Further Radiochemical Studies of Fission of the U^{236*} Compound Nucleus*

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Fission-product yields relative to the fission yield of Mo^{99} were measured for the fission of Th^{232} with He^4 ions of energies ranging from threshold to 45 MeV and for the fission of U^{235} with 14-MeV neutrons. The linear relationships between fission yields predicted by the two-mode fission hypothesis and previously observed for fission of U^{236*} at low excitation energies break down at the higher energies in many cases. The fission yield of Mo^{99} and the total fission cross section were evaluated as a function of He^4 ion energy.

I. INTRODUCTION

A PREVIOUS study[1] (hereafter referred to as Paper I) of the neutron-induced fission process in U^{235} and U^{238} has demonstrated linear relationships between the fission yields of any pair of fission products when the excitation energy of the U^{236} or U^{239} compound nucleus varied from about 8 MeV to about 16 MeV.

It was also shown in Paper I that if certain conditions were met such linear relationships could be derived from the two-mode fission hypothesis.[2] The assumptions made were that we were dealing with the fission of a single nuclear species whose two possible fission modes were characterized by mass-yield curves that remained unchanged as the excitation energy was varied. The changes in the observed mass-yield distribution were attributed to changes in the relative proportions of the two fission modes. Although Paper I included results for which it was reasonably certain that more than one nuclear species underwent fission, it was concluded that the mass distributions characterizing the fission modes were similar enough to yield the same linear relationships.

It was of interest to see to just what degree the relationships found in Paper I could be extended to higher excitation energies. With the exception of the 14-MeV neutrons obtainable from an $H^3(d,n)He^4$ reaction, neutron fluxes with mean energies greater than those utilized in Paper I are very difficult to obtain in intensities sufficient for extensive radiochemical studies of the mass distribution in fission. Fortunately, the U^{236*} compound nucleus can also be produced in good yield by the reaction of Th^{232} with He^4 ions. The present work describes the results obtained by bombarding Th^{232} with He^4 ions of energies ranging from slightly above threshold (about 20 Mev) to about 45 MeV and by irradiating U^{235} with 14-MeV neutrons. The range of excitation energies of U^{236*} studied has thus been extended up to approximately 38 MeV with an overlap in the region of excitation produced by 14-MeV neutron bombardment of U^{235}. This overlap permitted examination of the data for any marked differences in the mass distribution of the fission products which might result from the different distribution of spin states expected for U^{236*} formed at the same excitation energy by the two different methods.

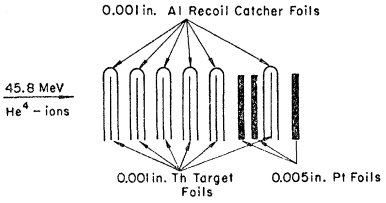

FIG. 1. Schematic diagram of foil assembly for $Th^{232}+He^4$ ion irradiations. In some bombardments, an additional aluminum absorber was placed in front of the first foil to degrade the incident He^{++} energy.

II. EXPERIMENTAL

The 14-MeV neutrons were produced by the $H^3(d,n)$-He^4 reaction using the Cockcroft-Walton accelerator at the Livermore site. Targets of 10 g of uranium (93% U^{235}) were wrapped twice with 0.001-in. 2S aluminum foil and taped as close to the tritium target as possible. After irradiation, the outer aluminum foil was discarded; the inner foil and the $\frac{1}{2}$-in.$\times\frac{1}{2}$-in.$\times\frac{1}{8}$-in. uranium targets were dissolved together in aqua regia with the same precautions as taken in Paper I.

The He^4-ion bombardments were usually made on stacked thorium metal foils and were carried out on the Crocker Laboratory 60-in. cyclotron. The targets for the He^4-ion bombardments were assembled according to Fig. 1. Target foils consisted of 0.001-in. thorium metal wrapped with 0.001-in. 2S aluminum foil to catch any recoils. Behind the fourth or fifth target foil was placed an amount of platinum foil sufficient to stop the He^4-ion beam. Behind this platinum foil was placed another aluminum-wrapped thorium foil to serve as a monitor of the fissions induced by the neutron background present. Behind the monitor foil was another platinum backing foil in thermal contact with cooling water. The 14 foils in the series designated Th+α−1, −2, and −5 were from stacked foil bombardments, while the five foils in the Th+α−3 and −4 series were bombarded singly behind measured thicknesses of aluminum chosen to slow the He^4 ions to the desired energies. The He^4-ion beam intensities were

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[1] H. B. Levy, H. G. Hicks, W. E. Nervik, P. C. Stevenson, J. B. Niday, and J. C. Armstrong, Jr., Phys. Rev. 124, 544 (1961).

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

TABLE I. R values for fission products of Th^{232} bombarded with He^4 ions and U^{235} bombarded with 14-MeV neutrons. (All standard deviations are ±5% unless otherwise noted.)

