found, giving further indication that the conversion coefficient is small. Hence, the last term in the equation was omitted and the equation solved for f at the experimental points shown in Fig. 2. A value of 0.68 $\pm$0.04 was obtained, with no significant trend being observed in the values of f as the amount of aluminum absorber was changed.

CONCLUSION

These data confirm the decay scheme proposed by Cheng and Pool.[1] However, the gamma-ray energy was found to be 157 kev instead of the reported 185 kev.

Nuclear shell theory predicts ground states of $f_{7/2}$ in both Sc^{47} and Ti^{47}. The logft value was calculated to be 6.0 for the 0.62-Mev β ray and 5.3 for the 0.46-Mev β ray. These logft values indicate[7] that both beta groups fall in the $\Delta I=0$, 1 (no) allowed classification. A level scheme consistent with these data is shown in Fig. 3. The assignment of $f_{5/2}$ to the excited state in Ti^{47} would thus make the 157-kev gamma a magnetic dipole transition. The only reported conversion coefficient,[1] 3.6×10^{-3}, falls within the range predicted for either electric or magnetic dipole.[8]

ACKNOWLEDGMENTS

The authors are indebted to Farno Green, who suggested the problem and supervised the irradiations.

[7] Mayer, Moszkowski, and Nordheim, Revs. Modern Phys. **23**, 315 (1951).
[8] M. E. Rose *et al.*, Phys. Rev. **83**, 79 (1951).

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Fission of U^{235} by 14-Mev Neutrons: Nuclear Charge Distribution and Yield Fine Structure*

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The following results were derived from experiments in which iodine was quickly separated from irradiated uranium metal at a known time after irradiation. The fractional chain yields are based on the cumulative chain yields at iodine.

	Ratio of cumulative yields, 14-Mev/thermal neutron fission	Fractional chain yield 14-Mev	Thermal
I^{131}	1.48 $\pm$0.07	<0.08	<0.01
Te^{131m}	5.5 $\pm$1.5	0.35 $\pm$0.05	<0.05
Te^{131}	...	<0.30	<0.03
I^{132}	1.12 $\pm$0.02	0.16 $\pm$0.01	<0.01
Te^{132}	0.97 $\pm$0.02	...	...
I^{133}	0.81 $\pm$0.03	<0.40	<0.05
Te^{133m}	0.64 $\pm$0.15	...	...
Te^{133}	...	<0.40	<0.1
I^{134}	0.65 $\pm$0.02	0.43 $\pm$0.02	0.11 $\pm$0.02
I^{135}	0.69 $\pm$0.02	...	...

The error given is the standard deviation of the average of the experimental results; it does not include the 4 percent standard deviation for the ratio of 14-Mev to thermal neutron fission yields of Mo^{99} used in the calculation of the ratio of cumulative yields. The results indicate that the pronounced peak in the mass-yield curve found at mass number 134 in thermal-neutron fission is nearly washed out in 14-Mev-neutron fission. The higher independent yields of late members of the chains in 14-Mev-neutron fission indicate a shift toward stability of the most probable initial nuclear charge. This shift is about 0.7 charge unit for the chains in the mass region studied.

As by-products of the work, the following half-life values were determined: I^{133}, 20.9$\pm$0.3 hour; I^{132}, 2.30$\pm$0.05 hour; fission precursor of I^{135}, <0.4 minute.

INTRODUCTION

THIS paper presents the results of experiments carried out during the summer of 1951. Prior to the start of this work, Ford and Gilmore[1] had measured the relative cumulative 14-Mev and thermal-neutron yields of I^{131}, and Glendenin[2] had reported values for the independent yields of I^{134} and I^{133} formed in thermal-neutron fission. Pappas,[3,4] in determining the thermal-neutron fission yields of the antimony, tellurium, and iodine isotopes, also obtained a value for the independent yield I^{133}(+2-min Te^{133}).

The general procedure followed was to irradiate for a short time a stack of two or more pieces of uranium metal in a flux of 14-Mev or thermal neutrons. One piece was dissolved immediately after the irradiation, and the iodine quickly separated. The iodine activities in this sample, corrected for the small growth from

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[1] G. P. Ford and J. S. Gilmore (private communication from the Los Alamos Scientific Laboratory).
[2] L. E. Glendenin, Technical Report No. 35, Laboratory for Nuclear Science, Massachusetts Institute of Technology, 1949 (unpublished).
[3] A. C. Pappas, Technical Report No. 63, Laboratory for Nuclear Science, Massachusetts Institute of Technology, 1953 (unpublished).
[4] A. C. Pappas, Progress Report, Laboratory for Nuclear Science, Massachustetts Institute of Technology, May, 1951 (unpublished).

antimony and tellurium precursors, gave a measure of the independent yields of the iodine isotopes. A second sample was dissolved and the iodine separated after the bulk of the antimony and tellurium precursors had decayed. The activities in this sample, after application of appropriate growth and decay corrections, gave a measure of the cumulative yields of the iodine isotopes.

EXPERIMENTAL METHODS

Irradiations

Thermal-neutron irradiations were carried out in the Los Alamos Homogeneous and Fast Reactors. For irradiations of less than one minute duration, the stacked uranium foils were injected into and out of an operating reactor by use of a pneumatic rabbit. The 14-Mev neutron irradiations were carried out in the Los Alamos Cockcroft-Walton accelerator. The $T(d,n)He^4$ reaction produced the fast neutrons. Two approximately 3-gram pieces of uranium metal were placed directly behind the tritium-zirconium target.

Chemistry

An irradiated piece of uranium metal (~0.3 g or ~3 g) was dissolved in a mixture of 5 or 10 ml of concentrated hydrochloric acid and 2.00 ml of KI carrier solution (20.0 mg of iodine). (The larger volume of hydrochloric acid, and of water and hydrogen peroxide used later, was for the ~3-gram samples.) The dissolving took place in an open-top separatory funnel fitted with a centrifugal stirrer. After solution was complete, 5 or 10 ml of water and 25 ml of carbon tetrachloride were added, the stirrer started, and 1.5 or 7 ml of 1 f H_2O_2 was added in a slow stream. The hydrogen peroxide oxidized the U^{+3}, formed on solution of the metal, to a mixture of U^{+4} and UO_2^{++} and most of the I^- to I_2, which was extracted into the carbon tetrachloride. The stirrer was stopped, and as the phases separated the carbon tetrachloride phase was drained into another separatory funnel.

The time the stirrer was stopped was taken as the time of separation of the iodine isotopes from their tellurium precursors. This time is in error by no more than 30 seconds since the stirrer was stopped within 30 seconds after the start of the hydrogen peroxide addition, and the phase separation was completed within 30 seconds after the stirrer was stopped. For the ~0.3-gram samples, these times were about 15 seconds.

