Effect of Increasing Excitation Energy on Nuclear Charge Distribution in Fission*

D. R. NETHAWAY AND H. B. LEVY
Lawrence Radiation Laboratory, University of California, Livermore, California
(Received 23 April 1965)

The independent fractional chain yields of Y^{90}, Nb^{96}, and La^{140} have been measured from fission of Th^{232} with alpha particles. The excitation energy was varied from about 20 to 40 MeV. Yields are also calculated for Cs^{136} from data previously published. In addition, the independent fractional chain yields of Y^{92}, Nb^{96}, and La^{140} were measured from fission of U^{235} with 14.9-MeV neutrons. From these data, smoothly varying plots of independent yield versus excitation energy were constructed for Y^{90}, Nb^{96}, Cs^{136}, and La^{140}. The data from 14.9-MeV neutron fission of U^{235} are in good agreement with those from alpha-particle fission of Th^{232} at the same excitation energy. It is shown that, within the limits of certain assumptions regarding the charge dispersion curves, the shift in Z_p (the most probable charge for a given mass number) with increasing excitation energy varies among the nuclides studied. In particular, it appears that dZ_p/dE may be significantly greater for heavy fragments than for light fragments. The data for Y^{90}, Nb^{96}, and La^{140} are consistent with linear plots of Z_p versus E^*, except that the value for Nb^{96} for thermal-neutron fission of U^{235} is off the curve by 0.1 charge unit. The value of dZ_p/dE for Cs^{136} decreases with energy in this energy range, possibly because of shell effects at low excitation energies.

INTRODUCTION

VARIOUS aspects of the complex phenomenon of nuclear fission have been studied intensively off and on for many years. One of the earliest problems to interest investigators and one which has been resistant to a complete theoretical understanding is the question of charge distribution in fission, that is, the relative balance of protons and neutrons in the two fission fragments.

Several attempts have been made to develop a theory to explain charge distribution. None has been successful enough to achieve complete acceptance. Some of the difficulties can no doubt be attributed to the nature of the available data. Most of the data from which charge-distribution information may be derived involve the measurement of independent fission yields. Although techniques have improved over the years, accurate measurements of independent yields are generally difficult, and it is rare that one can find more than a few data for fission occurring under a given set of circumstances for a given nucleus.

Considerably more success has been achieved by various attempts to correlate independent-yield data with charge distribution on a strictly empirical basis. In addition to variations of charge distribution with mass distribution in a given type of fission, one is concerned with the effects of change of excitation energy and the change of charge and mass of the fissioning nucleus.

There have been several papers published concerning the effect of excitation energy on nuclear charge distribution in fission. Pate *et al.*[1] studied the fission of Th^{232} with protons in the energy range 8 to 87 MeV. They found that the width of the charge dispersion curve is constant up to an excitation energy of about 30 MeV, but gradually increases as the excitation energy is raised further. This widening of the charge dispersion curve was ascribed to a wider distribution of fissioning nuclei. They also found that the most probable charge Z_p shifted towards stability with increasing excitation energy, due to increased neutron emission either before or after fission.

Kjelberg *et al.*[2] presented additional data on independent fission-yield measurements from the bombardment of thorium with protons in the energy range 13 to 82 MeV. Friedlander *et al.*[3] extended these results in a study of much higher energy fission, bombarding U^{235} with protons of energies up to 6.2 GeV.

The earlier work of Pate *et al.*[1] was supported by the measurements of Davies and Yaffe,[4] who studied the fission of uranium with protons in the energy range 10 to 85 MeV. They were able to construct charge dispersion curves for several bombarding energies, and found that the full width at half-maximum of the charge dispersion curve remained constant at a value of 2.2 up to an excitation energy of 40 MeV.

Wahl and his co-workers[5] have done a considerable amount of work on measuring independent yields and correlating charge distribution in low-energy fission, particularly thermal-neutron fission of U^{235}. Many of the attempts at deriving correlations of charge distribution utilize U^{235} thermal fission as a primary reference point. Building upon some of Wahl's empirical correlations, Coryell *et al.*[6] proposed a prescription for relating the Z_p values for any type of fission to the corresponding Z_p values for thermal-neutron fission of U^{235}. The independent yields for many fission processes can then be

* This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] B. D. Pate, J. S. Foster, and L. Yaffe, Can. J. Chem. **36**, 1691 (1958).

[2] A. Kjelberg, H. Taniguchi, and L. Yaffe, Can. J. Chem. **39**, 635 (1961).

[3] G. Friedlander, L. Friedman, B. Gordon, and L. Yaffe, Phys. Rev. **129**, 1809 (1963).

[4] J. H. Davies and L. Yaffe, Can. J. Phys. **41**, 762 (1963).

[5] A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1962).

[6] C. D. Coryell, M. Kaplan, and R. D. Fink, Can. J. Chem. **39**, 646 (1961).

estimated by assuming a value for the width of the charge dispersion curve.

We report here on the results of an investigation into the effects of increasing excitation energy on the distribution of nuclear charge in the fission of thorium with moderately low-energy alpha particles. We have measured the independent fission yields of several nuclides as a function of excitation energy in the range 20 to 40 MeV. From this study we hoped to determine as well as possible the shapes of the excitation functions of the independent-yield curves. Bombardment of thorium with alpha particles was chosen since the same compound nucleus is formed (U^{236*}) as in the irradiation of U^{235} with neutrons. In this manner advantage can be taken of the relative wealth of independent-yield measurements already reported for the fission of U^{235} with thermal neutrons, and thus our excitation functions can be extended down to lower energies.

There has been some speculation that thermal-neutron fission of U^{235} may be intrinsically different in some way from higher-energy fission. This is illustrated, for example, by the data of Levy *et al.*,[7] who found that data for thermal-neutron fission did not agree with certain linear relationships deduced from data for higher-energy, neutron-induced fission. The shape of the excitation function for independent yields at low energies might shed some light on this question. In addition to the thorium bombardments, we also report on some irradiations of U^{235} with 14-MeV neutrons. It is of interest to see if the independent yields show any variation with the manner in which the fissioning compound nucleus is formed.

In an unpublished thesis by McHugh,[8] a number of independent yields are reported for the fission of thorium with alpha particles in the energy range 20 to 60 MeV. He used a sensitive mass-spectrometric technique for the measurements and was able to define fairly accurately the excitation functions for independent yields of a number of nuclides. His work was done independently at about the same time as the work we report here. In many respects the two sets of results may be considered complementary to each other.

