Nuclear Charge Distribution in the Fission of Uranium and Thorium with 13.6-Mev Deuterons*

J. M. ALEXANDER† AND C. D. CORYELL
Laboratory for Nuclear Science and Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts
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Measurements have been made of independent fission yields and valley-to-peak ratios in 13.6-Mev deuteron fission and fast-neutron fission of U^{238} and Th^{232}. From these measurements the conclusion is drawn that deuteron capture precedes a large fraction of the deuteron-induced fission events. The primary distribution of yields along a mass chain can be adequately explained by the postulate of equal charge displacement with the same distribution curve found applicable in low-energy neutron fission, best results requiring a slightly greater tendency for neutron emission in the heavy fragments.

I. INTRODUCTION

EXPERIMENTAL studies of the fission process in heavy nuclei ($A > 230$ with less than about 30 Mev incident energy) have always revealed the familiar double-humped yield-mass curve. The valley yields (minimum at symmetric fission) have been found to be extremely sensitive to excitation energy and to be related in only a secondary way to the charge and mass of the fissile nucleus.[1] The fission yields of shielded nuclides have been measured in several cases of low-energy (<3 Mev) neutron fission, and these yields show a strong sensitivity to the charge and mass of the particular fissile nucleus.[2] The yields of shielded nuclides also have a strong energy dependence,[3,4] due to variation in the number of neutrons emitted in fission.

Measurements of valley yields and yields of shielded nuclides thus furnish two excellent supplementary tools for the study of fission induced by deuteron bombardment. This study was undertaken to obtain information concerning the nature of the deuteron interaction with the heavy target nucleus, and to extended knowledge of the distribution of fission yields along a mass chain.

II. EXPERIMENTAL PROCEDURE

The mounting of U and Th foils for deuteron bomment was identical to that described in the preceding paper.[1] For fast-neutron bombardment 10-gram samples of c.p. $Th(NO_3)_4 \cdot 4H_2O$ were placed in plastic tubes directly behind the Be target of the M.I.T. cyclotron. The energy of the neutrons extends from 0 to 19 Mev.

The irradiated samples of U or Th metal were dissolved in HCl (6 to 12M) in most experiments. Under these conditions a black residue is formed which must be dissolved before aliquots are taken. A few milligrams of NaF were sufficient to bring all the Th into solution, and 1 ml of HNO_3 completed the dissolution of U. When halide activities were separated, there was a possibility of loss by volatilization in the addition of HNO_3. This was avoided by having I^- and Br^- carriers present during the dissolution process which was carried out in a distilling flask with a 2M NaOH trap to catch any Br_2 or I_2 that may have been carried over. The basic solution was then added to the original solutions before any separations were made. Chemical separations were performed according to methods outlined in the appendix.

After short bombardments (1 to 10 minutes) samples were separated with measured chemical yields for the determination of Br^{83}, Br^{84}, Sr^{89}, Ag^{113}, Ag^{115}, I^{132}, I^{134}, I^{135}, Ba^{139}, and Ba^{140}; and after longer irradiations determinations were made of Br^{82}, Br^{83}, Rb^{86}, Sr^{89}, Ag^{111}, Ag^{112}, I^{131}, I^{132}, Cs^{136}, Cs^{137}, and Ba^{140}.

End-window proportional counters were used for the counting of radiation, usually from the first-shelf position. Corrections for scattering and absorption[5] relative to the standards (Ba^{140} and Sr^{89}) were calculated to be less than 5% in all cases except Br^{82}, Cs^{136}, and Cs^{137}. These nuclides have soft β radiation, and the absorption correction was in some cases as much as 40%. For the nuclides I^{132}, I^{134}, and I^{135}, γ radiation greater than 0.6 Mev was counted in order to eliminate the contribution of Xe daughters. Corrections due to the resolving time of the counters were never greater than 5%.

III. EXPERIMENTAL RESULTS

Table I summarizes the experimental results for fission yields relative to Ba^{140} for U^{238} fission and relative to Sr^{89} for Th fission. The deuteron energy in Mev is given in subscripts. The errors shown in the table represent the mean deviations of separate determina-

* Supported in part by the U. S. Atomic Energy Commission.
† Present address: Radiation Laboratory, University of California, Berkeley, California.
[1] Sugihara, Drevinsky, Troianello, and Alexander, Phys. Rev. **108**, 1264 (1957), preceding paper. See this paper for general references.
[2] 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/614.
[3] G. P. Ford, U. S. Atomic Energy Commission Report AECD-3597 (unpublished).
[4] A. C. Wahl, Phys. Rev. **99**, 730 (1955).
[5] A. C. Pappas, Technical Report No. 63, Laboratory for Nuclear Science, Massachusetts Institute of Technology, 1953 (unpublished); Atomic Energy Commission Report AECU-2806, 1953 (unpublished).

TABLE I. Deuteron and fast-neutron fission yields for natural uranium and thorium.