The aqueous phase containing the uranium and the bulk of the fission products was treated with excess hydrogen peroxide and with bromine. The resulting solution was used for the Mo^{99} determination,[5] which gave a measure of the number of fissions.

The iodine in the carbon tetrachloride phase was back-extracted into an aqueous solution of $NaHSO_3$, purified by two oxidation-reduction cycles, and precipitated as AgI for chemical yield determination and counting. The oxidation-reduction cycles, consisting of $NaNO_2$ oxidation of iodide ion in dilute nitric acid, extraction of I_2 into carbon tetrachloride, and back extraction of the iodine into an aqueous $NaHSO_3$ solution, and the precipitation procedure were essentially those outlined by Glendenin and Metcalf.[6]

The decay of the gamma activity of a sample was followed on a scintillation counter through 11.7 g/cm² of lead, which absorbed the soft gamma rays from Xe^{133} and Xe^{135} growing from I^{133} and I^{135}. After five days, when most of the xenon had grown, a sample was dissolved by digesting it along with the filter paper and Scotch tape cover for about 15 minutes in concentrated ammonium hydroxide containing metallic zinc. After reduction of silver iodide to silver was complete, the solution was acidified and boiled to remove any remaining xenon, and the iodine carried through another oxidation-reduction cycle. It was precipitated as AgI as before, the chemical yield determined, and the precipitate mounted for counting under 5 mils of aluminum. This aluminum cover plus the 0.7-mil aluminum window on the beta-proportional counter absorbed most of the soft beta particles and electrons from Xe^{133} and Xe^{131m} so that the beta-decay curve was easily resolved into its I^{133} and I^{131} components. It is estimated that <5 percent of the long-lived activity attributed to I^{131} could have been due to xenon.

A coincidence correction of 1 percent per 10^5 counts/min was applied to the counting data.

Uranium metal was used with the expectation[7] that radioactive iodine isotopes formed in the uranium metal would exchange readily and completely with the iodide ion carrier present during the solution process. This point was checked experimentally. A stack of four uranium metal foils were irradiated with thermal neutrons. Two of the foils were worked up for iodine by the method described in the previous paragraphs; two were carried through additional steps designed to further exchange. Oxidation in alkaline solution of I^- to IO_4^- by ClO^- was carried out as recommended by Glendenin and Metcalf,[6] and reduction in acid of the IO_4^- to I^- by H_2SO_3 and precipitation of PdI_2 was included as recommended by Ford and Gilmore.[1] The decays of the four samples mounted as AgI were followed under lead on the scintillation counter, and the decay curves were resolved into their I^{131}, I^{133}, and I^{135} components. The results summarized in Table I

[5] I wish to thank C. O. Minkkinen and J. S. Gilmore for running the Mo^{99} determinations.

[6] L. E. Glendenin and R. P. Metcalf in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 278, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 1625.

[7] Katcoff, Dillard, Finston, Finkle, Seiler, and Sugarman, Paper No. 280, p. 1635 of reference 6.

TABLE I. Exchange tests.

	Counts/min per mg U		
	From metal solution only	From alkaline oxidation	Ratio
I^{135}	1068, 1065	1072, 1083	0.99
I^{133}	243, 242	244, 248	0.99
I^{131}	3.34, 3.39	3.45, 3.50	0.97

show that, as expected, exchange is essentially complete during the solution process.

Resolution of Decay Curves

Decay curves were resolved by fitting the experimental points with lines of the correct slope. The half-life values[1,3,8–12] used are given in the second column of Table II. They and the values obtained by drawing the best line through the points agree within the ~1 percent error estimated for the latter. The 20.9-hour value for I^{133} was determined by following the decay over about 10 half-lives on the beta-proportional counter through 145 mg/cm² of aluminum to minimize the I^{131} component. The value agrees well with the 20.8-hour value reported by Katcoff and Rubinson.[13]

The beta-decay curves were easily resolved into their 8.07-day I^{131} and 20.9-hour I^{133} components. In order to subtract the I^{131} and I^{133} components from the gamma-decay curves, the relative counting efficiencies of these nuclides on the beta-proportional and scintillation counters were determined by following the decay of four remounted samples on both counters. In calculating the curves to be subtracted, the different chemical yields of the original and remounted samples had to be considered.

The 2.3-hour I^{132} activity which had grown from 77.7-hour Te^{132} before the tellurium-iodine separation was also subtracted from the gamma-decay curve. This activity was calculated from the I^{132} activity in equilibrium with Te^{132} 113 hours after the irradiation (samples *F3-A* and *B*, *T3-A* and *B*).

TABLE II. Half-life values.

Isotope	Used in resolution	Obtained from data
I^{135}	6.7 hr[a,b]	6.75 hr
I^{134}	52.5 min[c]	52.4 min
I^{133}	20.9 hr	20.9 hr
I^{132}	2.3 hr[d,e]	2.30 hr
I^{131}	8.07 days[f,g]	8.0 days

[a] See reference 8.
[b] See reference 9.
[c] See reference 3.
[d] See reference 10.
[e] See reference 11.
[f] See reference 1.
[g] See reference 12.

The residual gamma-decay curve consisted of the 52.5-minute I^{134}, the independently formed 2.3-hour I^{132}, and the 6.7-hour I^{135} activities. The I^{135} component was easily resolved and subtracted, but the remaining mixture of I^{134} and independently formed I^{132} activities was difficult to resolve by the usual method of drawing a line with a 2.3-hour half-life through the oldest points. Therefore, a method suggested by Ford[14] was employed, in which the activity at time t, $A(t)$, multiplied by $e^{\lambda_{132}t}$ is plotted against $e^{-(\lambda_{134}-\lambda_{132})t}$ as shown in Fig. 1. As seen from Eq. (2), the slope of the straight line gives the value of A^0_{134}, and the intercept gives the value of A^0_{132}.

$$A(t)=A^0_{134}e^{-\lambda_{134}t}+A^0_{132}e^{-\lambda_{132}t}. \quad (1)$$

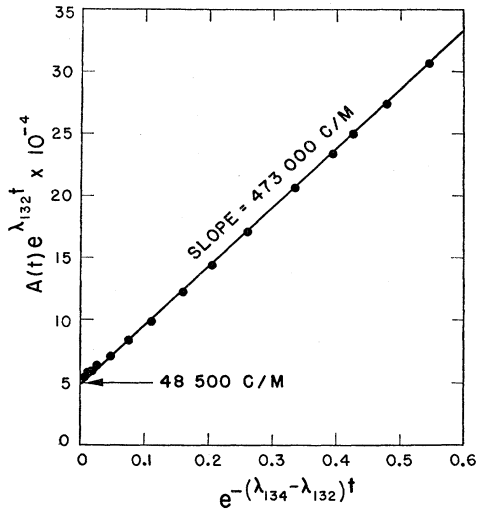

FIG. 1. Resolution of 52.5-min I^{134} and 2.3-hr I^{132} (independently formed) activities in sample *F2-A*. t is the time interval after the mean time of irradiation.