Bombardment	E^* (MeV)	Zn^{72a}	Sr^{89}	Y^{91}	Zr^{95}	Zr^{97}	Pd^{109}	Ag^{111}	Pd^{112}	Cd^{115}	Cd^{115m}	Cs^{136}	Ba^{140}	Ce^{141}	Ce^{144}	Nd^{147}	Sm^{153}	Eu^{156}	Tb^{161}
Th+α 3–1	17.7	···	···	···	1.06	1.00	38	56	71	64	···	···	···	···	···	···	···	···	···
4–1	19.0	···	0.81	1.04	···	···	42	66	86	82	···	39	0.85	0.86	0.69	0.85	2.01	5.4	80
5–5	19.6	270	0.83	1.03	1.00	1.00	40[a]	76[a]	94	91	91	45	0.83	0.79	0.65	0.83	1.84	5.1	82
3–2	20.0	···	···	···	1.10	1.07	41[a]	76	114	97	···	···	···	···	···	···	···	···	···
1–5	20.4	220	0.93	0.87	···	···	58	86	101	103	103	49	0.85	···	···	0.80	1.70	5.0	···
2–4	21.5	270	0.87	1.07	···	···	···	88	···	118	111	58	0.83	0.89	0.61	0.78	1.73	5.2	97
5–4	23.7	440	0.86	1.08	1.03	1.01	69	114[a]	149	150	150	86	0.82	0.77	0.61	0.76	1.77	5.7	116
3–3	23.9	···	···	···	1.05	1.04	86	124	161	153	···	···	···	···	···	···	···	···	···
1–4	25.9	430	0.95	1.12	···	···	93	135	174	180	176	97	0.86	···	···	0.80	1.86	6.3	137
2–3	27.1	470	0.83	1.01	···	···	···	149	···	194	196	111	0.75	0.75	0.57	0.70	1.81	6.2	144
3–4	29.1	···	···	···	1.00	1.00	123	164	221	217	···	···	···	···	···	···	···	···	···
5–3	29.6	740	0.89	1.05	1.01	1.00	103	176	213	224	231	141	0.77	0.72	0.56	0.70	1.79	6.4	159
1–3	30.4	610	0.98	1.07	···	···	115	182	226	233	240	139	0.81	···	···	0.73	1.81	6.7	168
2–2	31.7	650	0.83	1.03	···	···	···	188	···	249	269	162	0.72	0.71	0.51	0.64	1.74	6.5	176
5–2	33.8	1020	0.87	1.06	0.99	1.00	123	231	270	275	289	194	0.71	0.66	0.50	0.66	1.77	6.9	205
1–2	34.3	840	0.94	1.08	···	···	135	223	268	281	298	182	0.74	···	···	0.68	1.80	7.0	202
2–1	36.2	940	0.81	1.01	···	···	···	230	···	303	327	212	0.65	0.68	0.46	0.59	1.70	6.6	212
1–1	36.6	1040	0.88	1.02	···	···	147	235	301	308	339	211	0.66	···	···	0.61	1.67	6.8	222
5–1	37.9	1300	0.84	1.06	1.00	0.98	140	260[a]	317	324	350	238	0.66	0.75	0.47	0.61	1.79	7.2	239
U+n 1	20.4	220	0.97	0.98	0.98	1.01	53	81	86	103	···	46	0.96	0.95	0.86	0.90	1.93	5.1	94[a]
(Ref. 7)	21.1	···	1.09	0.99[b]	···	1.05	51	76	95	101	···	46	0.86	···	0.69	···	···	4.6	···

[a] Standard deviation = ±10%.
[b] Measured as Sr^{91}.

kept at about 7 μA and were measured with a Faraday cup. Bombardment durations varied from 1 to 5 h depending upon the product nuclides to be studied.

Each thorium foil was dissolved individually with its aluminum recoil catcher foil in nitric acid containing some hydrochloric acid and ammonium fluosilicate. In one bombardment (Th+α−5) perchloric acid was added when dissolution was complete, and the solution heated until strong perchloric acid fumes were evolved. These target solutions were then cooled and diluted to known volume with 6M hydrochloric acid. Duplicate aliquots were withdrawn and added to measured amounts of the proper carriers. Radiochemical purifications were performed by standard methods[3–5] with some minor modifications.

The radiations from the various nuclides were measured and the data calculated in the same way as in Paper I. Results are reported as R values, where

$$R_i=\frac{(\text{fission yield of nuclide } i/\text{fission yield of } Mo^{99})_{\text{experimental conditions}}}{(\text{fission yield of nuclide } i/\text{fission yield of } Mo^{99})_{U^{235}+\text{thermal neutrons}}}.$$

It was shown in Paper I that R values provide a convenient means of testing for linear relationships between pairs of fission products. The equation can be rewritten

$$R_i=\frac{(C_i/C_{Mo^{99}})_{\text{experimental conditions}}}{(C_i/C_{Mo^{99}})_{U^{235}+\text{thermal neutrons}}},$$

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

The fission products studied were Zn^{72}, Sr^{89}, Y^{91}, Zr^{95}, Zr^{97}, Mo^{99}, Pd^{109}, Ag^{111}, Pd^{112}, Cd^{115}, Cd^{115m}, Cs^{136}, Ba^{140}, Ce^{141}, Ce^{144}, Nd^{14}j, Sm^{153}, Eu^{156}, and Tb^{161}.