EXPERIMENTAL DETAILS

Bombardments

The bombardments with alpha particles were carried out at the 60-in. cyclotron which was then at the Crocker Laboratory of the University of California at Berkeley, California. The target assembly consisted of a stack of metal foils contained in a "cat's eye" water-cooled target holder. The targets consisted of 1-mil thorium foils, each surrounded by 1-mil aluminum foils used as catchers for recoil products. Spectrographic analysis of the thorium showed the presence of 200 ppm (parts per million) Fe, 100 ppm Ni and Zn, and 40 ppm Ti and Cr as the major impurities. A stack of three thorium foils was used so that we could obtain three different alpha energies in a single bombardment. Additional aluminum foils were usually included in the target assembly to further degrade the alpha energy to desired values. The alpha energy incident on the front foil was ~46 MeV, that leaving the last foil was as low as 20 MeV. We will describe below in the section on energy measurements the technique used for measuring the mean energy in each thorium foil. It involves the measurement of the Cd^{115}/Mo^{99} ratio, which is very sensitive to the bombarding energy. A typical target assembly is shown in Fig. 1. The length of a bombardment was about 50 min. The integrated alpha beam was usually about 6 μA h, and was measured with a Faraday cup.

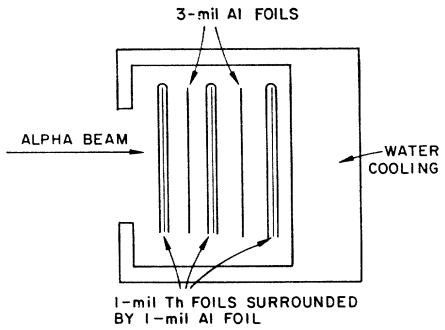

FIG. 1. A typical target assembly used for alpha bombardments.

The irradiations with 14-MeV neutrons were carried out at the Cockcroft-Walton accelerator at the Lawrence Radiation Laboratory at Livermore, California. A 400-keV deuteron beam was allowed to strike a tritium target, producing neutrons by the $t(d,n)\alpha$ reaction with a mean energy of 14.9 MeV. Our target assembly consisted of either uranium (enriched to 93% in U^{235}) foils wrapped in a 10-mil cadmium foil, or, for greater speed in processing, a bag of uranium nitrate hexahydrate wrapped in cadmium. The cadmium covering was used to reduce the low-energy neutron intensity. Irradiations lasted for 5 to 15 min for measurements of Y^{92}, 1 h for La^{140}, and 8 h for Nb^{96}. The neutron intensity was $\sim 2\times 10^{10}$ sec^{-1} in the target, and was monitored continuously by means of an alpha counter so that variations could be corrected for. The contribution to the yields of Y^{92}, Nb^{96}, and La^{140} from fission of U^{238} is $<0.5\%$, and has been neglected.

Chemical Separations—Niobium

After the cyclotron bombardment the thorium foils, together with their associated aluminum recoil-catcher foils, were dissolved in solutions of HCl, HNO_3, and HF. The solutions were then boiled and made up to 6 N

[7] 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).
[8] J. A. McHugh, Jr., thesis, University of California Radiation Laboratory Report UCRL-10673, 1963 (unpublished).

in HCl. Duplicate aliquots were taken and added to ~20 mg amounts of niobium carrier. HF was added to facilitate exchange between carrier and radio-niobium forms. $BaZrF_6$ was precipitated three times to provide a separation from zirconium. (We attempted to measure the independent yield of Nb^{95} as well as Nb^{96}, but it proved to be too small for our separation times.) Another aliquot was also taken and added to cadmium and molybdenum carrier solutions. Standard radiochemical procedures were then followed to purify the niobium, cadmium, and molybdenum samples. Samples were prepared in duplicate in all cases. A similar procedure was followed for the U^{235} foils.

Chemical Separations—Yttrium and Lanthanum

The thorium and aluminum foils were dissolved in solutions of HCl, HNO_3, and HF, with ~40 mg of yttrium and lanthanum carriers present. Two aliquots were taken for a precipitation of yttrium and lanthanum hydroxides in the presence of strontium and barium hold-back carriers, so that a quick separation from the beta-decay precursors of Y^{90} and La^{140} could be made. The hydroxide precipitation was repeated three times. Two other aliquots were also taken and saved for 2 weeks to allow appreciable decay of the fission product precursors to occur. A similar separation of yttrium from strontium and lanthanum from barium was then made. Y^{90} was later milked from the purified strontium fraction. The rare-earth fractions were purified radiochemically, and the individual rare earths separated from each other by selective elution from Dowex-50 cation exchange columns with ammonium lactate. As above, a separate aliquot was taken so that duplicate cadmium and molybdenum samples could be prepared.

For the measurement of Y^{92} from fission of U^{235} with 14-MeV neutrons, an extraction of yttrium into tributylphosphate from 8.V HNO_3 was used for the separation of Y^{92} from Sr^{92}. This separation was repeated on another aliquot several hours later after appreciable decay of the Sr^{92} had occurred. The procedure used for La^{140} from U^{235} fission was essentially the same as that used in the thorium bombardments.

CALCULATIONS AND EXPERIMENTAL DATA

Energy Measurements

A careful measurement of the alpha-particle energy is necessary because of the large loss in energy in passing through our mixed thorium-aluminum target stack. The calculation of energy degradation in thorium is not as accurate as that in aluminum. We decided to employ the internal method of energy measurement used by Hicks *et al.*[9] in their study of thorium fission. Their method involves the measurement of the ratio

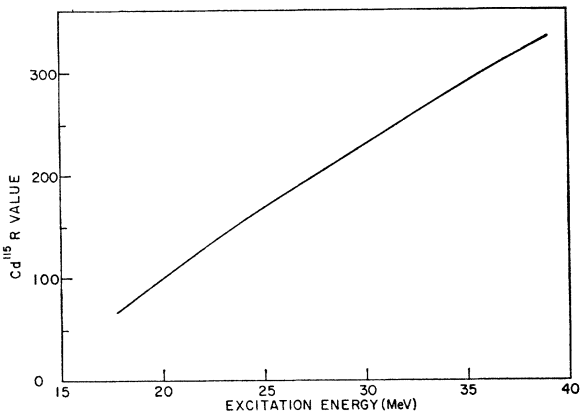

FIG. 2. Cd^{115} R value versus excitation energy.

of Cd^{115} to Mo^{99} produced in the thorium foil. This ratio is essentially the "valley-to-peak" ratio, and it is a very sensitive function of the bombarding energy in this energy range. This provides an effective mean excitation energy for fission averaged over the entire target foil.