Division by $e^{-\lambda_{132}t}$ gives

$$A(t)e^{\lambda_{132}t}=A^0_{134}e^{-(\lambda_{134}-\lambda_{132})t}+A^0_{132}. \quad (2)$$

For thermal-neutron fission, there was no measurable amount of I^{132} formed independently, as was shown by the small positive and negative values of the intercepts. A^0_{134} for the samples from thermal-neutron fission was therefore determined from the slope of the best line passing through zero.

Typical resolved gamma- and beta-decay curves are reproduced in Figs. 2 and 3. A count was taken every 15 minutes for the first six hours, but due to space limitations, only the points at 30-minute intervals are shown in Fig. 2.

[8] L. E. Glendenin and R. P. Metcalf, Paper No. 140, p. 992 of reference 6.
[9] S. Katcoff, Dillard, Finston, Finkle, Seiler, and Sugarman, Paper No. 141, p. 1005 of reference 6.
[10] J. S. Gilmore (private communication from the Los Alamos Scientific Laboratory).
[11] O. Hahn and F. Strassmann, Naturwiss. **27**, 451 (1939).
[12] Selger, Cavallo, and Culpepper, Phys. Rev. **90**, 443 (1953).
[13] S. Katcoff and W. Rubinson, Phys. Rev. **91**, 1458 (1953).
[14] G. P. Ford (private communication from the Los Alamos Scientific Laboratory).

Data

The activity, corrected for chemical yield, of each iodine isotope at the time of the tellurium-iodine separation was divided by the Mo^{99} activity at the mean irradiation time.[15] This ratio, D, is proportional to the number of iodine atoms per fission that were present at the time of the tellurium-iodine separation. The proportionality constant is of course different for each iodine isotope because of different counting efficiencies and decay constants; it also is different for fission by thermal and 14-Mev neutrons since the Mo^{99} yield is different.[16] For independently formed I^{132} and for I^{135}, for which there is no precursor of measurable half-life, it was more convenient to extrapolate the activity to the mean time of irradiation[15] and divide by the Mo^{99} activity to give a ratio denoted by d.

The data are summarized in Table III. Uranium containing >90 percent U^{235} was used in all of the 14-Mev runs and in thermal run $T6$. Normal uranium was used in the other thermal-neutron runs. Approximately 3 g samples were used in runs $T1$ and $T3$ so that the chemistry would be the same as for the 14-Mev runs. Also these runs were timed to match approximately the 14-Mev runs.

It was not possible to maintain a constant flux of 14-Mev neutrons. For runs $F1$ and $F2$ the average flux over one-minute intervals deviated as much as 30 percent from the six-minute average, the mean deviation being 10 and 15 percent, respectively. For run $T1$ the power of the Fast Reactor was varied at one-minute intervals to match the fluctuation of flux in run $F1$. The matching of runs $F1$ and $T1$ was done to maximize the cancellation of errors in comparison of 14-Mev and thermal-neutron fission yields.

The assumption is made in the calculations that follow that the neutron flux was constant. Although this is only approximately true for the 14-Mev irradiations, the approximation causes no apprecialbe error. All of the iodine isotopes studied and most of their precursors have half-lives long compared to the irradiation times. For these the uniformity of the flux is not important. Since a large fraction of the two short-lived precursors, 4.4-m Sb^{133} and 2-m Te^{133}, decayed during the time interval between the end of the irradiation and the tellurium-iodine separation, it is not highly important when during the irradiation they were formed.

CALCULATIONS AND RESULTS

Equations and Definitions

The equations developed below are adopted from the standard equations of radioactive transformation. A useful general equation given by Rubinson[17] for the nth member of decay chain whose first member is formed at a constant rate, r, for time, T, is

$$\lambda_n N_n = r\sum_{i=1}^{n} b_i(1-e^{-\lambda_i T})(e^{-\lambda_i t}),$$
$$b_i = \prod_{j\neq i}\frac{\lambda_j}{\lambda_j-\lambda_i} \quad (j=1,2,\cdots,n), \tag{4}$$

where t = time after end of irradiation, N = number of atoms, and λ = decay constant.

Consider the following fission-product chain in which member C has a cumulative yield of Y_C, and x, y, and z are the fractional chain yields based on Y_C ($x+y+z=1$).

$$\begin{array}{ccccc} & x & & y & & z & \\ & \downarrow\lambda_A & & \downarrow\lambda_B & & \downarrow\lambda_C & \\ A & \rightarrow & B & \rightarrow & C & \rightarrow. \end{array}$$

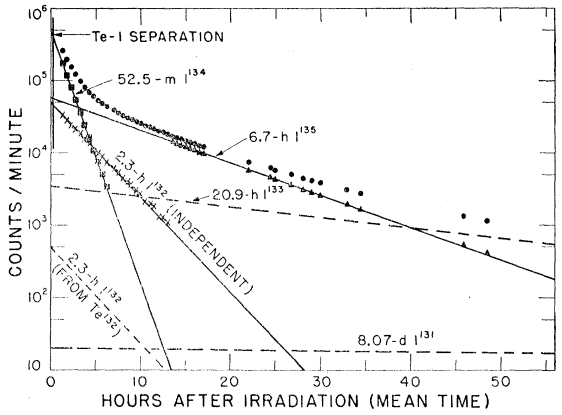

FIG. 2. Gamma-decay curve of sample $F2$-A followed on the scintillation counter through 11.7 g/cm² of Pb.

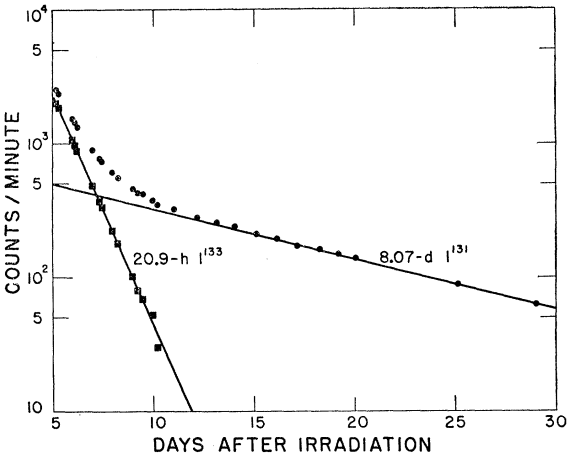

FIG. 3. Beta-decay curve of sample $F2$-A followed on the beta-proportional counter through 40 mg/cm² of Al.

