

## Fission Yields of Several Iodine Isotopes and Half-Life and Fission Yield of $^{135}\text{Te}$ †

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Measurements have been made of the independent fission yields of several iodine isotopes in thermal-neutron-induced fission of  $^{235}\text{U}$ . These yields were obtained by isolation of iodine-fission products at early times after fission by a new, fast radiochemical procedure. The fractional cumulative fission yield and half-life of  $^{135}\text{Te}$  were also determined from the experimental results. Fission yields measured are:  $^{131}\text{I}$ ,  $(4.2 \pm 0.4) \times 10^{-3}\%$  independent;  $^{132}\text{I}$ ,  $(1.74 \pm 0.11) \times 10^{-2}\%$  independent;  $^{133}\text{I}$ ,  $0.176 \pm 0.009\%$  independent;  $^{134}\text{I}$ ,  $0.90 \pm 0.08\%$  independent;  $^{135}\text{I}$ ,  $0.44 \pm 0.07$  fractional independent;  $^{135}\text{Te}$ ,  $0.52 \pm 0.07$  fractional cumulative. The half-life of  $^{135}\text{Te}$  was determined to be  $11.2 \pm 2.6$  sec.

### I. INTRODUCTION

THE purpose of this work is to extend the information on the distribution of nuclear charge in thermal-neutron-induced fission of  $^{235}\text{U}$  in the region of the 50-proton and 82-neutron closed shells. Isobaric-yield-distribution curves have previously been reported<sup>1</sup> from this laboratory for several chains in this region. The yields of individual closed-shell nuclides such as  $^{132}\text{Sn}$  and  $^{133}\text{Sb}$  showed no discernible enhancement over the yields of adjacent isobars; such enhancement had been predicted<sup>2-4</sup> and reported<sup>5</sup> by others. The only significant effect noted in this region was a decrease in the widths of the isobaric-yield curves with increasing mass number for the mass-131, -132, and -133 chains.<sup>1</sup>

For better definition of the yield distributions in these chains and in adjacent chains, the independent yields of several iodine isotopes have been measured. Such independent yields have not been previously measured, primarily due to the lack of a sufficiently fast iodine radiochemical procedure. For  $^{131}\text{I}$ ,  $^{133}\text{I}$ , and  $^{135}\text{I}$  a fast iodine procedure is needed to reduce the contribution from precursor tellurium. With  $^{132}\text{I}$ , earlier attempts to measure the independent yield have been unsuccessful because of the difficulty of assaying  $^{132}\text{I}$  in the presence of much larger amounts of interfering iodine radioisotopes. This difficulty has been overcome in these experiments by  $\gamma$ - $\gamma$  coincidence measurement of the  $^{132}\text{I}$  with a Compton-suppressed detector.

### II. EXPERIMENTAL

#### A. Irradiations

Irradiations were carried out in the 30-kW *t* reactor (NTR) of the General Electric Co. at Vallecitos, Calif. The thermal neutron flux in the region of the bombardment was approximately  $5 \times 10^{14}$  *n*/cm<sup>2</sup> sec and the cadmium ratio for fission of  $^{235}\text{U}$  was determined to be 14.6 under the conditions of the experiment. A gold foil was present on each rabbit as a flux monitor. The  $^{198}\text{Au}$  activity resulting from these irradiations was related to the number of fissions occurring in the target samples by a series of radiochemical determinations of  $^{99}\text{Mo}$  found in identical samples. The cumulative fission yield of  $^{99}\text{Mo}$  resulting from the thermal-neutron-induced fission of  $^{235}\text{U}$  was taken to be 6.25%.<sup>6</sup>

#### B. Chemistry

The iodine separations were made by a new, fast radiochemical procedure,<sup>7</sup> which permits the isolation of iodine fission products within 5 sec following an irradiation. Only a brief description of the procedure will be given: The sample to be irradiated was prepared by dissolution of uranium peroxide in periodic acid at 135°C in a weight ratio of 1:14, respectively. The clear melt was cooled and ground to a powder. For each sample, 600 mg of this powder was mixed with 90 mg of activated carbon and sealed into a thin-walled graphite capsule.

The capsule was irradiated in the reactor for a period of 10 sec at the above flux. After the irradiation, the capsule entered a reaction vessel (Fig. 1) where, upon being heated, the periodic acid decomposed to release gaseous iodine. The iodine passed through a heated sintered glass frit and was collected on a column of glass beads wet with  $\text{CCl}_4$ . The sample was washed off the glass beads with rinses of  $\text{CCl}_4$  and collected. The separation time was taken as the time from the

\* Work supported by the U. S. Atomic Energy Commission.