Mass No.	Nuclide isolated and measured	Uranium fission yields (relative to Ba^{140}) $d_{13.6}$	d_{10}	$d_{5(\text{fast } n)}$[a]	Thorium fission yields (relative to Sr^{89}) $d_{13.6}$	fast n
82	35.8-hr Br[b]	$(1.0\pm0.1)\times10^{-3}$	9.1×10^{-4}	$\ll7\times10^{-4}$	$(1.1\pm0.3)\times10^{-3}$	$<3.6\times10^{-5}$
83	143-min Br[c]	0.19 ± 0.01	0.15 ± 0.03	0.11 ± 0.01	0.34 ± 0.02	0.29 ± 0.01
84	31.5-min Br	0.23 ± 0.01	0.21 ± 0.02	0.20 ± 0.01	0.40	0.46
86	19.5-day Rb	$(<1.12\pm0.20)\times10^{-4}$	$\ll2.9\times10^{-4}$	...	$<2.5\times10^{-4}$	$\ll1.8\times10^{-4}$
89	54-day Sr	0.61 ± 0.07	0.52 ± 0.05	0.57 ± 0.04	*1.00*	*1.00*
111	7.6-day Ag	0.37 ± 0.02	0.30	0.040	0.31	0.046
112	3.2-hr Ag[d]	0.35 ± 0.03	0.30	0.044	0.31	0.051
113	5.3-hr Ag	0.25 ± 0.01	0.32	0.033	0.25	0.067
115	21.1-min Ag[e]	0.27 ± 0.02	0.30	0.035	0.27	0.071
131	8.07-day I	0.62 ± 0.01	0.77	0.48	0.36	...
132	2.3-hr I[f]	1.07 ± 0.10	1.21	0.96	0.82	...
134	52.5-min I	...	...	...	1.00[g]	...
135	6.7-hr I	...	...	...	1.20[g]	...
136	13.7-day Cs	0.057 ± 0.006	0.049 ± 0.001	$(1.7\pm0.7)\times10^{-3}$	0.04 ± 0.01	$(7.6\pm0.4)\times10^{-4}$
137	30-yr Cs	1.33 ± 0.16	1.52 ± 0.03	1.09 ± 0.08	1.25 ± 0.02	1.04
139	85.0-min Ba	1.24 ± 0.05	1.22 ± 0.10	1.20 ± 0.13	0.85	...
140	12.8-day Ba	*1.05*[h]	*1.05*[h]	*1.00*	0.87	<1.12[i]

[a] The accompanying paper (reference 1) gives evidence that the fission in this case is largely induced by fast neutrons.
[b] Measured relative to Br^{83}.
[c] The fraction of the 83 chain passing through 26-min Se^{83} was found to be 0.45 ± 0.09.
[d] 3.2-hr Ag^{112} was separated from 21-hr Pd^{112} during equilibrium; the yield is therefore that of Pd^{112}.
[e] A correction has been applied (references 17 and 18) for 28% of the 115 chain which β decays through ~20-sec Ag^{115m}.
[f] 2.3-hr I^{132} was separated from 77.7-hr Te^{132} during equilibrium; however, a correction was applied for the yield of I^{132}.
[g] The yields were measured by comparison to uranium fission and are based on estimated values of 1.50 for I^{134} and I^{135} from Fig. 2 of reference 1.
[h] Yields were normalized to 1.00 for Ba^{140}. A 5% correction was applied for the yield of La^{140} in deuteron fission.
[i] Upper limit, contamination from Ra^{226} possible.

tions. Duplicate analyses were made in two or three different experiments for all measurements of uranium yields in 13.6-Mev deuteron fission. When no error is given, the result is a single determination. Systematic errors due to relative counting efficiencies are probably about 10% for all activities except the soft β emitters Br^{82}, Cs^{136}, and Cs^{137}. In these cases this error may be as great as 25%. The yields given in Table I (with the exception of Br^{82}, Rb^{86}, and Cs^{136}) have been corrected for charge distribution[6] as will be described and isomeric decay.[7] They can therefore reasonably be assumed to represent the cumulative yields of the respective mass chains.

Figure 1 shows the dependence of cumulative fission yield on mass number for Th^{232} fission; a corresponding figure for U^{238} fission was shown and discussed in the preceding paper.[1] All measured yields for Th^{232} were reflected about mass number 114.5 by analogy to the deuteron and fast-neutron fission[1] of U^{238}, in which cases the reflection principle was justified for this mass region. A close similarity to pile-neutron fission[8] of Th^{232} is observed in the position of the peaks and in the general shape of the curve. The very broad region of low yields in the valley of near-symmetric fission is quite striking and a more detailed study of yields in this region would be interesting.