[15] When the irradiation time T is short compared to the half-life of a product, the simplifying approximation can be made that $(1-e^{-\lambda T})/\lambda T = e^{-\lambda T/2}$, which corresponds physically to an infinitely short irradiation occurring at time $T/2$. If the irradiation time is $\frac{1}{5}$ of a half-life, the approximation is in error by less than 0.1 percent.

[16] Terrell, Scott, Gilmore, and Minkkinen, Los Alamos Scientific Report LADC-1463 (1953).

[17] W. Rubinson, J. Chem. Phys. **17**, 542 (1949).

TABLE III. Summary of data.[a]

Sample No.	T	t	$d_{I^{135}}$	$D_{I^{134}}$	$D_{I^{133}}$	$D_{I^{132}}$ (from Te^{132})	$d_{I^{132}}$ (independent)	$D_{I^{131}}$
14-Mev-neutron fission								
F1-A	6.00 min	8.60 min	0.332	2.457	0.840	(0.0026)	0.284	0.0053
F1-B	6.00 min	125.0 min	(0.335)	1.394	1.722	(0.0221)	0.316	0.0290
F2-A	6.00 min	9.10 min	0.333	2.384	0.895	(0.0028)	0.287	0.0056
F2-B	6.00 min	125.3 min	0.339	1.372	1.804	(0.0222)	0.283	0.0302
F3-A	75 min	112.8 hr	···	···	0.0532	0.01711	···	0.0379
F3-B	75 min	112.8 hr	···	···	0.0513	0.01683	···	0.0341
Thermal-neutron fission								
T1-A	6.00 min	8.60 min	0.414	1.619	0.581	(0.0023)	0.016	0.00142
T1-B	6.00 min	125.0 min	0.412	2.137	1.799	(0.0193)	−0.014	0.0236
T3-A	60 min	112.5 hr	···	···	0.0548	0.01484	···	0.0207
T3-B	60 min	112.5 hr	···	···	0.0542	0.01485	···	0.0200
T4-A	0.17 min	1.83 min	0.413	0.837	0.109	(0.0004)	0.013	<0.0005
T4-B	0.17 min	120.6 min	0.417	2.245	1.811	(0.0185)	−0.012	0.0237
T5-A	0.42 min	2.20 min	0.411	0.896	0.139	(0.0005)	0.012	<0.0003
T5-B	0.42 min	4.45 min	0.404	1.044	0.250	(0.0009)	0.005	<0.0006
T5-C	0.42 min	181.8 min	0.400	1.244	1.846	(0.0244)	−0.016	0.0271
T6-A	0.25 min	8.50 min	···	···	0.457	···	···	0.00096
T6-B	0.25 min	15.75 min	···	···	0.726	···	···	0.00249

[a] T = duration of irradiation. t = time interval between end of irradiation and tellurium-iodine separation. d = ratio of iodine to Mo^{99} activity at mean irradiation time, activities corrected for chemical yield. D = ratio of iodine activity at time of tellurium-iodine separation to Mo^{99} activity at mean irradiation time, activities corrected for chemical yield. ( ) = value calculated from other data and used in resolution of the decay curve.

If a total of G fissions occur at a uniform rate during irradiation time T, the activity of C at time t after the irradiation is given by Eq. (5).[15]

$$\lambda_C N_C = (GY_C/T)\{x(aJ+bK+cL) + y(\bar{b}K+\bar{c}L)+zL\}, \quad (5)$$

$$J=(1-e^{-\lambda_A T})(e^{-\lambda_A t}),$$
$$K=(1-e^{-\lambda_B T})(e^{-\lambda_B t}),$$
$$L=(1-e^{-\lambda_C T})(e^{-\lambda_C t}),$$
$$a=\lambda_B\lambda_C/(\lambda_B-\lambda_A)(\lambda_C-\lambda_A),$$
$$b=\lambda_A\lambda_C/(\lambda_A-\lambda_B)(\lambda_C-\lambda_B),$$
$$c=\lambda_A\lambda_B/(\lambda_A-\lambda_C)(\lambda_B-\lambda_C),$$
$$\bar{b}=\lambda_C/(\lambda_C-\lambda_B),$$
$$\bar{c}=\lambda_B/(\lambda_B-\lambda_C).$$

The first term gives the activity of C that has grown from A, the second term gives the activity of C that has grown from independently formed B, and the third term gives the activity of independently formed C. If chain member C is identified with an iodine isotope, $\lambda_C N_C/G$ is proportional to D, the ratio of iodine to Mo^{99} activity.

$$\frac{\lambda_C N_C}{G}=\left(\frac{E_{Mo}}{E_C}\lambda_{Mo}Y_{Mo}\right)D, \quad (6)$$

$$D=\left(\frac{E_C Y_C}{E_{Mo}Y_{Mo}\lambda_{Mo}T}\right)\{x(aJ+bK+cL) + y(\bar{b}K+\bar{c}L)+zL\}, \quad (7)$$

where E represents the counting efficiency.

For two samples irradiated together, so that T is the same for both, but separated at different times t and t', the ratio of the activities of the samples at their respective separation times and normalized to the same number of fissions is

$$M=\frac{D'}{D}=\frac{\lambda_C N_C'/G'}{\lambda_C N_C/G} =\frac{x(aJ'+bK'+cL')+y(\bar{b}K'+\bar{c}L')+zL'}{x(aJ+bK+cL)+y(\bar{b}K+\bar{c}L)+zL}. \quad (8)$$

Solution of Eq. (8) for z gives

$$z=-\frac{(y\bar{c}+xc)(ML-L')+(y\bar{b}+xb)(MK-K')+ax(MJ-J')}{(ML-L')}. \quad (9)$$

For the purpose of calculating the relative cumulative yields of an iodine isotope from 14-Mev and thermal-neutron fission it is convenient to define a function D^0.

$$D^0=\left(\frac{E_C\lambda_C}{E_{Mo}\lambda_{Mo}}\right)\left(\frac{Y_C}{Y_{Mo}}\right) =\frac{DT\lambda_C}{x(aJ+bK+cL)+y(\bar{b}K+\bar{c}L)+zL}, \quad (10)$$

$$R=\frac{D^0_{14\text{–Mev}}}{D^0_{\text{thermal}}}=\frac{(Y_C/Y_{Mo})_{14\text{–Mev}}}{(Y_C/Y_{Mo})_{\text{thermal}}}. \quad (11)$$

An error listed for an average value obtained in the following calculations is the standard deviation of the average value. Where there is uncertainty in the mode of formation of a nuclide, e.g., an unknown branching ratio of a precursor, calculations using limiting values

have been made, and the error has been increased to include this uncertainty.