Monitor foils were used in all bombardments to determine the extent to which fission product activity may have been induced by a neutron background. The activity level of the monitor foils was always much less than those of the target foils, and as a general practice only Mo^{99} and Cd^{115} activities were determined for the monitor foils. However, in one bombardment (Th+α−4) a complete set of "matching" samples was taken from the monitor foil as well as from the target foil. Above incident He^4-ion energies of about 25 MeV the neutron-induced fission background was never more than one-half percent of that induced by the He^4 ions,

[3] M. Lindner, University of California Radiation Laboratory Report UCRL-4377 (unpublished).
[4] W. E. Nervik, J. Phys. Chem. **59**, 690 (1955).
[5] S. R. Gunn, H. G. Hicks, P. C. Stevenson, and H. B. Levy, Phys. Rev. **107**, 1642 (1957).

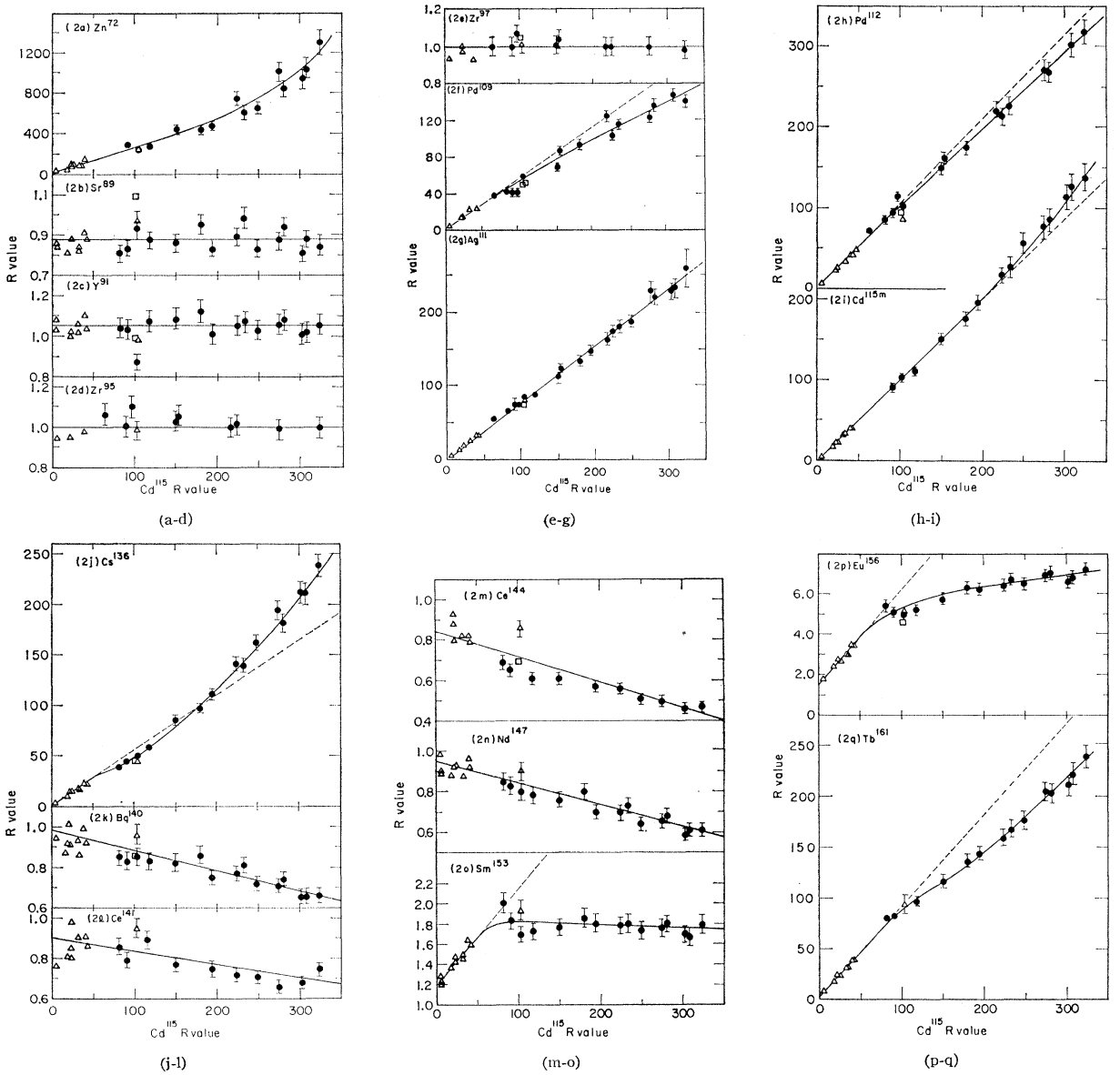

FIG. 2. (a)–(q). R values of various nuclides plotted vs Cd^{115} R values for fission of the U^{236*} compound nucleus: ● $Th^{232}+He^4$ ions; △ U^{235}+neutron reaction (reference 1 and this work); □ U^{235}+14-MeV neutrons (reference 7).

and was usually much less. Below 25 MeV He^4-ion energy, backgrounds varied from bombardment to bombardment but never ran above 5%. The background samples in Th+α−4 showed that the background corrections at these lower energies were approximately the same percentage for all nuclides studied and would tend to cancel out when ratios were taken for R values. Thus, possible errors arising from a back-ground of neutron-induced fission were much smaller than the over-all experimental error from other sources.