Table II gives the fractional chain yields for the six species accessible in this study. The estimation of fractional chain yields f of those nuclides shielded against formation in β decay by the occurrence of stable isobars of $Z-1$ (Br^{82}, Rb^{86}, Cs^{136}) requires knowledge of a cumulative chain yield by interpolation of the smooth yield-mass curve. The fractional chain yields of I^{132}, Te^{134}, and I^{134} were measured by the γ counting of iodine samples separated immediately after bombardment and comparing to samples separated much later. Only those γ's with energies greater than 0.6 Mev were counted, in order that the 2.3-hr I^{132}, 52.5-min I^{134}, and 6.7-hr I^{135} components would be favored with respect to 20.9-hr I^{133} and the 8.07-day I^{131}. The soft γ's of 5.3-day Xe^{133} and 9.2-hr Xe^{135} were completely eliminated and the decay curves resolved as suggested by Wahl.[4] The measurements of

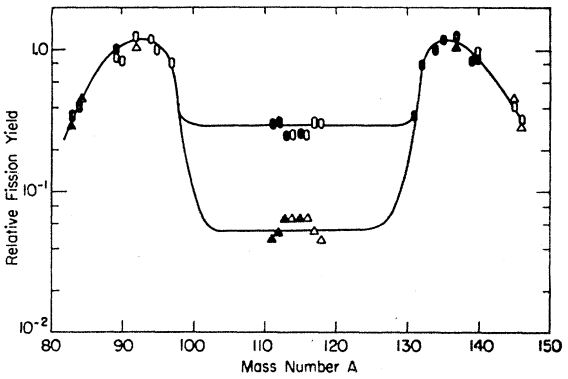

FIG. 1. Cumulative chain yields for Th^{232} fission *vs* mass number A, relative to Sr^{89}. ● measured points for 13.6-Mev deuterons, and ○ the same plotted against 229-A; ▲ measured points for fast neutrons (0–19 Mev), and △ the same plotted against 229-A. Compare Fig. 2 of reference 1 for corresponding data for U^{238} fission.

[6] The correction was less than 2% except for the yields of Ba^{140} and Te^{132}.
[7] Twenty-eight percent of the 115 chain decays through 20-sec Ag^{115m} directly to Cd^{115} (see references 17 and 18).
[8] A. Turkevich and J. B. Niday, Phys. Rev. 84, 52 (1951).

TABLE II. Fractional chain yields

	Uranium fission yields			Thorium fission yields	
	$d_{13.6}$	d_{10}	$d_{5(\text{fast } n)}$	$d_{13.5}$	fast n
Br^{82}	$(1.3\pm0.13)\times10^{-3}$	1.2×10^{-3}	$\ll1\times10^{-3}$	1.4×10^{-3}	$<5.2\times10^{-5}$
Rb^{86}	$<3.3\times10^{-4}$	$\ll8\times10^{-4}$	...	$<4\times10^{-4}$	$\ll3\times10^{-4}$
I^{132}	0.11 ± 0.01[a]	...	<0.04	0.15 ± 0.02	<0.04
Te^{134}	<0.45[b]	...	...	<0.43[b]	...
I^{134}	0.41 ± 0.04[a]	...	0.14 ± 0.02	0.43 ± 0.04	0.077 ± 0.04
Cs^{136}	0.037 ± 0.005	0.030 ± 0.001	$(1.2\pm0.5)\times10^{-3}$	0.033 ± 0.001	$(6.3\pm0.3)\times10^{-4}$

[a] Source of data described in the text.
[b] Fractional chain yield computed for $f_{Xe^{134}}=0.14$ according to calculations described in the text, and ignoring $f_{Sb^{134}}$, which may be about 0.07.

fractional chain yields for I^{132} and I^{134} are unaffected by counting corrections because only the relative activities of different samples are required. The major source of error in these measurements is the resolution of the decay curves with the aid of half-periods taken from the literature.[4]

A correction for the yields of Xe^{132} and Xe^{134} was applied by successive approximations as follows: The distribution of yields along these chains was assumed to be identical to the Gaussian charge distribution found in low-energy fission,[2] and successive approximations were based on this curve. First the Xe independent yields were assumed negligible; the measured $f_{I^{132}}$ or $f_{I^{134}}$ then determined a value of $Z-Z_P$; from this value of of $Z-Z_P$ the $f_{Xe^{132}}$ or $f_{Xe^{134}}$ was estimated from $(Z+1)-Z_P$; the measured $f_{I^{132}}$ or $f_{I^{134}}$ was corrected by this amount and the process was repeated until stable values of $f_{I^{134}}$ resulted. This correction was about 10% for the 13.6-Mev deuteron yields of I^{134} and negligible in all other cases.