Iodine-135

As can be seen in Table III, there is no detectable dependence of d on the separation time, showing that the half-life of the precursor of I^{135} is short compared to the separation times. For the d values of samples $T4$-A and $T5$-A to be within 2 percent of the other d values, as they are, more than 98 percent of a precursor must have decayed before the separation 2 minutes after the irradiation. The half-life of a precursor would have to be <0.4 minute for this to occur. Previous work[8,9,18] had shown that if Te^{135} is the precursor of I^{135} its half-life is less than 1 or 2 minutes. The ~0.8-minute antimony activity found in fission by Pappas[3,4] and assigned by him to mass number 134 or 135 must either not be Sb^{135}, or if it is, less than about one tenth of the I^{135} formed in fission comes from it.

An R value of 0.815±0.020 is obtained as the ratio of the average d values for 14-Mev and thermal-neutron fission.

Iodine-134

$$\begin{array}{ccccccc} x & & y & & z & & \\ \downarrow & & \downarrow & & \downarrow & & \\ Sb^{134} & \rightarrow & Te^{134} & \rightarrow & I^{134} & \rightarrow & \\ (\sim 0.8\ \text{min?})^{3} & & (44.5\ \text{min})^{4} & & (52.5\ \text{min})^{3} & & \end{array}$$

The results of the calculations for I^{134} are summarized in Table IV. Since it is not certain that the ~0.8-minute antimony is the parent of Te^{134} and since the fraction of the 134 chain formed at antimony and at tellurium is not known, calculations were made using Eq. (9) for the two extreme conditions, $y=0$ and $x=0$. As seen, the difference between the results of the two calculations is not large. A D^0 value listed for a pair of samples was calculated by using the z value calculated for the pair and the data for the sample with the greater time before separation.

In the samples that were separated soon after the irradiation, a large fraction of the I^{134} activity was due to independent formation of I^{134}. This fraction varied from 43 percent in $T1$-A to 87 percent in $T4$-A, and was 80 percent in $F1$-A and $F2$-A. Since the correction for growth from Te^{134} is not large, the fractional chain yields should be reliable. Glendenin[2] had previously determined the independent yield of I^{134} from thermal-neutron fission of U^{235}. His value as recalculated by Pappas[3] is (0.94±0.13) percent. This, divided by the (8.00±0.21) percent cumulative fission yield of I^{134} determined by Pappas,[3] gives 0.12±0.02 for the fractional chain yield of I^{134}, in good agreement with the value determined in this work.

TABLE IV. Relative yields of I^{134}.

Neutron energy	Samples	z_{max} ($y=0$)	z_{min} ($x=0$)	Selected z	D^0	R
14-Mev	$F1$-A and B	0.44	0.43	0.43 ±0.02	5.36	5.31 ±0.06
	$F2$-A and B	0.43	0.42		5.27	
						0.77 ±0.22
Thermal	$T1$-A and B	0.125	0.104	0.11 ±0.02	7.06	6.91 ±0.19
	$T4$-A and B	0.110	0.093		7.04	
	$T5$-A or B and C	0.127	0.111		6.38[a]	

[a] Given only half-weight in the average because t was an hour greater than for the other samples, so that growth and decay corrections were quite different. Therefore, less cancellation of half-life errors in the calculation of R was to be expected.

[18] R. W. Dodson and R. D. Fowler, Phys. Rev. **57**, 966 (1940).

Iodine-133

The formation scheme for I^{133} is shown in Fig. 4. Pappas,[3] from estimates of absolute fission yields, gives 0.92/0.08=(1)/(2) for the value of the branching ratio of Sb^{133}. This ratio is too high as will be shown in this section. Pappas[3] also determined for thermal-neutron fission the sum of the independent yields of I^{133} and the 2-min Te^{133}. His value and the values he recalculated from the data of Glendenin[2] and Katcoff *et al.*[9] are in the range of 20 to 30 percent of the total chain yield. These values are also too large because the fraction of Sb^{133} decaying by path 2 is larger than Pappas estimated.

The paths shown in Fig. 5 are considered. For the purpose of the calculations all paths through Te^{133m} are lumped into path 1 as an average. Equation (10) modified to include path 1 is given below.

$$D^0=\frac{DT\lambda_C}{(x_1+y')(a'J+b'K'+c'L)+x_2(aJ+bK+cL)+y(\bar{b}K+\bar{c}L)+zL}. \quad (12)$$

The terms a', b', c', and K' are the same as a, b, c, and K with $\lambda_{B'}$ substituted for λ_B.

The D values of the samples $F3$-A and B and $T3$-A and B separated after all the precursors of I^{133} had decayed can be used to calculate the values of D^0. There is about a 5 percent uncertainty in the results, depending on whether the I^{133} was formed rapidly via the short-lived paths 2, 3, and 4 or was held up at Te^{133m}. As a first approximation it was assumed that half was held up and half was not ($x_1+y'=0.5$, $z=0.5$). The

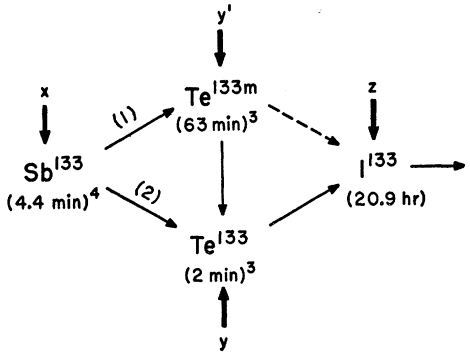

FIG. 4. Formation of I^{133}.

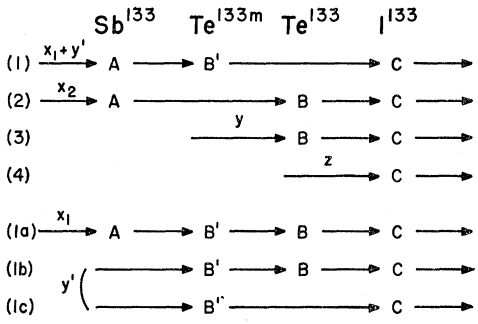

FIG. 5. Paths leading to the formation of I^{133}.

average D^0 values so obtained (2.19 and 2.25 for 14-Mev and thermal-neutron fission, respectively) were then used with the D values of the samples separated as soon as possible after the irradiation to calculate upper limits for $y+z$ ($x_1+x_2+y'=0$ assumed). These limits were 0.40 and 0.10 for 14-Mev and thermal-neutron fission, respectively.