Using their notation, we define an R value as

$$R_i=\frac{(Y_i/Y_{Mo^{99}})_{\text{experimental conditions}}}{(Y_i/Y_{Mo^{99}})_{U^{235}+n_{th}}}=\frac{(C_i/C_{Mo^{99}})_{\text{experimental conditions}}}{(C_i/C_{Mo^{99}})_{U^{235}+n_{th}}},\quad(1)$$

where Y_i is the fission yield of the nuclide in question, $Y_{Mo^{99}}$ is the fission yield of Mo^{99}, and the C's are the counting rates, both nuclides being measured in some definite counting arrangement (not necessarily the same for both nuclides). In this manner the rather uncertain corrections for counting efficiencies are cancelled out. Any other nuclide can, of course, be substituted for Mo^{99}.

Hicks *et al.*[9] measured the R value of Cd^{115} in a series of bombardments on thin thorium foils, using additional aluminum foils to degrade the alpha-particle energy to desired values. Figure 2 is a plot of their data for the R value of Cd^{115} as a function of excitation energy in the compound nucleus. It should be noted that this is a universal curve and is independent of the actual counting conditions. We have measured the R value of Cd^{115} for each thorium foil, and have used this curve to calculate the excitation energies. The energy calculated in this manner is an average for the whole foil, and is accurate to within ±1 MeV.

Independent Fission Yields of Y^{90}, Y^{92}, and La^{140}

Consider the generalized fission product decay chain $A \rightarrow B \rightarrow C$, in which x is the cumulative fission yield of A, and y and z are the independent yields of B and C,

[9] H. G. Hicks, H. B. Levy, W. E. Nervik, P. C. Stevenson, J. B. Niday, and J. C. Armstrong, Jr., Phys. Rev. **128**, 700 (1962).

respectively. These yields are fractional values based on the cumulative yield of C.[10] After a bombardment at constant intensity for a time T, separations of C from A and B are made from two aliquots at different times t and t'. The ratio M of the amount of C in the two aliquots, after normalization to the same number of fissions and to the same time, is given by the following equation[5,11] (Ref. 5 gives a useful extension of this equation).

$$M=\frac{x(aJ+bK+cL)+y(\bar{b}K+\bar{c}L)+zL}{x(aJ'+bK'+cL')+y(\bar{b}K'+\bar{c}L')+zL'}, \quad (2)$$

where $J=(1-e^{-\lambda_A T})(e^{-\lambda_A t})$, $K=(1-e^{-\lambda_B T})(e^{-\lambda_B t})$, $L=(1-e^{-\lambda_C T})(e^{-\lambda_C t})$, $a=\lambda_B\lambda_C/(\lambda_B-\lambda_A)(\lambda_C-\lambda_A)$, $b=\lambda_A\lambda_C/(\lambda_A-\lambda_B)(\lambda_C-\lambda_B)$, $c=\lambda_A\lambda_B/(\lambda_A-\lambda_C)(\lambda_B-\lambda_C)$, $\bar{b}=\lambda_C/(\lambda_C-\lambda_B)$, $\bar{c}=\lambda_B/(\lambda_B-\lambda_C)$, and the primed quantities are terms in t' rather than t, e.g., $J'=(1-e^{-\lambda_A T})(e^{-\lambda_A t'})$. After rearranging terms and noting that $x+y+z=1$, the independent yield of C is given by

$$z=\frac{a(1-y)(J-MJ')+[b(1-y)+y\bar{b}](K-MK')+[c(1-y)+y\bar{c}](L-ML')}{a(J-MJ')+b(K-MK')+(c-1)(L-ML')}. \quad (3)$$

This equation may be used to calculate the independent fractional chain yields of Y^{90}, Y^{92}, and La^{140} from a measurement of M, since the half-lives of their precursors are known. It is necessary to estimate the value of y, the independent yield of Sr^{90}, Sr^{92}, or Ba^{140}. The error involved in using the standard Gaussian charge-dispersion curve[5] for this estimation is negligible. The above equation for z was programmed for the IBM 650 computer.

In order to estimate the error in our calculated independent yields we also programmed the IBM 650 computer to solve for the expressions

$$\frac{1}{z}\frac{\delta z}{\delta T}, \frac{1}{z}\frac{\delta z}{\delta t}, \frac{1}{z}\frac{\delta z}{\delta t'}, \frac{\lambda_A}{z}\frac{\delta z}{\delta \lambda_A}, \frac{\lambda_B}{z}\frac{\delta z}{\delta \lambda_B}, \frac{\lambda_C}{z}\frac{\delta z}{\delta \lambda_C}, \text{ and } \frac{M}{z}\frac{\delta z}{\delta M}.$$

These partial-derivative expressions give the fractional change in z for a small change in the various parameters that enter into the previous equation used to calculate the value of z. For each calculation we made what we thought were reasonable guesses for the uncertainties in the times (T, t, and t'), decay constants (λ_A, λ_B, and λ_C), and counting ratio M. We then took the rms value of these calculated errors as the final over-all error. We treated the error in our estimated value of y separately; in all cases the contribution from this error is negligible.

The experimental data and the calculated independent fractional chain yields, z, for Y^{90} and La^{140} from the alpha-particle bombardment of thorium are given in Table I. The data for Y^{92} and La^{140} from the Cockcroft-Walton irradiations are given in Table II. The half-lives and their estimated uncertainties which were used

TABLE I. Independent fractional chain yields of Y^{90} and La^{140}. See text for explanation of symbols.