The measurement of the extremely low yields of Br^{82} and Rb^{86} is always difficult because of the possibility of interference from the same species produced by neutron activation of natural Br and Rb impurities in the target material. In Table II the observed activity was considered to be due mainly to impurities wherever the symbol $\ll$ appears. The yields could be examined for the presence of impurities by comparing the energy dependence of the fission cross section to the energy dependence of the (d,p) cross section. The $Br^{81}(d,p)$ cross section[9] decreases by a factor of ~2 from a deuteron energy of 13.6 Mev to 10 Mev whereas the fission cross section drops by ~5. Therefore, if Br^{81} impurity were responsible for the observed activity of Br^{82}, there would have been an apparent increase of the fission yield by the factor ~2.5 as the deuteron energy was dropped from 13.6 to 10 Mev. A slight decrease in the fission yield of Br^{82} in this energy region proves the absence of appreciable interfering contamination. On the other hand, the observed activity of Rb^{86} seems to have been largely the result of contamination and only an upper limit could be determined for its yield. Comparison of the fractional chain yields in U and Th fission indicates that Br^{81} contamination is negligible in Th and that Rb^{85} contamination is probably equally serious in Th and U.

IV. DISCUSSION

A. Charge of the Excited Fissile Nucleus

It has been shown by many investigators that the ratio of the cumulative yield in the valley of symmetric fission to the maximum peak yield is mainly determined by the excitation energy (E^*) of the fissile nucleus. A correlation of the experimental information published to date[1] indicates that a given value of the excitation energy fixes the valley-to-peak ratio (v/p) within a factor of about two. The data from all kinds of fission indicate that v/p for any particular kind of fission is a smoothly increasing function of E^* with no peaks or dips outside the experimental error in the energy region of interest. We have taken as a representation of the dependence of v/p on E^* the broken curve shown in the three parts of Fig. 2. This is a smooth curve passing through the points[2,10] for $U^{235}(n,F)$ [Fig. 2(A)], made

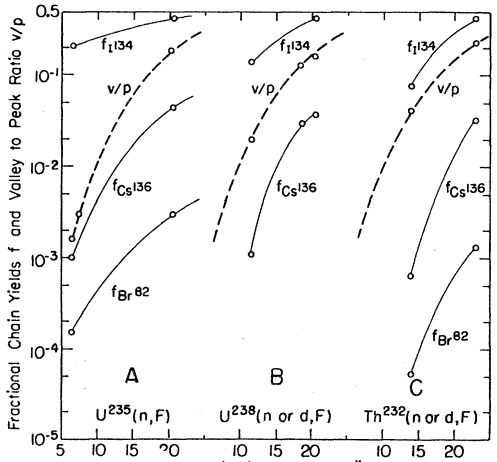

FIG. 2. Comparison of fractional chain yields f with valley-to-peak ratios v/p through the effective excitation energy E^*. Broken line, master curve for v/p vs E^*. Points in part A, literature data (references 2, 3, 4) for U^{235} neutron fission. Points in parts B and C are the data from Table II, with abscissas taken from the fit on the $v/p-E^*$ master curve.

[9] E. T. Clarke and J. W. Irvine, Jr., Phys. Rev. 66, 231 (1944).

[10] R. W. Spence, Atomic Energy Commission Report AECU-645, 1949 (unpublished).

TABLE III. Comparison of fractional chain yields f and valley-to-peak ratios v/p in different fission processes.

	$U^{238}+n$	U^{238}+13.6-Mev d	Observed ratio	$U^{235}+n$	$U^{235}+n$	Reference ratio
v/p	0.021±0.004	0.17±0.03	8.1±2.1	0.021	0.17	8.1
$f_{Cs^{136}}$	(1.2±0.6)×10^{-3}	0.037±0.01	31±16	0.006	0.041	6.8
$f_{I^{134}}$	0.14±0.03	0.41±0.05	2.9±0.7	0.28	0.41	1.46
	$Th^{232}+n$	Th^{232}+13.6-Mev d	Observed ratio	$U^{235}+n$	$U^{235}+n$	Reference ratio
v/p	0.043±0.010	0.24±0.5	5.6±1.9	0.043	0.24	5.6
$f_{Cs^{136}}$	(6.3±3.0)×10^{-4}	0.033±0.006	52±28	0.011	0.067	6.1
$f_{I^{134}}$	0.08±0.04	0.43±0.05	5.4±2.8	0.32	0.45	1.4
$f_{Br^{82}}$	<5.2×10^{-5}	(1.4±0.3)×10^{-3}	>27	9×10^{-4}	4.1×10^{-3}	4.56

parallel to curve A in Fig. 3 of Sugihara *et al.*[1] With the aid of this curve, the E^* values were estimated from v/p ratios for $U^{238}(n$ or $d,F)$ and $Th^{232}(n$ or $d,F)$ from this work.

