unpublished).

TABLE V. Calculated cross sections for excited-nucleus formation, the source of fission, and probabilities of decay for the U^{238} target.

	Mode	13.6 Mev	20 Mev
σ_1	(d,p) stripping	450±200 mb	500±200 mb
σ_2	(d,n) stripping	70±50 mb	70±50 mb
σ_3	d capture	375±200 mb	700±350 mb
f_1	fractional source of fission	0.1±0.1	0.2±0.1
f_2		$0.04_{-0.04}{}^{+0.1}$	$0.02_{-0.02}{}^{+0.1}$
f_3		0.8±0.2	0.75±0.25
${}^1q_n$	competitive decay probabilities	0.5±0.2	0.45±0.25
${}^1q_F$		0.1±0.1	0.35±0.20
${}^2q_n$		0.5±0.2	0.45±0.25
${}^2q_{2n}$		0	0.1±0.1
${}^2q_F$		0.2±0.2	0.30±0.15
${}^2q_\gamma$		0.3±0.2	0.15±0.10

$${}^2q_\gamma=\frac{\sigma_{(d,n)>}}{\sigma_{(d,n)}}\equiv\frac{\sigma_{(d,n)}(E_n>E_d-2.23)}{\sigma_{(d,n)}}. \quad (10)$$

Since (d,p) stripping and (d,n) stripping are similar processes, differing only in the effect of the Coulomb field, their q's should be similar. Relative magnitude of these ${}^1q_x$ and ${}^2q_x$ values can be estimated from proton and neutron spectra and fission thresholds.

$${}^1q_n=K_1\,{}^2q_n, \quad (11)$$

$${}^1q_\gamma=K_2\,{}^2q_\gamma. \quad (12)$$

Unlike stripping processes, the deuteron-capture process gives a compound nucleus with a definite excitation energy E^*. The probability of fission of this heavy nucleus is so great (0.9 to 1.0) at the excitation energies of interest that it can be accurately estimated from α-particle excitation functions[3,33,38] of Np^{237}, Pu^{239}, Pu^{240}, and U^{238}.

$${}^3q_F=[\sigma_{(\alpha,F)}/\textstyle\sum_x\sigma_{(\alpha,x)}]_{E^*}. \quad (13)$$

The probabilities of neutron boil-off (not followed by fission) are so small that a large error (factor of 2 or 3) is permissible in their estimation. Relations analogous to Eq. (13) can be used for this purpose.

The present status of experimental information severely limits the accuracy of the calculation. Nevertheless, as far as the source of fission is concerned [percent of the fission events resulting from (d,p), (d,n) and d capture], an informative solution can be obtained. This is true because one process (d capture) is the predominant source of fission and its error is thus canceled out in the calculation of its fission contribution.

$$(f_i)=(\text{fraction of fission from }\sigma_i)=\sigma_i\,{}^iq_F/\textstyle\sum_i\sigma_i\,{}^iq_F. \quad (14)$$

Calculations were made for $E_d=20$ Mev and 13.6 Mev; at lower energies the uncertainty in the ${}^1q_\gamma$ factor was too great. Table IV summarizes the experimental quantities used and the approximations; Table V gives the results. The solution was obtained by

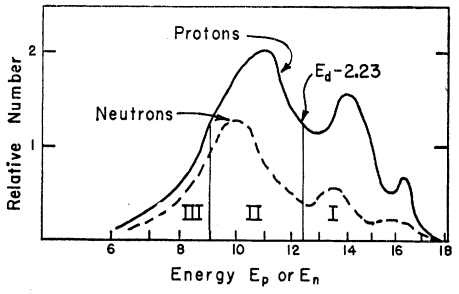

FIG. 5. A comparison of proton and neutron spectra from stripping reactions. The proton spectrum was taken from reference 34 and the neutron spectrum is hypothetical. Excited nuclei which result after events in region I can only emit γ's; nuclei from events in region II probably emit only one neutron; those in region III probably emit two neutrons or undergo fission.

successive trials; a value was taken for σ_3, the unknowns were determined, and the resulting calculated value of σ_3 was required to be consistent with the trial value.

The fission cross sections were taken from a smooth curve drawn through the values of Hicks and Gilbert[7] and the values reported here. The (d,n) and $(d,3n)$ cross sections for U^{238} have not been measured, so an arithmetical average was taken[3,4,39] of $\sigma_{(d,n)}$ and $\sigma_{(d,3n)}$ for Th^{232}, Pu^{238}, Pu^{239}, Pu^{240}, and U^{233}.

The proton spectra measured by Aschenbrenner were used to estimate ${}^1q_\gamma$ with the assumption that protons from electric breakup are unimportant.[34] If this assumption is incorrect, one would expect the electrically broken deuterons to contribute more protons to the energy region $E_p < E_d - 2.23$ which would increase the value of ${}^1q_\gamma$. This in turn would decrease the calculated value of σ_1 and its corresponding contribution to the fission processes. Similar experiments now in progress at Los Alamos by Stokes, Boyer, and Northrup[4] should give more direct information on electric breakup. The Los Alamos group is also measuring the coincidences of protons and fission events‖ induced by 14-Mev deuterons on U^{235} and U^{238}.