	Time (h)			Energy		Y^{90}		La^{140}		
Run	T	t	t' (La^{140})	Cd "R"	E^* (MeV)	M	z	M	z	% Error
10 a	3.417	11.75	491	335	38.9			1.758	0.0709	4.0
b				232	30.3			0.996	0.0279	6.4
c				112	21.0			0.645	0.0067	18.0
11 a	1.083	2.75	367	240	30.9	1.480	3.9×10^{-4}	0.566	0.0307	3.6
b				180	26.0	0.499	1.2×10^{-4}	0.348	0.0163	4.4
c				103	20.4	0.800	2.1×10^{-4}	0.201	0.0063	6.8
12 a	0.500	2.00	505	337	39.1	7.22	19.0×10^{-4}	1.496	0.0653	3.0
b				230	30.1	1.84	4.8×10^{-4}	0.630	0.0258	3.5
c				112	21.0			0.233	0.0065	5.6
14 a	0.867	2.40	315	330	38.4	6.65	18.0×10^{-4}	1.039	0.0688	3.1
b				223	29.5	1.921	5.1×10^{-4}	0.438	0.0266	3.6
c				104	20.4	0.768	2.0×10^{-4}	0.163	0.0060	6.4
17 a	0.800	0.65	415	331	38.5	5.30	14.1×10^{-4}	1.343	0.0726	2.9
b				264	32.9	1.865	4.9×10^{-4}	0.758	0.0414	3.0
c				127	22.0	0.956	2.5×10^{-4}	0.199	0.0095	3.9
18 a	0.867	0.55	291	322	37.7	5.18	13.8×10^{-4}	0.977	0.0697	2.9
b				243	31.2	2.42	6.4×10^{-4}	0.483	0.0345	3.1
c				106	20.6	0.682	1.8×10^{-4}	0.119	0.0071	4.2
24 a	0.917	1.13	317	297	35.5	3.58	9.5×10^{-4}	0.849	0.0569	3.0
b				199	27.6	1.874	5.1×10^{-4}	0.380	0.0243	3.4
c				134	22.5	1.203	3.1×10^{-4}	0.184	0.0102	4.1

[10] Whenever we refer to "independent yield" in this article, it is expressed as the fraction of the total chain yield.
[11] A. C. Wahl, Phys. Rev. **99**, 730 (1955). (Note that his value of M is the reciprocal of ours.)

TABLE II. Independent fractional chain yields of Y^{92} and La^{140} from fission of U^{235} with 14.9-MeV neutrons. See text for explanation of symbols.

Run	Time T	Time t	Time t'	M	z Y^{92}	z La^{140}	% Error
1	60 min	15.0 min	16.0 day	0.165		8.8×10^{-3}	6.9
2	60 min	15.5 min	16.0 day	0.141		7.3×10^{-3}	2.3
3	60 min	14.5 min	15.8 day	0.137		7.2×10^{-3}	1.2
4	15 min	9.7 min	4.98 h	0.179	7.3×10^{-3}		67.0
6	5 min	4.8 min	3.38 h	0.0887	6.8×10^{-3}		38.0
7	5 min	4.6 min	1.37 h	0.136	7.5×10^{-3}		37.0
10	5 min	2.9 min	3.20 h	0.0742	7.9×10^{-3}		36.0

in the calculations are: Rb^{90} 2.74±0.08 min, Sr^{90} 28.0±0.3 yr, Y^{90} 2.67±0.01 days, Rb^{92} 5.3±2.1 sec, Sr^{92} 2.71±0.05 h, Y^{92} 3.57±0.05 h, Cs^{140} 1.1±0.2 min, Ba^{140} 12.80±0.06 days, and La^{140} 40.23±0.08 h. There is an error of ±1 min in the cyclotron bombardment times, and the separation times, t and t', have errors of about ±3 and ±6 min, respectively. We estimate that the counting ratio M is known to about ±3% for La^{140} and ±20% for Y^{90}. The latter error is due mainly to the difficulty in resolving a minor component from a complex-decay curve. In the case of Y^{90} the over-all error in the calculation of z is due practically completely to this error in M, and is not listed in Table I. The errors in z for La^{140} were calculated as described previously and are listed in the last column of Table I.

For the Cockcroft-Walton irradiations the errors in T, t, and t' are ±0.1, ±0.4, and ±1 min, respectively, for Y^{92}, and ±0.5, ±1, and ±3 min, respectively, for La^{140}. The counting ratio M is known to about ±5 to ±8% for Y^{92} and La^{140}. The calculated errors in z for Y^{92} and La^{140} are listed in the last column of Table II.

Independent Fission Yields of Nb^{96} and Cs^{136}

The nuclides Nb^{96} and Cs^{136} are both shielded from decay of fission-chain precursors by stable nuclides, hence the calculation of fractional chain yields can be made directly with no correction for formation by β^- decay. We have measured the R value of Nb^{96} at various bombarding energies. We also include here the R value data for Cs^{136} reported by Hicks *et al.*[9] Referring back to Eq. (1), we see that

$$R_i=\frac{(Y_i/Y_{Mo^{99}})_{\text{experimental}}}{(Y_i/Y_{Mo^{99}})_{U^{235}+n_{th}}}$$

which we will take to mean the R value for independent formation of nuclide i referred to the Mo^{99} cumulative yield. We also define an R value for total chain yield

$$R_i(T)=\frac{[Y_i(T)/Y_{Mo^{99}}]_{\text{experimental}}}{[Y_i(T)/Y_{Mo^{99}}]_{U^{235}+n_{th}}}.$$

Dividing one by the other, we have

$$\frac{R_i}{R_i(T)}=\frac{[Y_i/Y_i(T)]_{\text{experimental}}}{[Y_i/Y_i(T)]_{U^{235}+n_{th}}}.$$

Thus, the independent yield that we wish to calculate is given by

$$Y_i/Y_i(T)=[R_i/R_i(T)](Y_i/Y_i(T))_{U^{235}+n_{th}}.$$

The values of the independent yields of Nb^{96} and Cs^{136} for thermal-neutron fission of U^{235}, $[Y_i/Y_i(T)]_{U^{235}+n_{th}}$, have been found to be 9.9×10^{-5} (Ref. 12) and 1.0×10^{-3} (Ref. 13), respectively. We next need information on $R_i(T)$, the R value for the total chain yields of masses 96 and 136. Hicks *et al.*[9] measured the $R_i(T)$ values for Zr^{95} and Zr^{97} in the same range of excitation energies that we are considering and found that both values were constant over the whole energy range. The average value for both masses is 1.02±0.03. We will make the reasonable assumption that $R_{96}(T)$ has this same value over the energy range studied.