The other curves in Fig. 2(A) show the energy dependence of fractional chain yields in $U^{235}(n,F)$ for Br^{82}, Cs^{136}, and I^{134} from the literature.[2-4] In Figs. 2(B) and 2(C) are shown the corresponding fractional yields from Table II with E^* calibrations obtained as described above. From the figure it is evident that the fractional chain yields increase much more rapidly in the deuteron fission of Th^{232} and U^{238} than one would expect from the normalization of excitation energy of $U^{235}(n,F)$. Table III gives the comparison of this variation of fractional chain yields and v/p ratio. The second and third columns give the values for v/p and f observed for fast neutrons and 13.6-Mev deuterons on U^{238} and Th^{232}, and the fourth column the ratio of the value at high energy to that at low energy. The fifth and sixth columns give the corresponding U^{235} reference values interpolated on the curves in Fig. 2(A) for the same excitation energies E^*. The last column gives the ratios for the U^{235} reference curves. The estimated errors shown in columns 2 and 3 have been increased from those in Table II to take into account systematic errors. The high errors in column 4 represent the difficulties of quantitative determinations of very low-yield species.

In essence the ratios of f values in column 4 referred to those in column 7 indicate how fast fractional chain yields increase in deuteron fission compared to neutron fission of U^{235} for a given increase in v/p ratio. It is seen that the f values increase with energy in deuteron fission much faster than do the v/p ratios.

From these large increases of f, the conclusion is drawn that in deuteron-induced fission either the whole deuteron or at least its proton is captured by the target nucleus. This decreases the neutron-to-proton ratio of the excited fissile nucleus and correspondingly decreases the neutron-to-proton ratio of the fission fragments, thus raising the yields of the shielded nuclides near β stability ($Z>Z_P$). The excitation energies[1] of 16 to 21 Mev and 14.5 to 18 Mev which correspond to the experimental v/p values for $Th^{232}(d_{13.6},F)$ and U^{238} $(d_{13.6},F)$ suggest that most of the fission events follow compound-nucleus formation by deuteron capture. This conclusion is born out by a more quantitative analysis[1] of the mechanism of fission based on measured cross sections for various deuteron reactions.

B. Charge Distribution

In the correlation of fractional chain yields, the assumption[11] is usually made that the charge distribution is independent of the mass of the fission fragments. This assumption has led to a remarkable correlation of the data from low-energy neutron fission when used in conjunction with the postulate of equal charge displacement of a primary fission-fragment pair.[2,5,11] This postulate requires the following relation for the most probable charge Z_P of a fission product of final mass A_1 after the emission of ν_1 neutrons[2]:

$$Z_P = Z_{(A_1+\nu_1)} - \tfrac{1}{2}[Z_{(A_1+\nu_1)} + Z_{(A_C-A_1-\nu_1)} - Z_C] \quad (1)$$

where Z_C and A_C denote the charge and mass of the compound nucleus formed by the target plus bombarding particle, and Z_A is the most stable charge[12] associated with mass number A.

The postulate of equal charge displacement has been put in question in the region of the 50-proton shell by measurements[13] of an appreciable fractional chain yield of I^{128} in the pile fission of U^{235} and Pu^{239}. Kennett and Thode[13] have shown that a postulate of maximum energy release in fission predicts variations in Z_P near the 50-proton shell which would account for the surprisingly high yield of I^{128}. However, it is clear from any mass formula that this method of calculating Z_P predicts longer chain lengths in the heavy fragments than in the light. The reason for this is the increasing flatness of the isobaric mass parabolas with increasing mass. We have tried to correlate the measured fractional

[11] L. E. Glendenin, C. D. Coryell and R. R. Edwards, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 52, National Nuclear Energy Series, Plutonium Project Report Vol. 9, Div. 4; L. E. Glendenin, Technical Report No. 35, Laboratory for Nuclear Science and Engineering, Massachusetts Institute of Technology, 1949 (unpublished).

[12] C. D. Coryell, Annual Review of Nuclear Science (Annual Reviews, Inc., Stanford, 1953), Vol. 2, p. 305.

[13] T. J. Kennett and H. G. Thode, Phys. Rev. **103**, 323 (1956).

chain yields in low-energy neutron fission[2] with $Z-Z_P'$, calculating Z_P' by the method of maximum energy release.[13] The Levy mass formula[14] was used in this calculation, but similar results would be expected from other mass formulas. There was a great scatter of the data, which seemed to preclude clear dependence of f on $Z-Z_P'$, although the points for heavier masses tend to show longer chain lengths. No improvement was observed by making computations for maximum energy release before neutron boil-off. Until more evidence is adduced, we shall continue with the postulate of equal charge displacement to determine Z_P.

As the excitation energy of the compound nucleus is increased the number of neutrons emitted also increases, and the analysis of fractional chain yields is made more difficult. Large uncertainties due to shell closure are introduced in the quantity $Z_{(A_1+\nu_1)}$ of Eq. (1). If post-fission neutron emission is more pronounced from either the heavy or the light fragments, the assumption of a charge distribution curve independent of mass is more likely to be in error. Any correlation of the independent yields must be made with the knowledge of these sources of difficulty.