III. RESULTS

The results of our investigations are summarized in Table I. In Fig. 2 the R values of the various fission products are plotted vs the corresponding Cd^{115} R values in the same manner as in Paper I. Data from the neutron bombardments reported in paper I are also included; the broken lines are the straight-line relationships derived therein. For many of the cases in which straight lines were not determined in Paper I, straight lines have been drawn through the data for the sake of comparison even when a curve seemed to fit the data better. Data from Ford and Gilmore[6,7] are shown both in Table I and Fig. 2. The data of Newton[8] and of Foreman,

[6] G. P. Ford and J. S. Gilmore (private communication).
[7] G. P. Ford and J. S. Gilmore, Los Alamos Scientific Laboratory Report LA-1997 (unpublished).
[8] A. S. Newton, Phys. Rev. **75**, 17 (1949).

Gibson, Glass, and Seaborg[9] agree with the present work within experimental error.

A. Charge Distribution

There exists the possibility, especially at the higher He^4-ion energies, that members of a mass chain closer to stability than those measured here will be formed in significant abundance as primary fission products. If this occurs, then the behavior of the chosen nuclide will no longer represent the behavior of the total mass chain. We have attempted to examine this possibility by using the MIT prescription[10] to obtain values of Z_p, the most probable charge, for the several mass chains at the various excitation energies. This prescription gives the relationship:

$$Z_p^{\text{corr}}=Z_p^0-\tfrac{1}{2}(Z_c-92)+0.21(A_c-236) \\ -0.19\times 0.12(E^*-6.5),$$

where Z_p^{corr} is the most probable charge of the mass chain in question, Z_p^0 is the most probable charge for thermal-neutron fission of U^{235}, and Z_c, A_c, and E^* are the charge, mass number, and excitation energy of the compound nucleus. The Z_p^0 values for this prescription were obtained from Wahl's[11] empirical Z_p function for thermal-neutron fission of U^{235}. Independent and cumulative yields were then obtained by assuming a charge distribution around Z_p^{corr} of the same shape as given by Wahl.[11] The results obtained from these calculations indicate that even at the highest excitation energies more than 98% of the total mass chain yield was measured in all the chains studied except 97 and 136. The shielded nuclide Cs^{136} represents an independent yield, of course. For Zr^{97} the prescription gives the result that 10 to 15% of the chain is formed in nuclides with Z greater than 40.

These calculations indicate that loss of chain yield is unimportant in this work. However, some recent unpublished results on independent yields[12,13] in He^4-ion induced fission of Th^{232} show that the MIT prescription underestimates the fraction of chain yield formed in members close to stability, at least in a few cases. Unfortunately, there is still no better over-all method for estimating independent and cumulative yields. Rather than making corrections that are dubious, we present the uncorrected data, while keeping in mind that the data for the highest excitation energies may be affected slightly by loss of chain yield.

Both in Paper I and in this paper we have used the R value of the 53-h Cd^{115} to represent the behavior of the 115 mass chain. This is valid so long as both isomeric states of Cd^{115} are formed entirely by β decay. Under such conditions the ratio of 43-day Cd^{115m} to 53-h Cd^{115} will remain constant, and a plot of the R value of Cd^{115m} vs the R value of Cd^{115} will follow a straight line that passes through the origin and has a slope of one. When some of the 115 mass chain is formed directly as Cd, the ratio of the two isomers is likely to change and there will be a deviation from the straight line. Figure 2(i) shows the R values of Cd^{115m} plotted vs the corresponding R values of Cd^{115}. Most of the points follow the line representing a constant ratio of the two isomers. However, at the higher excitation energies there is some deviation from the line, indicating some direct formation of the Cd^{115} isomers. The deviation at the highest energy is about 10%. Since only about 7% of the 115 chain goes through Cd^{115m} by beta decay,[14] the change in the ratio of the two isomers at the highest excitation energy represents a correction to be made to the Cd^{115} R value of slightly less than 1%. Since such a correction is much less than the experimental error, we have ignored it.

B. Excitation Energy of the Compound Nucleus

Without an internal monitor, it would be necessary to calculate the energy degradation of He^4 ions through the stack of aluminum and thorium foils in order to determine the particle energy in a particular foil. Accurate range-energy relationships have been determined for aluminum,[15] but such range-energy relationships can only be estimated for thorium from certain formulae relating other materials to aluminum. Thus, although the energy degradation through the aluminum can be calculated fairly accurately, the degradation through the thorium can only be estimated with an error that is both unknown and likely to be cumulative with several thorium foils in the stack.