The above information was then used to calculate the fraction of I^{133} formed from Te^{133m} (path 1). Data were used from samples separated 8 to 16 minutes after the end of the irradiation when most of the 4.4-min Sb^{133} and 2-min Te^{133}, but little of the 63-min Te^{133m}, had decayed. Two calculations were made for each sample, one for $y+z=0$ and one for $y+z$ equal to the upper limit calculated above. The average of the calculated limits are listed in Table V; the error for each value shows the spread of the limits.

The limits of 0.70 to 0.74 for the fraction of I^{133} formed from Te^{133m} in thermal-neutron fission were used in Eq. (12) to recalculate the limits on y and z. The results of these calculations are summarized in Table VI.

Finally, the values of D^0 were recalculated by using the determined limits on x_1+y'. The data used were from samples separated two hours or more after the irradiation; paths 2, 3, and 4 were therefore essentially equivalent. Two calculations were made for each sample, one for each x_1+y' limit. The average of the two limiting values are listed in Table VII; the errors show the spread of the limits.

The principal uncertainties in the reliability of the values calculated in this section, not included in the listed errors, are (1) the uncertainty in the separation time, 15 to 30 second maximum error, and (2) the error introduced in the approximation that path 1 adequately

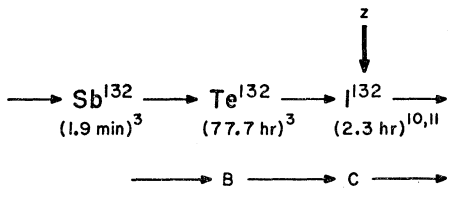

FIG. 6. Formation of I^{132}.

TABLE V. Fraction of I^{133} formed from Te^{133m}.

Neutron energy	Sample	x_1+y'
14-Mev	*F1-A*	0.59±0.08
	F2-A	0.56±0.08 } 0.57±0.11
Thermal	*T1-A*	0.72±0.02
	T6-A	0.73±0.04 } 0.72±0.02
	T6-B	0.71±0.01

represent all possible paths, 1, 1*a*, 1*b*, and 1*c*, through Te^{133m}. A 30 second error in the separation time would change the value of x_1+y' by about 0.02, z_{max} by about 0.01, $(y+z)_{max}$ by about 0.03, $(y+z)_{min}$ by about 0.01, and D^0 not at all. If path 1*a* is the only important path through Te^{133m}, limits on x_1 would be 0.03 to 0.04 lower than those calculated for x_1+y'. If path 1*c* is the important path, y' limits would be 0.04 to 0.07 higher than those calculated for x_1+y'. The values of D^0 and the upper limits on z and $(y+z)$ would be affected very little by use of path 1*a* or 1*c* instead of path 1 in the calculations. The minimum value of $(y+z)$ would be reduced to ~ 0 if path 1*c* were the important one. The calculated minimum value for $(y+z)$ of 0.02 may be due entirely to uncertainties in timing and chain path.

Iodine-132

The formation scheme for I^{132} is shown in Fig. 6.

In the samples that were allowed to stand 113 hours after irradiation before the tellurium-iodine separation, the I^{132} was in transient equilibrium with Te^{132}.

$$\lambda_C N_C = (\lambda_B N_B{}^0)\frac{\lambda_C}{\lambda_C-\lambda_B}e^{-\lambda_B\tau}, \tag{13}$$

where $\tau=t+T/2$. Division of both sides of Eq. (13) by the Mo^{99} activity at irradiation time gives

$$D=D_B{}^0\frac{\lambda_C}{\lambda_C-\lambda_B}e^{-\lambda_B\tau}. \tag{14}$$

$D_B{}^0$ is proportional to the ratio of Te^{132} and Mo^{99} cumulative fission yields.

$$D_B{}^0=\left(\frac{E_C}{E_{Mo}}\frac{\lambda_B}{\lambda_{Mo}}\right)\left(\frac{Y_B}{Y_{Mo}}\right). \tag{15}$$

TABLE VI. Independent formation of I^{133} and 2-min Te^{133} in thermal neutron-fission.

Sample	z_{max}	$(y+z)_{max}$	$(y+z)_{min}$
T4-A	0.03	0.07	0.03
T5-A	0.04	0.07	0.03
T5-B	0.04	0.05	0.02
	0.04	0.07	0.02

TABLE VII. Relative yields of I^{133}.

Neutron energy	Sample	D^0		R
14-Mev	*F1-B*	2.14±0.07	2.19+0.04	0.96±0.03
	F2-B	2.25±0.07		
	F3-A	2.21±0.01		
	F3-B	2.14±0.01		
Thermal	*T1-B*	2.33±0.22	2.28±0.05	
	T4-B	2.40±0.02		
	T5-C	2.22±0.01		
	T3-A	2.24±0.00		
	T3-B	2.21±0.00		

It has the same form as D^0, except that the decay constant and yield of tellurium replace those of iodine; the counting efficiency of the iodine radiation, which is measured, is retained.

The relative yields of Te^{132} are shown in Table VIII. The R value for Te^{132} is in agreement with the value of 1.13±0.03 obtained by Gilmore.[10]

For the other samples, which were separated a few hours or less after the irradiation, the activity of the I^{132} that had grown from Te^{132} was calculated from the $D_B{}^0$ values given above and subtracted from the gross decay curves. Any 2.3-hour period remaining was attributed to independently formed I^{132}. The independent yield of I^{132} relative to the cumulative yield of Te^{132} is given by

$$\frac{z}{1-z}=\left(\frac{d}{D_B{}^0}\right)\left(\frac{2.3\text{ hr}}{77.7\text{ hr}}\right), \tag{16}$$

and the results are listed in Table IX.

From 14-Mev-neutron fission, the activity of independently formed I^{132} was as a maximum between 15 and 20 percent of the total iodine activity; from thermal-neutron fission it was <1.5 percent of the total. If, as seems reasonable, the reliability of a resolved component of a decay curve is taken to be about 1 percent of the total activity, the ±0.01 error placed on the independent yields of I^{132} is a good indication of the reliability of the values.

An R value for I^{132} of 1.33±0.03 is obtained by multiplying the R value for Te^{132} by 1.16.

Iodine-131

The formation scheme for I^{131} is shown in Fig. 7. The paths shown in Fig. 8 are considered. For the purpose of the calculation all paths through Te^{131m} are lumped into path 1 as an average.