Goeckerman and Perlman[15] have shown that fission of Bi by 190-Mev deuterons is best explained by the postulate of unchanged charge distribution rather than that of equal charge displacement. This postulate requires equal charge-to-mass ratio for fission products and the fissile nucleus,

$$Z_P''=\left(\frac{Z_C}{A_C-\bar{\nu}}\right)A_1, \tag{2}$$

where $\bar{\nu}$ is the average number of neutrons per fission, or if primary products are considered,

$$Z_P'''=\left(\frac{Z_C}{A_C}\right)(A_1+\nu_1). \tag{3}$$

These equations predict shorter chain lengths in the heavy fragments than in the light, which the equal-charge-displacement postulate (as modified by Pappas[5]) also predicts if many more neutrons are boiled off the heavy fragment than the light.

There must be a transition in the charge distribution from equal chain lengths observed in low-energy fission to shorter chain lengths observed for heavy fission products in the 190-Mev region. Independent yield measurements by Gibson[16] in the fission of Np^{237} suggest that the transition to unchanged charge distribution is almost complete when the excitation energy of the compound nucleus is ~25 Mev. This conclusion is based on the fractional chain yields of 3.2-hr Ag^{112} and might be invalidated by the presence of an isomer of Ag^{112}, which is not unlikely in the light of the large number of isomers in this region.[17,18]

Fractional chain yields have also been measured[3,4] in the 14-Mev neutron fission of U^{235}. Figure 3 shows these yields in relation to position in chain as calculated by Eq. (1), filled squares for equal charge displacement, and by Eq. (2), open squares for unchanged charged distribution. The latter postulate is seen to give a discontinuity between data for heavy and light fragments, while the former gives a reasonable fit to the charge distribution curve found to apply to low-energy neutron fission. The assumption was made in Fig. 3 that $\nu_1=0.5\bar{\nu}=2.5$. The fit would be improved if ν_1 were taken as 3 for the heavy fragment and 2 for the light. This tendency has been predicted by the statistical theory of fission.[19]

The excitation energy of the compound nucleus in this case of neutron fission is ~21 Mev, which is about the same as the excitation energy in 13.6-Mev deuteron fission of U^{238} and Th^{232}. Therefore the yields from deuteron fission reported in this paper would be expected to lead to a similar correlation. However, the postulate of unchanged charge distribution has been reported to be more successful[16] in the α fission of Np^{237} at an excitation energy of ~25 Mev.

Figure 4 shows the fractional chain yields from this work for U^{238} fission correlated with $Z-Z_P$ predicted by the two postulates in question, and Fig. 5 is a similar plot for Th^{232} fission. The species whose yields are plotted are identified by symbols along the left side at the same ordinate level. The smooth curves shown on the two figures are the same, and identical with that of

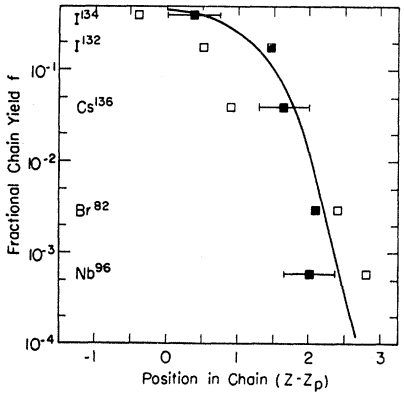

FIG. 3. Comparison of fractional chain yields for 14-Mev neutron fission of U^{235} with postulated $Z-Z_P$ values. Literature data (references 3, 4), assuming that $\nu_1=0.5\bar{\nu}=2.5$ and that Z_P is calculated by two different postulates: ■ Eq. (1), equal charge displacement, and □ Eq. (2), unchanged charge distribution. Horizontal bar, Z_P uncertain because of shell effect on Z_A. Smooth curve taken from Glendenin (reference 2).

[14] H. B. Levy, Phys. Rev. **106**, 1265 (1957); J. Riddell, Atomic Energy of Canada, Ltd. Report CRP-654, 1956 (unpublished).
[15] R. H. Goeckerman and I. Perlman, Phys. Rev. **76**, 628 (1949).
[16] W. M. Gibson, University of California Radiation Laboratory UCRL-3493, 1956 (unpublished).
[17] A. C. Wahl and N. A. Bonner, Phys. Rev. **85**, 570 (1952).
[18] Alexander, Schindewolf, and Coryell, Massachusetts Institute of Technology Laboratory for Nuclear Science Progress Report for May 1957 (to be published).
[19] P. Fong, Phys. Rev. **102**, 434 (1956).

Glendenin.[2,11] Capture of the whole deuteron in 13.6-Mev deuteron fission followed by the emission of 2.5 neutrons from each fragment was assumed for the calculation of Z_P by equal charge displacement, and for fast-neutron fission, the emission of 1.75 neutrons from each fragment. Because of the effects of nucleon shells[12] on Z_A, the value of Z_P computed by Eq. (1) has uncertainties for primary fragments with compositions very near a shell edge, as reflected in the abscissa spread for certain species. This point is discussed further by Pappas.[5] It is clear that a charge distribution independent of mass cannot be obtained by the conventional application of unchanged-charge-distribution equations (2) or (3). A ratio of ~4:1 must be taken for $\nu_{\text{light}}:\nu_{\text{heavy}}$ in order to get a reasonable fit if primary products (before neutron emission) are considered. This alternative seems very unlikely.