In order to minimize such errors, we used an internal energy monitor. Since the Cd^{115} R value was to be used as a reference point for the correlation between R values, we decided to determine the excitation function of the Cd^{115} R value and to use Cd^{115} as an internal energy monitor.

The R values for the Cd^{115} excitation function were measured in thorium target foils that had only aluminum interposed between the target foil and the beam emerging from the cyclotron.[16] The energy of the He^4 ions incident on the target foil was varied by changing the thickness of aluminum in front of the target.[17]

In these runs the background was carefully monitored, and corrections made in the Cd^{115} and Mo^{99} activities.

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[9] B. M. Forman, Jr., W. M. Gibson, R. A. Glass, and G. T. Seaborg, Phys. Rev. **116**, 382 (1959).
[10] C. D. Coryell, M. Kaplan, and R. D. Fink, Can. J. Chem. **39**, 646 (1961).
[11] A. C. Wahl, J. Inorg. Nucl. Chem. **6**, 263 (1958).
[12] M. C. Michel (private communication).
[13] D. R. Nethaway and H. B. Levy (private communication).
[14] S. Katcoff, Nucleonics **18**, 201 (1960).
[15] Hans Bichsel, Phys. Rev. **112**, 1089 (1958).
[16] The range-energy relationship in thorium was estimated by using formulas given by R. M. Sternheimer, Phys. Rev. **115**, 137 (1959) and Phys. Rev. **118**, 1045 (1960), for converting ranges in Al to ranges in other materials.
[17] We acknowledge the assistance of D. R. Nethaway in determining the Cd^{115} R-value excitation function.

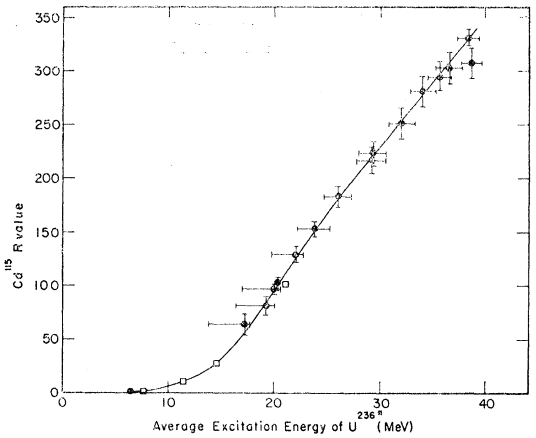

FIG. 3. Cd^{115} R value as a function of the excitation energy of U^{236*}: ● $Th^{232}+He^4$ ion (first foils of stack); △ 14-MeV neutron-induced fission of U^{235} (this work); □ neutron data from reference 7. Limits of the excitation energies are calculated from the incident and emergent energies of the He^4 ions as given in Table II.

The corrected Cd^{115} R values are given in Table II and plotted versus the excitation energy of the U^{236} compound nucleus in Fig. 3 along with Cd^{115} R values measured for U^{235} bombarded with neutrons of various energies. The limits indicated for the excitation energies in Fig. 3 were calculated from the incident and emergent energies of the He^4 ions. The effective average energies were obtained by weighting according to the fission cross-section results reported below. In the lower energy region the limits are not symmetric because of the sharp variation of cross section with energy.

The excitation energies given in column 2, Table I, are those values read from the smooth curve in Fig. 3.

TABLE II. R values of Cd^{115} vs U^{236} excitation energy (MeV).[a]

Particle energy (MeV) ⟨E⟩	Reference	Excitation energy (MeV) E*	R(Cd)115
	Neutrons		
1.2	7	7.6	1.84
5.0	7	11.4	9.47
8.1	7	14.5	26.5
14	This paper	20.4	103
14.7	7	21.1	101.4
	Helium ions		
Incident	Emergent	⟨E⟩[b]	E*	R(Cd115)
22.6	18.8	22.3	17.3	64
24.9	21.4	24.3	19.3	82
25.4	21.9	25.0	19.9	97
27.9	24.8	27.1	22.1	129
30.3	27.2	29.0	23.9	153
32.4	29.8	31.3	26.1	183
35.8	33.0	34.5	29.3	217
35.8	33.2	34.6	29.4	224
38.6	36.1	37.4	32.1	251
40.6	38.1	39.4	34.1	281
42.1	39.9	41.0	35.7	294
43.2	40.6	41.9	36.6	303
44.9[c]	42.5[c]	43.7	38.4	331±8[c]
45.0	43.0	44.0	38.7	308

[a] Standard deviations for this work are 5% unless otherwise noted.
[b] Effective average helium ion energies obtained by weighting according to the cross-section results of Table IV and Fig. 5.
[c] Average of nine determinations.

TABLE III. Yield of Mo^{99} from fission of the U^{236*} compound nucleus.