Neutron spectra have not been measured; therefore, Eq. (9) could not be used. The K values in Eqs. (10) and (11) were assumed to be $K_1=1$, $K_2=0.75$, on the following reasoning. The only difference between (d,p) stripping and (d,n) stripping is the effect of the Coulomb field on the proton. Therefore, one would expect the protons from (d,p) stripping to carry away from the excited nucleus more kinetic energy than neutrons from (d,n) stripping. This implies that the neutron spectra would be similar to the proton spectra with, however, a larger contribution from lower energy particles. Thus, neglecting the differences in excited nuclei 1 and 2, one would expect ${}^1q_\gamma$ to be larger than ${}^2q_\gamma$; the sum of ${}^1q_F$ and ${}^1q_{2n}$ to be smaller than the sum of ${}^2q_F$ and ${}^2q_{2n}$; and ${}^2q_n$ to be rather close to ${}^1q_n$. This is illustrated in Fig. 5 which shows a comparison of the proton spectrum and the neutron spectrum which may reasonably be expected. Vertical lines separate regions where the residual nucleus is expected to yield mainly (1) γ emission, (2) one-neutron emission, (3) fission and two-neutron emission.

Considering the above approximations and the precision of the available cross sections, one sees that the calculated σ's and q's are accurate to only about 50%. Thus a detailed discussion of the variation of these quantities with energy is not possible at this time. It is considered significant, however, that a large fraction of the fission events follow deuteron capture. This result is borne out by the deuteron fission yields: (a) the v/p ratio discussed in Sec. B which shows an excitation energy of the compound nucleus only slightly less than that calculated by deuteron capture, and (b) fractional chain yields which show[1] that the charge of the average excited nucleus which undergoes fission is greater than the charge of the target nucleus.

The determination of σ_1, σ_2, and σ_3 involves a number of approximations. However, the reliability of the results may be substantiated by the following consideration. Since σ_1, σ_2, and σ_3 are determined by the target-deuteron interaction and are independent of the mode of decay, their values for U^{238} should be nearly the same as those for a nucleus of comparable size such as Bi^{209}. More complete cross-section data[41] are available for deuteron reactions with Bi^{209} and α-particle reactions with several Pb isotopes. Thus, a more accurate calculation of cross sections for excited-nucleus formation may be made. Indeed it is found that σ_1, σ_2, and σ_3 for Bi^{209} are within 25% of those obtained for U^{238}. Hence the approximations shown in Table IV are justified.

V. ACKNOWLEDGMENTS

The advice, encouragement, and aid of Professor C. D. Coryell made this work possible. We are indebted to Professor N. S. Wall, Professor D. H. Frisch, and Professor J. W. Irvine, Jr. for many hours of fruitful consultation. The authors are especially grateful to Dr. P. T. Demos for sponsoring this inter-university collaboration and for his generosity in making special equipment available. The Ordnance Corps and Charles Barton and William Coomey of the Rice Barton Corporation, Worcester, Massachusetts, contributed generously in time, materials, and labor in constructing the massive low-level shields. Earl White and Frank Fay were responsible for providing copious quantities of deuterons. One of us (JMA) is

[39] L. M. Slater, University of California Radiation Laboratory Report UCRL-2441, 1954 (unpublished).

[40] Stokes, Boyer, and Northrup, Bull. Am. Phys. Soc. Ser. II, 2, 207 (1957).

‖ *Note added in proof.*—Dr. J. A. Northrop and Dr. R. H. Stokes inform us that their value of f_1 is ~0.04, in accord with the estimate of Table V.

[41] E. L. Kelly, University of California Radiation Laboratory Report UCRL-1044, 1950 (unpublished); W. John, University of California Radiation Laboratory Report UCRL-3093, 1955 (unpublished).

indebted to E. I. du Pont de Nemours and Company for a teaching fellowship.

APPENDIX. SUMMARY OF CHEMICAL SEPARATION PROCEDURES

The chemical procedures used are based on standard methods [42–44] for the removal of fission-product elements from solutions of the U target. Minor modifications were introduced in some cases to reduce contamination. A summary of the methods and specific references follow.

Zinc.—Precipitated as $ZnHg(SCN)_4$, scavenged with Bi_2S_3, precipitated as ZnS, repeated evaporation with HBr, scavenged with $Fe(OH)_3$+$BaCO_3$. Final sample[45] $ZnHg(SCN)_4$.

Arsenic.—Precipitated as As_2S_3, extracted as AsI_3 from 3*M* HCl+conc. HI into $CHCl_3$,[46] reduced with Cr^{++} to elementary As. Final sample As.

Bromine.—Oxidized with $KMnO_4$ to Br_2, extracted into CCl_4, purified from I_2 by reduction with $NH_2OH \cdot HCl$ and back-extracted into H_2O, extracted into CCl_4. Final sample[47] AgBr. When the target was UO_3, dissolved with cold 6*M* HCl; when the target was metallic U, dissolved with HCl—HNO_3 containing H_2O_2. An NaOH trap was used in the latter case to catch any volatile Br species. Under these circumstances no Br loss is expected.