For mass 136 we look at the mass-yield curves of Davis[14] and Katcoff[15] and calculate the (mass 136)/(mass 99) yield ratios for fission of Th^{232} with alpha particles and for fission of U^{235} with thermal neutrons. The $R_{136}(T)$ values calculated from these ratios are approximately constant at 0.98±0.05 over the energy range that we are interested in.

The experimental data and the calculated independent fractional chain yields z for Nb^{96} and Cs^{136} are given in Tables III and IV, and we estimate that the over-all error of these values is about ±10%. The values of z for Cs^{136} have been corrected for a small loss of total chain yield due to independent formation of Ba^{136}. This correction was estimated from the standard Gaussian charge-dispersion curve.[5]

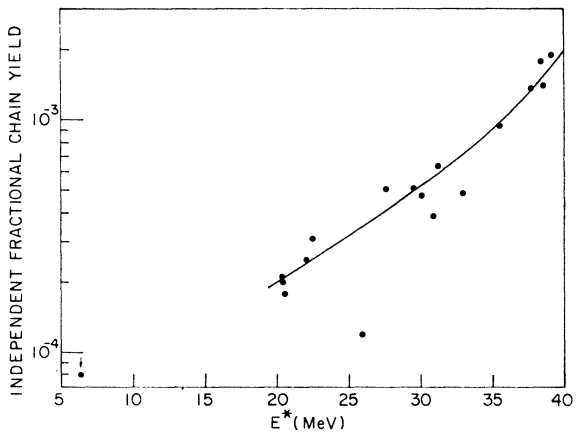

FIG. 3. Independent fractional chain yield of Y^{90} from U^{236}*.

[12] The average of values reported by I. F. Croall, J. Inorg. Nucl. Chem. 16, 358 (1961) and A. C. Wahl *et al.*, Ref. 5.

[13] The average of values reported by W. E. Grummitt and G. M. Milton, J. Inorg. Nucl. Chem. 20, 6 (1961); A. P. Baerg, R. M. Bartholomew, and R. H. Betts, Can. J. Chem. 38, 2147 (1960); and A. C. Wahl *et al.*, Ref. 5.

[14] M. E. Davis, thesis, Purdue University, TID-18131, 1963 (unpublished).

[15] S. Katcoff, Nucleonics 18, 201 (1960).

TABLE III. Independent fractional chain yields for Nb^{96}.

Run	Cd "R"	E^* (MeV)	$R_{Nb^{96}}$	$z_{Nb^{96}}$
25 a	326	38.4	35.8	3.47×10^{-3}
b	276	33.9	26.1	2.53×10^{-3}
c	142	23.1	11.0	1.07×10^{-3}
26 a	303	36.0	30.6	2.97×10^{-3}
b	212	28.6	17.3	1.68×10^{-3}
c	107	20.6	8.3	0.81×10^{-3}
27 a	342	39.6	37.9	3.68×10^{-3}
b	262	32.7	23.4	2.27×10^{-3}
c	174	25.5	12.5	1.21×10^{-3}
CW 11		21.3	8.12	0.79×10^{-3}
12		21.3	8.30	0.81×10^{-3}

TABLE IV. Independent fractional chain yields for Cs^{136}. The values of $R_{Cs^{136}}$ are taken from Ref. 9.

Run	E^* (MeV)	$R_{Cs^{136}}$	$z_{Cs^{136}}$	Corrected $z_{Cs^{136}}$
4–1	19.0	39	0.041	0.041
5–5	19.6	45	0.047	0.047
1–5	20.4	49	0.051	0.051
2–4	21.5	58	0.060	0.060
5–4	23.7	86	0.090	0.090
1–4	25.9	97	0.101	0.101
2–3	27.1	111	0.116	0.116
5–3	29.6	141	0.147	0.146
1–3	30.4	139	0.145	0.144
2–2	31.7	162	0.169	0.168
5–2	33.8	194	0.202	0.200
1–2	34.3	182	0.189	0.188
2–1	36.2	212	0.221	0.219
1–1	36.6	211	0.220	0.218
5–1	37.9	238	0.248	0.245

DISCUSSION

The independent fractional chain yields of Y^{90}, Nb^{96}, Cs^{136}, and La^{140} are plotted versus excitation energy E^* in the compound nucleus in Figs. 3 through 6. The yields of these nuclides have also been measured for thermal-neutron fission of U^{235} ($E^*=6.4$ MeV) and are: $Y^{90}<8\times10^{-5}$ (Ref. 16), Nb^{96} 9.9×10^{-5} (Ref. 12), Cs^{136} 1.02×10^{-3} (Ref. 13), and La^{140} 7.0×10^{-4} (Ref. 16). These data are included in Figs. 3 through 6. The data of McHugh[8] for Cs^{136} are also included in Fig. 5 and are in very good agreement with ours. The curves are drawn only as an aid to inspection of the data. The data are all consistent with a smoothly increasing independent yield as the energy is increased. However, there are some differences in the shapes of the curves.

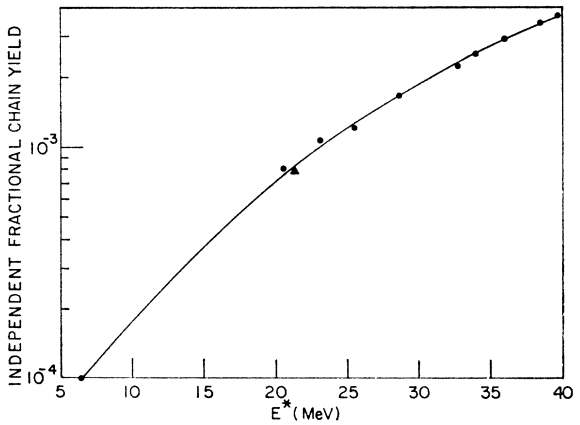

FIG. 4. Independent fractional chain yield of Nb^{96} from U^{236}*. The point for U^{235}+14-MeV n is indicated by a ▲.