E* (MeV)	Cd^{115} R value	Thermal fission yield / fission yield	Percent fission yield
6.4[a]	1	1.00	6.06
7.4[b]	...	1.01	6.02
7.6[c]	1.84	1.01	6.02
11.4[c]	9.47	1.02	5.97
14.5[c]	26.5	1.06	5.7
16.4	50	1.16	5.2
20.2	100	1.26	4.81
20.4	103	1.23[d]	4.95
21.1[e]	101	1.19	5.10
23.8	150	1.36	4.46
27.9	200	1.44	4.21
32.0	250	1.51	4.01
36.2	300	1.57	3.86
38.6	330	1.60	3.8

[b] Reference 14.
[a] Reference 20.
[c] Reference 6.
[d] Reference 19.
[e] Reference 7.

C. Fission Yields of Mo^{99}

If one multiplies the R values by the published fission yields for U^{235} at thermal energies,[14] a pseudofission yield curve can be drawn.[2,18] Integrating under the curve and normalizing to 200% enables "true" fission yields to be calculated subject to the additional error from the integration. This technique was applied using R values taken from the smooth curves in Fig. 2 at Cd^{115} R values of 50, 100, 150, 200, 250, 300, and 330. Table III and Fig. 4 present the fission yields of

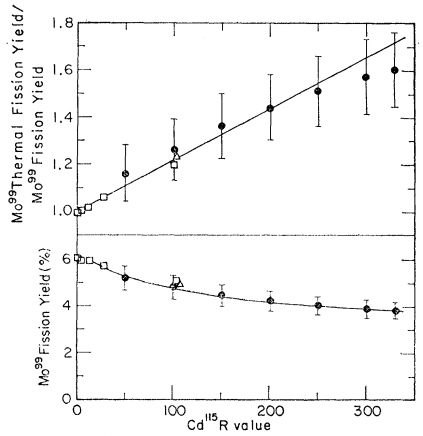

FIG. 4. Yield of Mo^{99} from fission of the U^{236*} compound nucleus plotted *vs* the Cd^{115} R value: ● $Th^{232}+He^4$ ions; △ $U^{235}+14$-MeV neutrons (reference 19); □ $U^{235}+$neutrons (see Table III and references 6, 7, 14, and 20).

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

Mo^{99} calculated from these results and from neutron data.[6,7,19,20] Standard deviations of fission yields of Mo^{99} determined in this way were estimated to be 10%. The value of the Mo^{99} fission yield obtained in this way from data in Table I for fission of U^{235} with 14-MeV neutrons agrees to within 2% of the value given in reference 19.

In paper I the linear relationships implied by the two-mode-fission hypothesis result in the following equation:

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

where c_i and d_i are constants, y_i is the absolute fission yield of mass chain i, and y_0 is the absolute fission yield of the mass chain chosen as a "standard." This equation states that the reciprocal of the Mo^{99} fission yield is a linear function of the fission yield of mass chain i measured relative to that of Mo^{99}; hence the reciprocal of the Mo^{99} fission yield is also a linear function of the R value of mass chain i. This relationship is compatible with our data, as shown in Fig. 4.

TABLE IV. Fission cross section of Th^{232} with He^4 ions.

Av He^4 ion energy[a] (MeV)	Cd^{115} R value	σ_f (b)[b]
19.0	···	$(9.3\pm2.4)\times10^{-4}$[c]
(20.3)	50	$(9\pm1)\times10^{-3}$
22.3	64	0.039
24.3	82	0.078
(24.6)	91	0.14
25.0	97	0.20
(25.5)	103	0.37
(26.5)	118	0.46
27.7	···	0.59±0.15[c]
27.1	129	0.66
(28.8)	150	0.77
29.0	153	0.75
(31.2)	180	0.75
31.3	183	1.04
(32.3)	194	1.00
34.5	217	1.35
(34.9)	224	1.25
34.6	224	1.42
(35.6)	233	1.19
36.3	···	0.93±0.24[c]
(37.0)	249	1.41
37.4	251	1.53
(39.2)	275	1.50
(39.8)	281	1.50
39.4	281	1.64
41.0	294	1.76
41.9	303	1.62
44.0	308	1.76
43.7	324	1.63
44.6	···	1.66±0.41[c]

[a] Weighted average energies as taken from Table II and reference 9. Values in parentheses are based on the Cd^{115} R value and taken from the smooth curve of Fig. 3.
[b] Standard deviations are ±10% unless otherwise noted.
[c] Reference 9.

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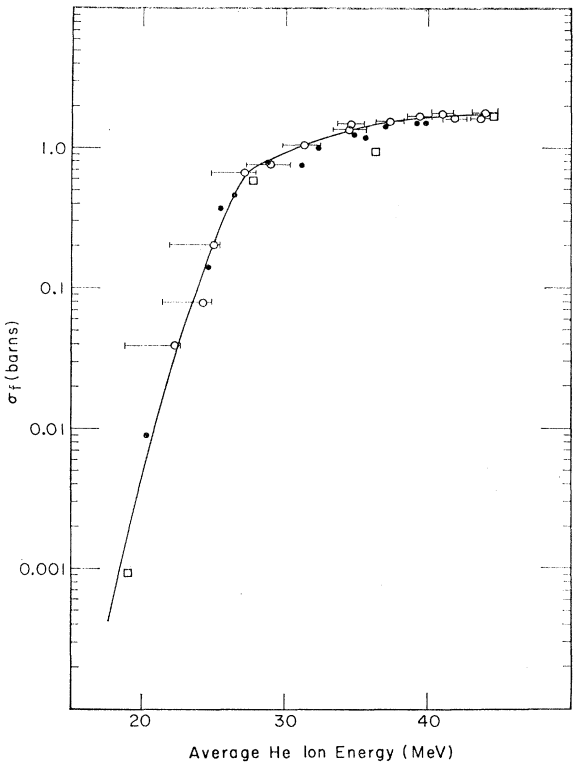