TABLE VIII. Relative yields of Te^{132}.

Neutron energy	Sample	$D_B{}^0$		R
14-Mev	*F3-A*	0.0457	0.0453±0.006	1.15±0.02
	F3-B	0.0449		
Thermal	*T3-A*	0.0395	0.0395	
	T3-B	0.0395		

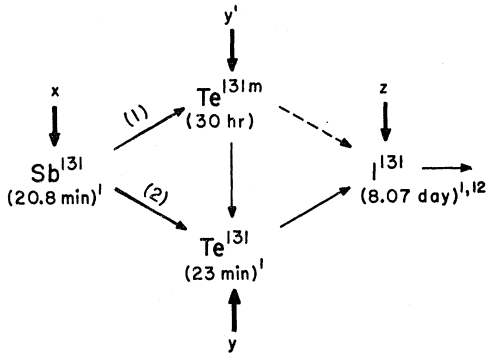

FIG. 7. Formation of I^{131}.

Calculations were made using Eq. (12) by two series of approximations similar to the method used for the 133-chain. In the first approximation, D^0 was calculated from the data of the samples separated 113 hours after the irradiation, with the use of the assumption that $x_1+y'=0.5$, which introduced a 3 percent uncertainty in the calculations. The values obtained, 0.0527 and 0.0297 for 14-Mev and thermal-neutron fission, respectively, were then used with the data from the samples separated shortly after irradiation to calculate upper limits for y and z. These limits, $z<0.10$ and <0.01, and $(y+z)<0.35$ and <0.15 for 14-Mev and thermal neutron fission, respectively, were then used to calculate the fraction of I^{131} formed from Te^{131m}. The results were $x_1+y'=0.41\pm0.03$ and 0.11 ± 0.04 for 14-Mev and thermal-neutron fission, respectively.

The results of the second series of calculations are summarized in Tables X, XI, and XII. For D^0 and x_1+y' two calculations were made for each sample, one for each of the possible limiting values for the other fractional chain yields. The average of the two calculations are listed along with an error indicating the spread.

For the 131 chain, whose members have half-lives of >20 minutes, the <0.5-minute timing error is not significant. The approximation that path 1 adequately

TABLE IX. Independent yield of I^{132}.

Neutron energy	Sample	$\frac{z}{1-z}$		z
14-Mev	*F1-A*	0.186	0.191+0.006	0.16±0.01
	F1-B	0.206		
	F2-A	0.188		
	F2-B	0.185		
Thermal	*T1-A*	0.012	0.001±0.004	<0.01
	T1-B	−0.010		
	T4-A	0.010		
	T4-B	−0.009		
	T5-A	0.009		
	T5-B	0.004		
	T5-C	−0.012		

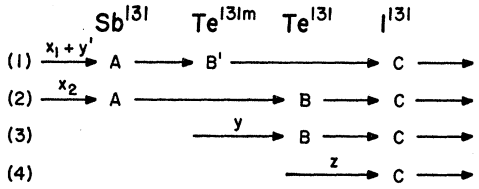

FIG. 8. Paths leading to the formation of I^{131}.

represents all paths through Te^{131m} is a good one and introduces no appreciable uncertainty in the values of the constants calculated. For samples separated a few hours or less after the irradiation, the contribution from any path through Te^{131m} is small, and it makes little difference which is used; for the samples separated 113 hours after an irradiation all paths through Te^{131m} lead to equivalent results.

There is reasonable agreement between the R value for I^{131} determined in this work and the value of 1.70 $\pm$0.03 determined by Ford and Gilmore.[1] However, Ford and Gilmore assumed that the same fraction of I^{131} was formed from Te^{131m} in 14-Mev and thermal-neutron fission. This is not the case, and if a correction is made, Ford and Gilmore's value is lowered to 1.61.

There is also reasonable agreement between the value of 0.11$\pm$0.03 for the fraction of I^{131} formed from Te^{131m} in thermal-neutron fission and the value of 0.14 reported by Pappas.[3] The branching ratio of Sb^{131} of 0.15/0.85 = (1)/(2), also reported by Pappas, more than accounts for all of the Te^{131m} formed in thermal neutron fission, so the independent yield of Te^{131m} must be small, probably <5 percent of the total chain yield. However, in 14-Mev-neutron fission the yield of Te^{131m} is high, 41$\pm$2 percent of the cumulative I^{131} yield. Since only 15 percent of Sb^{131} decays to Te^{131m}, the independent yield of Te^{131m} in 14-Mev-neutron fission must be large.

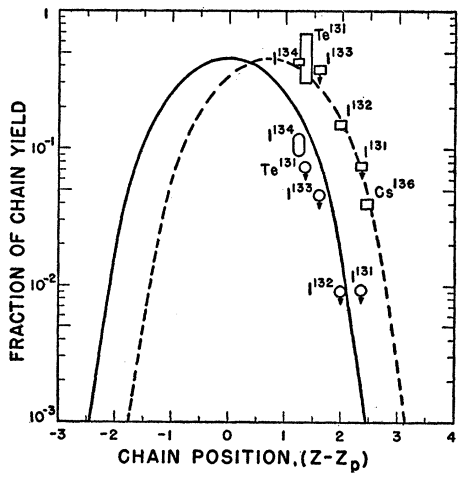

FIG. 9. Variation of yield with nuclear charge. The circles and ovals refer to thermal-neutron fission, the rectangles to 14-Mev neutron fission. The vertical arrows indicate upper limits. Except for Cs^{136} (reference 24), fractional chain yields are based on cumulative yields at iodine.

TABLE X. Relative yields of I^{131}.

Neutron energy	Sample	D^0	R
14-Mev	*F1-B*	51.8±3.5	1.76±0.08
	F2-B	53.9±3.7	
	F3-A	55.6±0.1	52.9±2.1
	F3-B	50.1±0.1	
Thermal	*T1-B*	29.0±1.5	
	T4-B	29.6±1.5	
	T5-C	31.2±1.4	30.1±0.7
	T3-A	30.8±0.1	
	T3-B	29.8±0.1	

The calculated value is 35$\pm$5 percent of the cumulative I^{131} yield.

DISCUSSION

Independent Fission Yields

The fractional chain yields determined in this research are plotted in Fig. 9 as a function of their chain position. The function used is $(Z-Z_P)$ in which Z_P is the most probable initial charge of the chain calculated according to the postulate[19] that the most probable mode of nuclear charge division is one that leads to equal idealized chain lengths (Z_A-Z_P) for both primary fragments.

$$Z_P=Z_A-[Z_A+Z_{(236-A)}-92]/2. \qquad (17)$$

The Z_P values were taken from the tabulation of Pappas,[3] who assumed that one neutron was emitted from each fragment. He obtained the Z_A values from the lines of maximum stability given by Coryell, Brightsen, and Pappas.[20] Those lines are discontinuous at shell edges.