<sup>1</sup> P. O. Strom, D. L. Love, A. E. Greendale, A. A. Delucchi, D. Sam, and N. E. Ballou, *Phys. Rev.* **144**, 984 (1966); *Naval Radiological Defense Laboratory Report No. USNRDL-TR-935*, 1965 (unpublished).

<sup>2</sup> P. Armbruster, in *Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, 1965* (International Atomic Energy Agency, Vienna, 1965).

<sup>3</sup> H. Greisner and K. Wildermuth, *Phys. Letters* **2**, 212 (1962).

<sup>4</sup> D. M. Wiles, B. W. Smith, R. Horsley, and H. G. Thode, *Can. J. Phys.* **31**, 419 (1953).

<sup>5</sup> E. Konecny, H. Opower, H. Gunther, and H. Goebel, in *Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, 1965* (International Atomic Energy Agency, Vienna, 1965).

<sup>6</sup> H. Farrar and R. H. Tomlinson, *Nucl. Phys.* **34**, 367 (1962).

<sup>7</sup> A. E. Greendale, D. L. Love, and A. A. Delucchi, *Ann. Chim. Acta* **34**, 32 (1966); *Naval Radiological Defense Laboratory Report No. USNRDL-TR-831*, 1965 (unpublished).

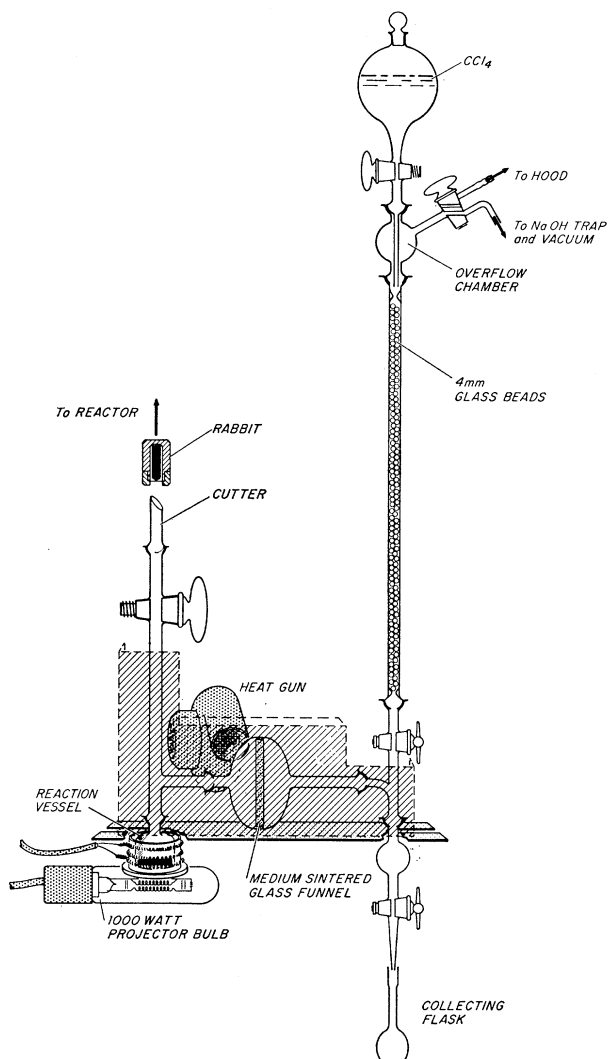


Fig. 1. Iodine-separation apparatus.

end of irradiation to the time that the capsule ruptured, releasing the iodine. With this procedure, we were able to separate iodine fission products from their precursors as early as 5 sec after the end of irradiation.

The iodine was extracted from the  $\text{CCl}_4$  into water containing sodium bisulfite. This solution was transferred to a Lucite container of about 4 ml volume in the shape of a short cylinder for counting measurements. The iodine chemical yield for each run was determined by gravimetric measurement of an  $\text{AgI}$  precipitate.

### C. Counting

In a source of iodine fission products separated at early times following irradiation, there is a considerable activity due to the 52.8-min  $^{134}\text{I}$  and 6.75-h  $^{135}\text{I}$ . Interferences from these nuclides make the measurement of the low-yield 2.29-h  $^{132}\text{I}$  very difficult. For assay of

the  $^{132}\text{I}$  it was necessary to do two-parameter  $\gamma$ - $\gamma$  coincidence counting with a Compton-suppressed detector assembly. It was convenient to use these same coincidence measurements also to assay the  $^{134}\text{I}$  and the  $^{135}\text{I}$ .