FIG. 5. Fission cross section of Th^{232} with He^4 ions (see Sec. IIID and Table IV): ○ calculated from first foils of stack; ● from other foils and Cd R values; □ data from reference 9.

D. Fission Cross Section for $Th^{232}+He^4$

The number of fissions occurring in a foil can be calculated from the fission yield of Mo^{99} and its counting rate. The fission cross section is calculated from the number of fissions, the measured mass thickness of the thorium foil, and the beam intensity.

The fission cross section vs average helium ion energy is shown in Table IV and Fig. 5. In the case of the first foil of a stack, the helium ion energy was known from range-energy curves,[15] but for foils deeper in the stack, the helium ion energy was derived from Fig. 3, and the Cd^{115} R value. The data of Foreman, Gibson, Glass, and Seaborg[9] agree well with the present data.

IV. DISCUSSION

The linear relationships between the R values of pairs of fission products predicted by the two-mode fission hypothesis and observed in Paper I break down at higher excitation energies (Fig. 2). There are several factors to be considered that could contribute to such a breakdown without necessarily invalidating the general concept of the two-mode fission hypothesis. First, there is the question of the validity of the radiochemical observations as a proper test. Are we measuring the same quantities at the higher energies as we are at the lower energies? Then there is the possibility of the breakdown of some of the assumptions that were needed

to derive the linear relationships without necessarily implying the complete breakdown of the two-mode fission hypothesis. Thirdly, although we are most likely dealing with compound-nucleus systems throughout the entire energy range,[21] we should consider possible effects of differences in the angular momentum of the same system produced in the two different ways ($U^{235}+n$ and $Th^{232}+He^4$).

Concerning the validity of the radiochemical measurements, the possibility that we are no longer measuring total chain yields because of a shift toward stability in the charge distribution of the fission fragments has been discussed in Sec. IIIA. The calculations made for the magnitude of this effect were too uncertain to warrant any changes in the observed data, but indicated that only a small part of the observed deviations from linearity could be accounted for. Further evidence that such a change in charge distribution is not the dominant source of these deviations can be found in the behavior of the isotopes Pd^{109} and Pd^{112}. By any of the postulates of charge distribution currently in vogue (minimum potential energy, equal chain length, or constant charge-to-mass ratio), independent yields of isotopes with $Z>46$ should be larger for mass 112 than for mass 109 and should also become significant at lower U^{236*} excitation energies. Since the Pd^{109} R values fall below the extrapolated lines, one would expect the corresponding R values for Pd^{112} to fall proportionally lower if such a loss of chain yield were the principal source of the deviations from linearity. This is not the case, and on a percentage basis the Pd^{112} points deviate only about one-third as much as the Pd^{109} values.

Another question regarding the validity of the chain yield measurements arises from the additional neutrons which are emitted as the excitation energy of the fission process is increased. If these additional neutrons come from the primary fission fragments, then the mass chains observed radiochemically may represent heavier primary fission products for high-energy fission than they do for low-energy fission ("primary" is used here to mean before neutron emission).

Starting with the general concept of two basic modes of fission, it was necessary to make some further assumptions[1] in order to derive the linear relationships under discussion. We assumed that for a single fissioning nuclide each of the two basic fission modes led to characteristic mass distribution curves. If some of the additional neutrons that are emitted as the excitation energy is increased come out of the compound nucleus, we will observe significant numbers of fissions from lower mass nuclei, such as U^{235*}, U^{234*}, etc., as well as from U^{236*}. Each fissioning nuclide might very well have but two basic fission modes, and the corresponding mass distributions would probably be similar in general shape. However, the observed yields would be averaged over several fission modes and any variation in the details of the individual mass distributions would tend to destroy the linear relationship between R values.

An important aspect of the initial assumptions was that the characteristic mass distributions of the two basic fission modes remained unchanged with increasing excitation energy. Of course, it is quite possible that the mass distribution of one or both modes may change over a broad energy range,[22] thus causing a breakdown of the predicted linear relationships.