The solid curve is the one that has been found[3] to best fit other thermal-neutron data. The thermal-neutron points reported in this paper generally fall below this line. The I^{134} and I^{133} points might be expected to fall low because of the abnormally large 134- and 133-chain yields, presumably due to a preference for

TABLE XI. Independent yields of I^{131} and 23-min Te^{131}.

Neutron energy	Sample	z_{max}	$(y+z)_{max}$	$(y+z)_{min}$
14-Mev	*F1-A*	0.08	0.29	0.07
	F2-A	0.08	0.30	0.08
		0.08	0.30	0.07
Thermal	*T1-A*	0.02	0.07	0.00
	T4-A	<0.02	<0.31	···
	T5-A	<0.01	<0.13	···
	T5-B	<0.01	<0.07	···
	T6-A	0.01	0.03	0.00
	T6-B	0.01	0.02	0.00
		0.01	0.03	0.00

[19] Glendenin, Coryell, and Edwards, Paper No. 52, p. 489, of reference 6.

[20] Coryell, Brightsen, and Pappas, Phys. Rev. 85, 732 (1952).

TABLE XII. Fraction of I^{131} formed from Te^{131m}.

Neutron energy	Sample	x_1+y'	
14-Mev	*F1-B*	0.42±0.02	0.41±0.02
	F2-B	0.40±0.01	
Thermal	*T1-B*	0.14	0.11±0.03
	T4-B	0.12	
	T5-B	0.08	

closed-shell configuration in fission[21,22] and the excessive evaporation from primary fission fragments of the loosely bound neutrons just outside closed shells.[2,3,23] If the fractional chain yields are calculated on the basis of the smooth mass-yield curve, the I^{134} and I^{133} points would be raised 40 and 30 percent, respectively, to bring them near the line. In addition, the independent formation of I^{134} may be less than that predicted from charge-displacement considerations because I^{135}, which has 82 neutrons, may emit fewer prompt neutrons than most other primary fragments.

No such explanation exists for Te^{131}. To bring it up to the line would require that most of the 0.11±0.03 cumulative fractional chain yield of Te^{131m} be attributed to independent formation, which in turn would require the branching ratio of Sb^{131} to be much less than the 0.15/0.85 reported by Pappas.[3]

The independent yields from 14-Mev-neutron fission, including the yield of Cs^{136} determined by Comstock,[24] all fall considerably above the solid curve. The dashed curve, which is the solid curve displaced by 0.7 charge units, fits the points well. The I^{134} point is a little high, but it would be brought onto the dashed curve if the fractional yield were based on the total chain yield rather than the cumulative yield at iodine, since, as estimated from the dashed curve, about 12 percent of the 134 chain is formed at Xe^{134}.

A possible explanation for the apparent shift in Z_P toward stability in 14-Mev neutron fission is that more neutrons are boiled off. Since in the mass region 130 to 138 the slope[3] of Z_P/A is 0.37, the ~0.7 shift in Z_P would correspond to the loss of approximately two more neutrons from the heavy fragment in 14-Mev-neutron fission than in thermal-neutron fission. However, there is little evidence for the assumption inherent in the explanation that the nuclear charge distribution for primary fragments is the same in 14-Mev and thermal-neutron fission.

TABLE XIII. Yields from 14-Mev neutron fission.

Isotope	$R\left(\frac{5.17}{6.14}\right)$	Thermal yield,[a] %	14-Mev yield, %	Estimated fraction of total chain yield	Estimated total chain yield, %
I^{131}	1.48	3.02	4.47	1.00	4.5
I^{132}	1.12	4.49	5.03	1.00	5.0
I^{133}	0.81	6.62	5.36	0.97	5.5
I^{134}	0.65	8.00	5.20	0.90	5.8
I^{135}	0.69	6.31	4.35	0.79	5.5

[a] See reference 3.

[21] Wiles, Smith, Horsley, and Thode, Can. J. Phys. **31**, 419 (1953).
[22] Glendenin, Steinberg, Ingram, and Hess, Phys. Rev. **84**, 860 (1951).
[23] L. E. Glendenin, Phys. Rev. **75**, 337 (1949).
[24] A. Comstock, reported by G. P. Ford in U. S. Atomic Energy Commission Report AECD-3597, 1953 (unpublished).

Yields from 14-Mev-Neutron Fission

In Table XIII are given the ratios of 14-Mev to thermal-neutron fission yields. These were obtained by multiplying the R values by 5.17/6.14, the ratio of Mo^{99} yields in 14-Mev and thermal-neutron fission determined by Terrell, Scott, Gilmore, and Minkkinen.[16] This allowed calculation of absolute 14-Mev-neutron fission yields from known thermal-neutron fission yields. The total chain yields were estimated from the charge distribution curve for 14-Mev-neutron fission (dashed curve in Fig. 9).

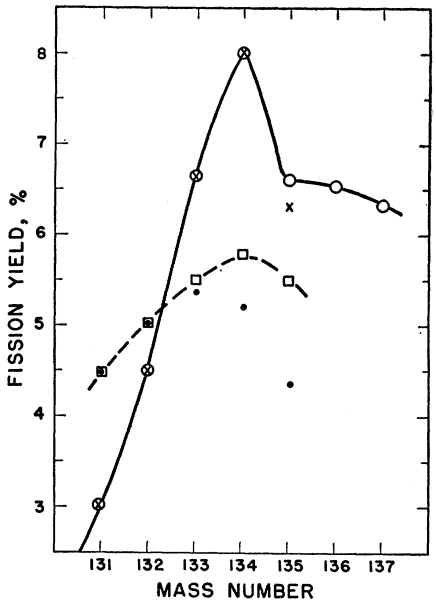

FIG. 10. Mass-yield curves. 14-Mev-neutron fission: ● iodine cumulative yield, □ total chain yield (estimated). Thermal-neutron fission: × iodine cumulative yield,[3] ○ total chain yield.[3]

In Fig. 10 are plotted both the 14-Mev and thermal neutron yields *vs* mass number. It may be seen that the prominent fine structure in thermal neutron fission is pretty well washed out at the higher energy.

ACKNOWLEDGMENTS

It is a pleasure to thank Dr. R. W. Spence and the members of his group for their hospitality and their interest and assistance in this work. I also wish to thank the groups responsible for the operation of the Cockcroft-Walton accelerator and the homogeneous and fast reactors for the irradiations they provided.