Unfortunately, in the application of equal charge displacement [Eq. (1)] only Br^{82} and I^{132} are unaffected by uncertainty in $Z_{(A1+\nu1)}$ due to shell closure. Within the uncertainty, all the points except $f_{I^{134}}$ in Th(fast n,F) lie on the distribution curve found to apply at low energies. As in neutron fission of U^{235}, a better fit would be obtained if $\nu_{\text{heavy}}/\nu_{\text{light}} \approx 2.8/2.2$. The explanation for the low yield of I^{134} in this case might be a fine-structure peak at mass 134 due to preference for the 82-neutron species Te^{134}. Such is the explanation given for the same effect in U^{235}(thermal n,F), although it is not apparent why fine structure would be found in $Th^{232}(n,F)$ but not $U^{238}(n,F)$.

It should be noted that if (d,p) stripping preceded fission in most cases, the application of equal charge displacement would move all points 0.5 unit toward greater $Z-Z_P$ which would give a smooth charge-distribution curve that would sum to more than 1.3, which is impossible.

We conclude that equal charge displacement gives a reasonable picture of the results. The question now

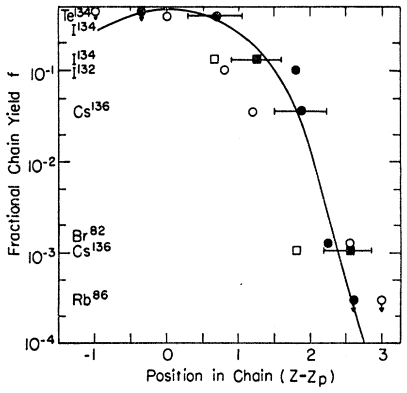

FIG. 4. Comparison of fractional chain yields in U^{238} fission with postulated $Z-Z_P$ values. Circles, 13.6-Mev deuterons, $\bar{\nu}=5.0$, squares, fast neutrons, $\bar{\nu}=3.5$; filled symbols Z_P by Eq. (1), equal charge displacement, and open symbols Z_P by Eq. (2), unchanged charge distribution. Smooth curve taken from Glendenin (reference 2).

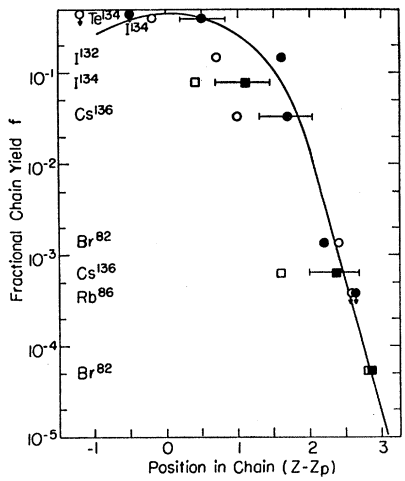

FIG. 5. Comparison of fractional chain yields in Th^{232} fission with postulated $Z-Z_P$ values. Circles, 13.6-Mev deuterons, $\bar{\nu}=5.0$; squares, fast neutrons, $\bar{\nu}=3.5$; filled symbols Z_P by Eq. (1), equal charge displacement, and open symbols, Z_P by Eq. (2), unchanged charge distribution. Smooth curve taken from Glendenin (reference 2).

arises about the best choice of $Z_{(A1+\nu1)}$ for those nuclides affected by shell closure. It might be expected that $Z_{(A1+\nu1)}$ would be taken from a single Z_A–vs–A line,[20] or that it would possibly show a gradual transition with excitation energy from the Z_A of one shell region to that of the next. The latter effect would cause the yields of Cs^{136}, I^{134}, and Rb^{86} to rise more rapidly with energy than other yields the same distance from Z_P. Comparison of Cs^{136} and Br^{82} yields in this work and in $U^{235}(n,F)$ does show this effect.

Wahl[4] treated the fractional chain yields in U^{235} (14-Mev n,F) and showed that the $Z-Z_P$ correlation could be taken to be the same as that for low-energy fission by assuming $\bar{\nu}$ of about 5, but he considered that there was insufficient evidence that the nuclear-charge-distribution pattern remains the same. Ford[3] presents the case for close similarity in the distribution patterns, using data for Br^{82}, I^{132}, I^{134}, and Cs^{136} from U^{235} (14-Mev n,F). The conclusion from the present work, based on still more extended study, is that the postulate of equal charge displacement can be extended to fissile compound nuclei with atomic numbers 90 to 93 and excitation energies 10 to 20 Mev, with no large change being required in the charge-distribution curve derived from low-energy fission yields.