The same compound nucleus system is formed by $Th^{232}+He^4$ as by $U^{235}+n$, but the angular momentum brought into the system will differ for the two particles. Thus, at the same excitation energies the distribution of spin states in the compound nucleus will be different for the two methods of formation. In considering the possibile effects of this difference it appears most reasonable to retain our original assumptions[1] for the derivation of linear relationships, with the simple modification that the relative proportions of the two modes may depend upon the distribution of spin states in the compound nucleus as well as on the excitation energy. In such a case, the U^{235} (n,f) points should then fall on a different curve from the $Th^{232}(\alpha,f)$ points in the plot of Cd^{115} R value vs excitation energy given in Fig. 3. Such an effect is not apparent from our data.[23] However, even if the proportions of the two fission modes did depend on the angular momentum in the compound nucleus system, the linear relationship between R_i and R_j should not be affected if our other assumptions remain valid. The points for any set of R_i and R_j values that can be considered a combination of two fixed fission modes will still fall on the same straight line. The behavior of the various plots of R_i vs R_j in the region of overlap seems to imply that the deviations from linearity cannot be explained by simple effects of angular momentum alone. It is, of course, quite possible that increasing angular momentum brought into the compound nucleus adds to the effect of increasing excitation energy in causing a general breakdown of the conditions necessary for the linear relationships.

On examining the plots in Fig. 2 in the light of the various possibilities discussed, we find that the devia-

[21] In their study of helium-ion bombardment of thorium, Foreman, Gibson, Glass, and Seaborg (reference 9) conclude that all fission observed with helium ions of energies up to 46 MeV proceeds through the compound nucleus.

[22] Actually, Fairhall and co-workers have recently examined [A. W. Fairhall (private communication)] the symmetric fission of several nuclei with $Z<90$ at excitation energies comparable to those used in this work and observed that the widths of the symmetric mass distributions became broader as the excitation energy was increased above the fission threshold.

[23] However, Ford and Leachman [Bull. Am. Phys. Soc. **6**, 376 (1961)] report finding such a difference in the ratios of certain "symmetric" fission yields ($Pd^{109,112}$ and $Ag^{111,113}$) to "asymmetric" yields (Zr^{97} and Ba^{140}) above approximately 20-Mev excitation energy. They found an exponential increase with energy in these ratios with a large decreasing step which was greater for $U^{235}(n,f)$ than for $Th^{232}(\alpha,f)$. Our Cd R values increase smoothly with energy at a rate less than exponential for $Th^{232}(\alpha,f)$, but further comparison is restricted by our lack of additional neutron points and by the poor resolution and uncertainty of our He^4-ion energies in the region of interest.

tions from linearity in all cases except that of Tb^{161} are qualitatively consistent with the effect of additional neutrons coming out of either the compound nucleus or the fission fragments. The behavior of Tb^{161} suggests that some other factor is playing a significant role. Most probably the mass distrigutions associated with the basic fission modes are changing with energy.

In Paper I the exactness of the two-mode fission hypothesis was questioned because the results from thermal neutron fission of U^{235} did not fall on the corresponding lines in the R value plots. Despite this, the hypothesis appeared to serve as a good approximation at low energies even though the fission of more than one nuclear species was probably observed. The general concepts of the two-mode fission hypothesis are attractive ones and may be valid. However, it would appear that those conditions that are necessary for quantitative treatments within the framework of the hypothesis can be approximated only over limited energy ranges for a particular fissioning system.

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d-He^4 Elastic Scattering from 6 to 14 MeV*

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Elastic differential cross sections for d-He^4 scattering have been measured for deuteron laboratory energies between 6 and 14 MeV. Both deuterons and alpha particles were accelerated in the Los Alamos variable-energy cyclotron and focused at the center of the Los Alamos multiplate camera. The charged-reaction products were identified on the basis of range measurements in processed $K2$ emulsions. The present results are at variance with previous work at 8 MeV and also indicate significant deviations at large angles from earlier experiments at 13.7 MeV. Around 10 MeV, previous results have been substantiated.

I. INTRODUCTION

DEUTERON–HELIUM elastic scattering has been the subject of extensive investigation at deuteron energies below 5 MeV.[1] At these energies, elastic scattering has proven to be a powerful tool for the study of low-lying excited states in the compound system, Li^6, and has shed some light on the coupling of the two p-shell nucleons. At higher energies, however, angular distribution measurements were made only at 8.0,[2] 10.3,[3] and 13.7 MeV.[4]

Although no recent experimental work has been performed on this problem, theoretical interest has continued to such an extent that a complete bibliography would be excessively lengthy. The analysis of the low-energy data[1] by Galonsky and McEllistrem[5] in 1955 and a general treatment of the deuteron (spin-one) problem by Lakin[6] and Stapp[7] have been both preceded and followed by many excellent papers on the spin polarization of the deuteron.[8]

In 1960, Gammel, Hill, and Thaler developed a

TABLE I. Estimated errors.

Y: particle identification and counting efficiency	1.5%
n: number of incident particles	1.0%
N: number of target nuclei	2.0%
G: slit geometry	1.0%
beam energy	1.0%
statistical	variable
background	25% background correction
water vapor	25% of correction where applicable

* Work performed under the auspices of the U. S. Atomic Energy Commission.

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[8] In addition to the many references given by G. R. Satchler in Oak Ridge National Laboratory Report ORNL-2861, 1960 (unpublished), an excellent treatment can be found in J. Hamilton, *The Theory of Elementary Particles* (Clarendon Press, Oxford, 1959).