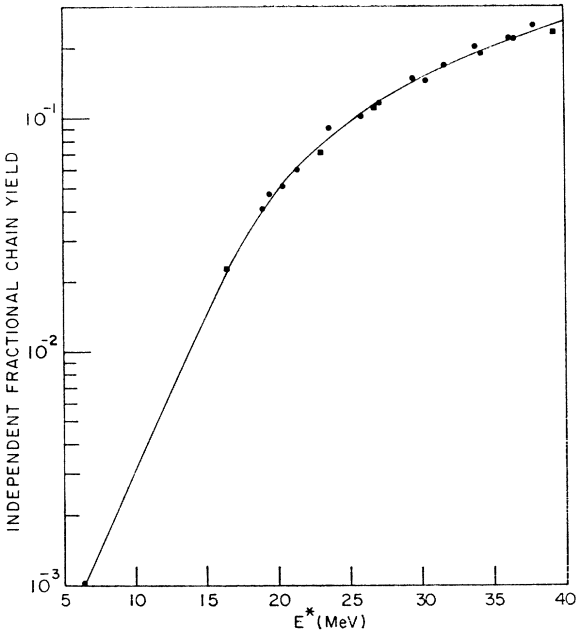

FIG. 5. Independent fractional chain yield of Cs^{136} from U^{236}*. The data of McHugh (Ref. 8) are indicated by a ■.

The data for Nb^{96} and La^{140} at an excitation energy of 21.3 MeV, which were obtained from the 14-MeV neutron irradiations of U^{235}, are in very good agreement with the data from the alpha-particle bombardments of thorium at this same excitation energy. This is somewhat gratifying as it indicates that the method of forming the fissioning compound nucleus has little or no effect on the resulting charge distribution.

We can next look at the change in Z_p, the maximum in the charge-dispersion curve, for the four mass numbers studied. We do this by assuming that a single charge-dispersion curve[5,17] is valid for each mass number, and read off the value of $Z-Z_p$ from the curve for each fractional-chain-yield measurement. Knowing Z, then the value of Z_p can be calculated. The assumption that a single charge-dispersion curve is valid for these mass numbers is critical. However, it has been shown to be true for all mass chains studied where more

[16] W. E. Grummitt and G. M. Milton, J. Inorg. Nucl. Chem. 5, 93 (1957).

[17] We have used a Gaussian curve,

$$z=(c\pi)^{-1/2}\exp[-(Z-Z_p)^2/c],$$

with $c=0.86$.

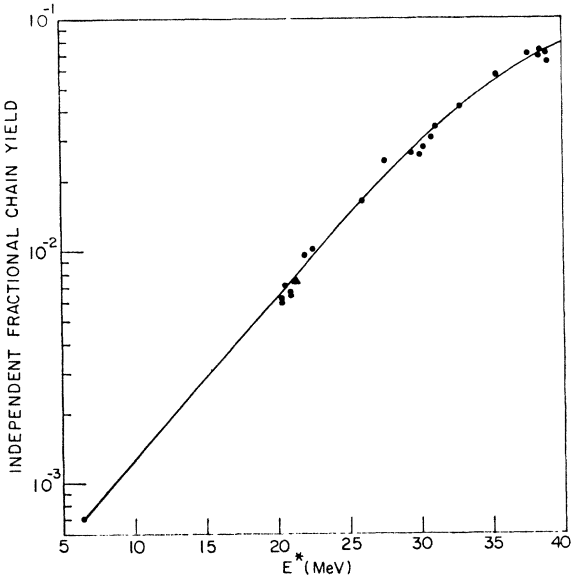

FIG. 6. Independent fractional chain yield of La^{140} from U^{236*}. The point for $U^{235}+14$-MeV n is indicated by a▲.

than one independent yield measurement has been made ($A=91$ to 95 and 139 to 143).[5,18] Furthermore, it was shown by Pate *et al.*,[1] Davies and Yaffe,[4] and McHugh,[8] that the width of the charge-dispersion curve remains constant in this energy range. We have plotted the data in Fig. 7 in the form ΔZ_p versus E^*, where $\Delta Z_p=Z_p-Z_p(U^{235}+n\text{th})$. No attempt has been made to indicate the errors in the ΔZ_p or E^* values. In calculating ΔZ_p for Y^{90}, we have assumed that the independent yield for $U^{235}+n$th is the upper limit (8×10^{-5}) actually measured. If the true value is indeed lower, then all ΔZ_p values for Y^{90} will be shifted up from the zero value by a constant amount. The data of McHugh[8] for Cs^{136} are included in Fig. 7c.

It was proposed by Coryell *et al.*[6] that the only effect of an increase in excitation energy on Z_p would be the effect due to an increase in the neutron emission connected with the fission process. In deriving their prescription for calculating Z_p values Coryell *et al.* assumed that $d\nu_T/dE$ is constant at about 0.12 MeV^{-1} in this energy range. They also assumed that $\nu_L=\nu_H$. If these assumptions held, the shift in Z_p would be a linear function of the excitation energy with a slope $dZ_p/dE=0.023$. Alternately, as proposed by Kaplan,[19] if $d\nu_T/dE$ is a decreasing function of the excitation energy, then the shift in Z_p would tend to level off somewhat at higher excitation energies.

Our data for La^{140} are indeed consistent with a linear change in Z_p with E^*. The slope of the line passing through all points from $E^*=6.4$ to 39 MeV is 0.033 MeV^{-1}. The data for Nb^{96} are linear with a slope of 0.017 MeV^{-1}. However, the point for thermal-neutron fission is off the line by 0.10 charge unit. It is difficult to say how significant this discrepancy is, as we are uncertain about the error in a "ΔZ_p" value.

The data for Cs^{136} do not lie on a straight line at all, and exhibit a slope ranging from 0.065 to 0.028 MeV^{-1}. The data for Y^{90}, which scatter badly, seem to indicate a reverse curvature, but in reality are consistent with a straight line of slope 0.020 MeV^{-1}. In order to lie on the line the value of the independent yield of Y^{90} for thermal-neutron fission must be $\sim3\times10^{-5}$, which is in agreement with the measured upper limit of $<8\times10^{-5}$.