V. ACKNOWLEDGMENTS

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[20] Pappas (reference 5) proposes that this will be the Z_A line corresponding to the shell region of the stable end member of the decay chain.

and Company for a teaching fellowship. Thanks are due Earl White and Frank Fay for the numerous and efficient cyclotron irradiations.

APPENDIX. SUMMARY OF CHEMICAL SEPARATION PROCEDURES

The chemical procedures for the separation of Br, Sr, I, and Ba have been outlined in the preceding paper.[1] The procedures employed for the separation of Rb, Ag, and Cs are standard methods[21,22] with slight modifications as indicated in the following outline.

Rubidium and cesium.—Precipitated as cobaltinitrites, Cs separated from Rb as $CsBi_2I_9$; supernate containing Rb scavenged by 5 more $CsBi_2I_9$ precipitations. Bi_2S_3 and I_2 removed from the Rb fraction. Ba and Sr specifically removed as carbonates. Rb_2SnCl_6 precipitated. Final sample $RbClO_4$.

$CsBi_2I_9$ dissolved in HCl and scavenged with $Fe(OH)_3$ and $Bi(OH)_3$. Cs precipitated as the silicotungstate followed by several precipitations of $CsClO_4$. Final sample $CsClO_4$.

Silver.—Precipitated as AgCl. Two cycles of Ag_2S precipitation followed by $Fe(OH)_3$ scavenging. Ag reduced to metal by Zn powder. Final form AgCl.

[21] Selected papers from *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

[22] Selected papers from Los Alamos Report LA-1721 (rev.), 1954 (unpublished).

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Gamma-Ray Excitation of Ta^{181} Isomer*

J. E. Draper and R. L. Hickok†
Electron Accelerator Laboratory, Yale University, New Haven, Connecticut

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The 615-kev isomeric state in Ta^{181} was excited by 6-Mev bremsstrahlung gamma rays. The half-life and gamma ray spectra were measured with a NaI(Tl) scintillation detector. The measured half-life is 18.1±0.3 μsec. The relative intensities of the 134-kev, 346-kev, and 482-kev gamma rays and the 56-kev x-ray from internal conversion are 57, 16, 100, and 32 and agree with the results obtained from the excitation of this state by beta decay of Hf^{181}. An estimate of the yield is given.

INTRODUCTION

THE (γ,γ') process in nuclei has been demonstrated[1] with incident gamma rays of discrete energies by the process of resonance elastic scattering (the γ and γ' having the same energy). A search for this elastic scattering using incident gamma rays with a continuous energy spectrum was unsuccessful[1] until recently[2] because of the large background from nonresonant interactions. In the present work, information about the related process of inelastic nuclear gamma-ray scattering is obtained by using a pulsed beam, the inelastic gammas causing transitions to an isomeric state which persists after the beam pulse. The purpose is to investigate the reaction $Ta^{181}(\gamma,\gamma')Ta^{181*}$ and to compare the properties of the isomer with those of Ta^{181*} produced by β decay of Hf^{181}.

Excitation of the 615-kev isomeric state in Ta^{181} by bremsstrahlung gamma rays has been reported[3–5] but owing to background difficulties no detailed analysis of the gamma-ray spectrum has been obtained. The Ta^{181*} half-life as determined by pulsed gamma-ray excitation has been reported as 16±3 μsec, 18.5±2.5 μsec, and 20.5±0.4 μsec.

Ta^{181*} is also formed by beta decay of Hf^{181} and the isomeric activity has been studied extensively from sources obtained in this manner.[6–9] Gamma-ray multipolarities have been assigned on the basis of internal conversion coefficients and angular correlation measurements. The decay scheme of the 615-kev isomeric state as reported by Boehm and Marmier[9] is shown in Fig. 1. Half-life determinations by delayed-coincidence measurements between the Hf^{181} beta rays and the Ta^{181*} gamma rays have varied from 22 μsec to 18.8±0.5 μsec.

METHOD

The Yale linear electron accelerator was used to produce electron pulses which had an amplitude of 250 milliamperes, a duration of 1.2 μsec, a repetition

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† Socony Mobil Postdoctoral Fellow.

[1] K. G. Malmfors, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), p. 521.
[2] E. Hayward and E. G. Fuller, Phys. Rev. **106**, 991 (1957).
[3] S. H. Vegors, Jr., and P. Axel, Phys. Rev. **101**, 1067 (1956).
[4] T. F. Godlove and J. G. Carver, Phys. Rev. **99**, 1634(A) (1955).
[5] R. A. Becker and N. H. Brown, Phys. Rev. **90**, 328 (1953).
[6] S. DeBenedetti and F. K McGowan, Phys Rev **70**, 569 (1946).
[7] H. S. Murdoch, Proc. Phys. Soc. (London) **A66**, 944 (1953).
[8] De Brunner, Heer, Kundig, and Rüetschi, Helv. Phys. Acta **29**, 463 (1956).
[9] F. Boehm and P. Marmier, Phys. Rev. **103**, 342 (1956).