Strontium and Barium.—Precipitated as $(Sr,Ba)(NO_3)_2$ with fuming HNO_3, scavenged with $Fe(OH)_3$, precipitated Ba as chromate, Sr as oxalate, final samples[48] $BaCl_2 \cdot H_2O$ and $SrC_2O_4 \cdot H_2O$.

Zirconium.—Scavenged with LaF_3, precipitated $BaZrF_6$ and $Zr(OH)_4$ followed by $BaSO_4$ scavenging, extracted Zr from 2*M* HCl into 0.4*M* thenoyltrifluoracetone (TTA) solution in benzene.[49] Final sample ZrO_2.

Ruthenium.—Distilled RuO_4 from $HClO_4$—H_3PO_4 solution plus $NaBiO_3$, precipitated Ru_2O_3 and RuO_2 with ethanol, reduced to elementary Ru with Mg. Final sample[50] Ru.

Cadmium.—Precipitated CdS, dissolved in 6*M* HCl, scavenged with PdS and basic Fe acetate. Final sample[51] $CdNH_4PO_4 \cdot H_2O$.

Antimony.—Extracted Sb(V) into isopropyl ether from 1*M* HCl, reduced with $N_2H_4 \cdot HCl$ to Sb(III) and back-extracted into aqueous KSCN solution, scavenged with elementary Te, reduced with Cr^{++}. Final sample[52] Sb.

Iodine.—Carrier as I^-, oxidized by ClO^- to IO_4^-, reduced to I_2, extracted with CCl_4, reduced with $NaHSO_3$ and back-extracted, oxidized with $NaNO_2$ (Br^- not oxidized) to I_2, extracted, and cycle repeated. Final sample[53] AgI. Iodine was separated only from metallic U targets. Target was dissolved in 20 ml 4*M* HCl+10 ml conc. HNO_3+1–3 drops 30% H_2O_2. In some irradiations an NaOH trap was used to catch possible volatile I species, in others there was no trap. The observed I fission yields were the same within experimental error with and without the trap.

Rare earths (except cerium) and yttrium.—About 20 mg each of carrier Y, Gd, Eu, Sm, Nd, and Pr added to aliquot of target solution. Rare-earth hydroxides precipitated in presence of Sr and Ba. Hydroxides dissolved in conc. HCl and solution passed through the anion exchanger Dowex-1. Rare earths precipitated as fluoride, dissolved in H_3BO_3, scavenged with $Zr_3(PO_4)_4$. Successively precipitated as fluoride and hydroxide, passed in conc. HCl through Dowex-1, precipitated as hydroxide, adsorbed on the cation exchanger Dowex-50 from acid solution,[54] transferred resin to column (1 $cm^2 \times 45$ cm) of Dowex 50-X12, colloidal mesh, in ammonium form. Rare earths eluted with 0.70*M* ammonium lactate, *p*H 3.32, at room temperature, flow rate[55] 0.15–0.20 ml/min. Radiochemically pure fractions obtained in chemical yields of 60–80%. Y eluted after 2 column volumes, Pr after 12. Final sample $(RE)_2(C_2O_4)_3 \cdot 10H_2O$. Pm carried on $La_2(C_2O_4)_3 \cdot 10H_2O$.

Cerium.—Precipitated as Ce (III) hydroxide, scavenged with $Zr(IO_3)_4$, oxidized by BrO_3^-, precipitated $Ce(IO_3)_4$, passed in conc. HCl through Dowex-1. Repeated hydroxide-iodate cycle. Final sample[56] $Ce_2(C_2O_4)_3 \cdot 10 H_2O$. Mass 143 measured by separating 13.7-day Pr^{143} from previously separated Ce fraction. Final sample $Pr_2(C_2O_4)_3 \cdot 10 H_2O$.

[42] Selected papers from reference 25.
[43] Selected papers from Los Alamos Report LA-1721 (rev.), 1954 (unpublished).
[44] Selected papers from University of California Radiation Laboratory Report UCRL-4377, 1954 (unpublished).
[45] J. M. Siegel and L. E. Glendenin, reference 25, Paper 226.
[46] R. J. Prestwood, reference 43, p. 91.
[47] Glendenin, Edwards, and Gest, reference 25, Paper 232.
[48] L. E. Glendenin, reference 25, Paper 236.
[49] G. M. Iddings, reference 44, p. 45.
[50] L. E. Glendenin, reference 25, Paper 260; M. A. Melnick, reference 43, p. 91.
[51] Reference 43, p. 142.
[52] A. C. Pappas, Technical Report No. 63, Laboratory for Nuclear Science, Massachusetts Institute of Technology, 1953 (unpublished); U. S. Atomic Energy Commission Report AECU-2806, 1953 (unpublished).
[53] L. E. Glendenin and R. P. Metcalf, reference 25, Paper 278.
[54] H. G. Hicks, reference 44, p. 29.
[55] E. J. Troianello and T. T. Sugihara (to be published).
[56] H. G. Hicks, reference 44, p. 12.