We can go one step further in our analysis of the change in Z_p with excitation energy. There exist a number of measurements of independent yields from fission of U^{236*} at an excitation energy of ~21 MeV. We can compare the values of Z_p calculated from these fractional chain yields and a Gaussian charge-dispersion curve with the corresponding Z_p values for thermal-neutron fission of U^{235}. For the latter values we have chosen to use the empirical Z_p function given by Norris,[18] partly because independent yields for some of the nuclides have not been measured for thermal-neutron fission, and partly to try to avoid any effects due to shell structure. Essentially identical results are obtained if one calculates the Z_p value using measured independent yields for thermal-neutron fission together with the charge dispersion curve. The nuclides for which we feel that sufficiently accurate data exist are listed in Table V, together with their independent yields and Z_p values. Some of the independent-yield data have been obtained by a small interpolation or extrapolation from data at nearby energies. The change in $Z_p(\Delta Z_p)$ between thermal-neutron fission ($E^*=6.4$ MeV) and fission at $E^*=21$ MeV is given for each nuclide. For

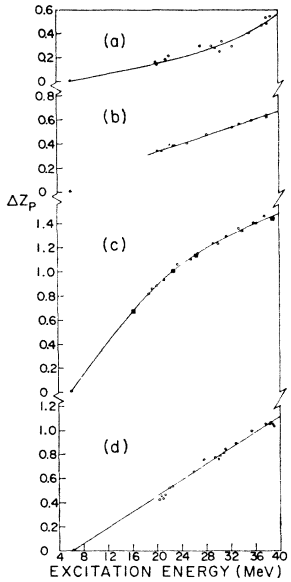

FIG. 7. Plots of $\Delta Z_p=Z_p-Z_p$ ($U^{235}+n$th) versus excitation energy (E^*) for (a) Y^{90}, (b) Nb^{96}, (c) Cs^{136}, and (d) La^{140}. Data of McHugh (Ref. 8) for Cs^{136} are indicated by a ■.

[18] A. E. Norris, thesis, Washington University, St. Louis, Missouri, 1963 (unpublished).

[19] M. Kaplan, thesis, Massachusetts Institute of Technology, Cambridge, Massachusetts, 1960 (unpublished), mentioned in Ref. 5.

TABLE V. Shift in Z_p for fission with $E^*=6.4$ and 21 MeV.

Fragment	Nuclide	Fractional chain yield[a]	Reference	Z_p[b] $E^*=21$ MeV	Z_p[c] $E^*=6.4$ MeV	ΔZp
Light	Br^{82}	1.6×10^{-3}	d	32.74	32.43	0.31
Light	Y^{90}	2.2×10^{-4}	e	36.40	35.86	0.54
Light	Kr^{91}	3.6×10^{-1}*	f	36.71	36.32	0.39
Light	Kr^{92}	1.56×10^{-1}*	f	37.10	36.78	0.32
Light	Y^{92}	7.4×10^{-3}	e	37.05	36.78	0.27
Light	Kr^{93}	3.9×10^{-2}*	f	37.54	37.30	0.24
Light	Nb^{96}	8.3×10^{-4}	e	38.62	38.30	0.32
Heavy	I^{128}	1.8×10^{-3}	8	50.77	50.30	0.47
Heavy	I^{130}	2.7×10^{-2}	8	51.37	50.48	0.89
Heavy	I^{132}	1.6×10^{-1}	11	51.93	51.30	0.63
Heavy	I^{134}	4.3×10^{-1}	11	52.46	51.81	0.65
Heavy	Xe^{135}	3.0×10^{-1}	8	53.22	52.45	0.77
Heavy	Cs^{135}	1.1×10^{-2}	8	53.14	52.45	0.69
Heavy	Cs^{136}	5.8×10^{-2}	e	53.58	52.70	0.88
Heavy	Xe^{138}	7.5×10^{-1}*	f	54.12	53.51	0.61
Heavy	Xe^{139}	4.29×10^{-1}*	f	54.61	53.82	0.79
Heavy	Xe^{140}	2.04×10^{-1}*	f	54.99	54.34	0.65
Heavy	La^{140}	7.3×10^{-3}	e	55.05	54.34	0.71
Heavy	Xe^{141}	5.6×10^{-2}*	f	55.44	54.97	0.47

[a] Fractional chain yields for fission of U^{236}* at $E^*=21$ MeV. Starred values are cumulative yields, others are independent yields.
[b] Z_p values calculated from a Gaussian charge-dispersion curve $z=(c\pi)^{-1/2}\times\exp[-(Z-Z_p)^2/c]$, with $c=0.86$, and the measured independent yields. Z_p values for cumulative yields were taken from footnote f.
[c] Z_p values from the empirical Z_p function of Norris (Ref. 18).
[d] G. P. Ford and J. S. Gilmore, Los Alamos Scientific Laboratory Report LA-1997, 1956 (unpublished).
[e] This work.
[f] K. Wolfsberg, Phys. Rev. 137, B929 (1965).

the light fragment the average ΔZ_p is 0.34 ± 0.04, with $dZ_p/dE=0.023\pm0.003$, and for the heavy fragment the average ΔZ_p is 0.68 ± 0.04, with $dZ_p/dE=0.047\pm0.003$. The choice of a particular Gaussian charge-dispersion curve does not change the conclusion that dZ_p/dE is greater for the heavy fragment. However, the actual values of dZ_p/dE do depend on this choice.

Another way of looking at the relative change in Z_p with energy in the two fragments is shown in the following argument. We assume for the moment that the independent fractional chain yield of a nuclide is given by the Gaussian expression

$$z=(c\pi)^{-1/2}\exp[-(Z-Z_p)^2/c],$$

that c is the same for all mass numbers in low-energy fission, and that Z_p changes linearly with energy $Z_p=Z_p{}^0+bE^*$ in the energy range 20 to 40 MeV. Then the logarithm of the ratio of the independent yields of two nuclides can be expressed as

$$\ln R=\ln z_1-\ln z_2=(-1/c)[(Z_1-Z_{p1}{}^0-b_1E^*)^2-(Z_2-Z_{p2}{}^0-b_2E^*)^2],$$

the derivative of $\ln R$ with respect to E^* as

$$\frac{d\ln R}{dE^*}=(2/c)[b_1(Z_1-Z_{p1}{}^0)-b_2(Z_2-Z_{p2}{}^0)-E^*(b_1{}^2-b_2{}^2)],$$

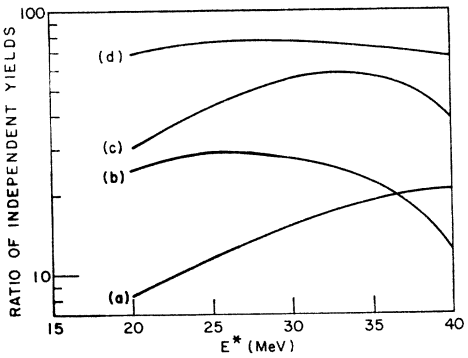

FIG. 8. Ratio of the independent yields (heavy fragment/light fragment). Curve (a), La^{140}/Nb^{96}; Curve (b), Cs^{136}/Y^{90}; Curve (c), La^{140}/Y^{90}; Curve (d), Cs^{136}/Nb^{96}. The independent yields have been read off the smooth curves in Figs. 3 through 6.

and finally the second derivative of $\ln R$ as

$$\frac{d^2\ln R}{dE^{*2}}=\left(-\frac{2}{c}\right)(b_1{}^2-b_2{}^2).$$

Thus, in a plot of $\ln(z_1/z_2)$ versus E^*, a curve that is concave downward implies that $b_1>b_2$. We show in Fig. 8 the logarithm of the ratios of the heavy fragments to the light fragments. All four curves exhibit a downward concavity, which, if our assumptions are valid, implies that $b_1>b_2$, or that dZ_p/dE is greater for the heavy fragment than for the light fragment. One important difference between this treatment and that in the preceding paragraph is that here we have not had to make a choice of a particular Gaussian charge-dispersion curve.

We can reach three conclusions concerning these results:

(1) The values of Z_p for different mass numbers can change with energy in different ways. However, it was pointed out by McHugh[8] that these differences tend to become smaller at higher excitations (above $\sim$23 MeV). It is possible that some of the curvature in the ΔZ_p versus E^* plots is due to shell effects which are less important as the excitation energy is increased. The data for mass 136 show the greatest curvature found to date. The value of Z_p for thermal-neutron fission would have to be raised by 0.57 charge unit to be in agreement with data in the 23–40 MeV range. This would imply a factor of $\sim$13 increase in the independent yield, and it may be that the missing yield is going instead to Xe^{136} which has a closed shell of 82 neutrons. Wahl *et al.*[5] indicated that the independent yield of Xe^{136} may be higher than that expected from the normal charge-dispersion curve.

(2) The shift in Z_p with excitation energy appears to be significantly greater in the heavy fragment than in the light fragment. If we assume, as did Coryell *et al.*,[6] that the only effect on Z_p caused by a change in energy is the result of a change in the number of neutrons

emitted, then we must conclude that with increasing excitation energy neutron emission increases faster for the heavy fragment than it does for the light fragment. This supports the evidence presented by McHugh,[8] who concluded that $d\nu/dE$ was greater for the heavy fragment than for the light. Therefore, the assumption that $\nu_L=\nu_H$ throughout the energy range cannot hold. The method of calculating independent fission yields at different excitation energies proposed by Coryell *et al.*[6] is based in part on the assumption that $\nu_L=\nu_H$ and particularly implies that $d\nu_L/dE=d\nu_H/dE$. Hence it appears to be in error in this respect. On the basis of what limited evidence we have, it appears reasonable to use a value of $\sim$0.047 for dZ_p/dE for heavy fragments, and a value of $\sim$0.023 for light fragments. However, one must exercise caution in extrapolating to or from thermal-neutron fission, especially for nuclides near closed shells.

We would like to point out that the difference in dZ_p/dE for the light and heavy fragments has some implications concerning the use of the equal charge displacement (ECD) rule. As is well known, most of the data for thermal-neutron fission are in agreement with a single charge dispersion curve, with Z_p values calculated from the empirical ECD rule. The fact that the Z_p value for a heavy fragment may change with energy faster than that for a light fragment implies that the ECD rule will be increasingly less successful in correlating data for different mass numbers as the excitation energy is raised.

(3) The inescapable conclusion is that the prediction of independent fission yields at different excitation energies is still fraught with danger. We would like to point out the need for independent yield data from fission with excitation energies of 8 to 18 MeV. The large gap that exists here makes the interpolation from thermal-neutron fission data to higher energy data difficult, and may be hiding important details resulting from shell effects. Unfortunately, experimental data in this area are very difficult to obtain.

PHYSICAL REVIEW VOLUME 139, NUMBER 6B 20 SEPTEMBER 1965

Cross Sections for the Production of Li^9, C^{16}, and N^{17} in Irradiations with GeV-Energy Protons*

I. DOSTROVSKY,† R. DAVIS, JR., A. M. POSKANZER, AND P. L. REEDER
Chemistry Department, Brookhaven National Laboratory, Upton, New York
(Received 8 April 1965)

Twenty-five targets ranging from boron to uranium were irradiated with 1.0- and 2.8-GeV-energy protons. Cross sections for the production of the nuclides Li^9, C^{16}, and N^{17} were measured by counting the delayed neutrons which they emit. No longer lived delayed-neutron emitters were observed except those produced as fission products from uranium. Cross sections for the latter are presented and interpreted in terms of low-deposition-energy processes. Cross sections for the nuclides Li^9, C^{16}, and N^{17} from the lightest targets are interpreted as simple spallation products, mainly from (p,xp) reactions. Several pairs of targets with similar mass number and differing neutron-to-proton ratio were studied. A strong dependence of the cross sections on the neutron-to-proton ratio of the target was observed, especially for the lighter mass regions. Relative cross-section calculations were performed for the heavier targets assuming these and other light fragments were evaporated from excited knock-on cascade products. The effect of secondary evaporation from excited evaporated fragments was included. The experimental and calculated relative cross sections agree well with respect to their dependence on the mass number and neutron-to-proton ratios of the target. It is concluded that the mass-energy surface, which is included in the evaporation formalism, is important in determining the relative yields of light fragments.

INTRODUCTION

THE formation of light fragments (mass 6–24) in considerable yields is one of the characteristics of the interaction of high-energy particles with complex nuclei. Previous investigations of these reactions included measurements of the cross section for the formation of He^6,[1] Be^7,[2] Li^8,[3] C^{11},[4] N^{13},[5] F^{18},[6] and Na^{24} [6] from a range of target nuclei bombarded with GeV-

* Research performed under the auspices of the U. S. Atomic Energy Commission.
† On leave of absence from the Weizmann Institute of Science, Rehovoth, Israel.

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[2] E. Baker, G. Friedlander, and J. Hudis, Phys. Rev. **112**, 1319 (1958); J. Hudis, 1964 (private communication).
[3] S. Katcoff, Phys. Rev. **114**, 905 (1959).
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