The coincidence detector that was used for the measurements<sup>8</sup> was designed to reduce as many interferences as possible caused by Compton interactions. The sample rests between two identical, large  $\text{NaI}(\text{Tl})$  assemblies. Each assembly consists of a central  $6\frac{1}{2}$  in. diam by  $4\frac{1}{2}$ -in.-high  $\text{NaI}(\text{Tl})$  crystal viewed by a single 5-in. photomultiplier. Surrounding this crystal is an annular  $\text{NaI}(\text{Tl})$  crystal with a 13 in. diam, which is viewed by six 3-in. photomultipliers. Coincidence measurements were obtained with this detector system by acceptance of coincidence events from the two central crystals which were not

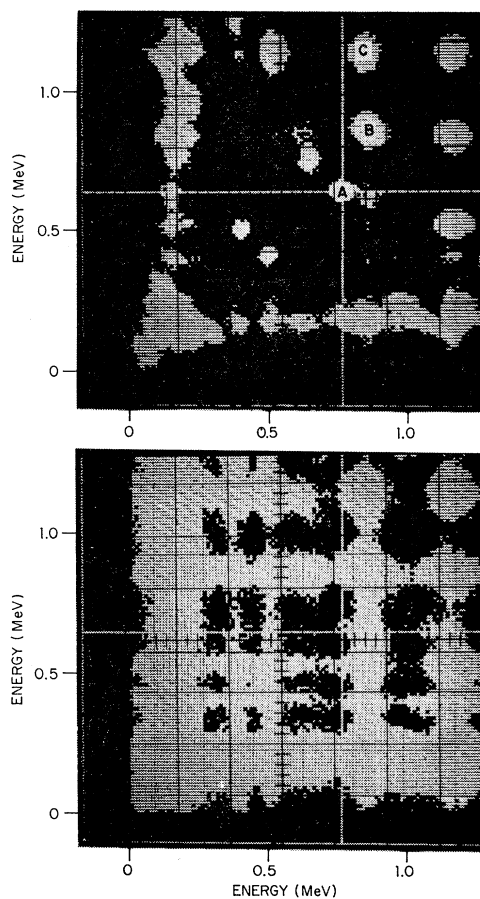


Fig. 2. Coincidence pulse-height spectra: Upper—approximately equal activities of  $^{132}\text{I}$ ,  $^{134}\text{I}$ , and  $^{135}\text{I}$ ; Lower—spectrum of iodine-fission products separated at early times after fission. Coincidence photopeaks: A— $^{132}\text{I}$ , 0.67 and 0.77 MeV, B— $^{134}\text{I}$ , 0.85 and 0.89 MeV, C— $^{135}\text{I}$ , 1.2 and 0.85 MeV.

<sup>8</sup> B. A. Euler, D. F. Covell, and S. Yamamoto, Naval Radiological Defense Laboratory Report No. USNRDL-TR-68-150, 1968 (unpublished).

TABLE I. Independent fission yields.

Run No.	Iodine separation time (sec) <sup>a</sup>	$^{131}\text{I}$ ( $10^{-5}$ atoms/fission)	$^{132}\text{I}$ ( $10^{-4}$ atoms/fission)	$^{133}\text{I}$ ( $10^{-3}$ atoms/fission)	$^{134}\text{I}$ ( $10^{-3}$ atoms/fission)
1	4.8	4.08	1.65	1.80	...
2	4.8	3.98	...	1.77	...
3	5.8	...	1.76	1.72	...
4	6.0	4.63	...	...	9.33
5	10.0	...	...	...	8.58
8	13.3	...	1.75	1.75	...
9	15.8	...	...	...	8.84
10	20.6	...	...	...	9.33
11	29.8	...	...	...	8.68
12	64.4	...	1.67	...	...
Weighted averages:		$4.2 \pm 0.4$	$1.74 \pm 0.11$	$1.76 \pm 0.09$	$9.0 \pm 0.8$

<sup>a</sup> From end of 10-sec irradiation.

accompanied by events in either of the annular crystals. The outputs from the detector system were processed on a two-parameter 16 384 channel analyzer and stored on magnetic tape.

Figure 2 shows two coincidence pulse-height spectra obtained with this counting system. The upper spectrum is of a source containing about equal activities of  $^{132}\text{I}$ ,  $^{134}\text{I}$ , and  $^{135}\text{I}$ ; the intensified regions show the prominent coincidence photopeaks from these nuclides. The photopeaks that were used to assay the three iodine nuclides are identified in the figure. The crosshairs are centered on the  $^{132}\text{I}$  photopeak.

The difficulty of the  $^{132}\text{I}$  measurement is clearly illustrated in the lower spectrum of Fig. 2. This spectrum is of iodine fission products separated at an early time (6 sec) following fission and counted 3.5 h later. In this source, the  $^{132}\text{I}$  activity is less than 1% of the activity of the  $^{134}\text{I}$  and  $^{135}\text{I}$  combined. The crosshairs are still on the  $^{132}\text{I}$  photopeak in this spectrum, but the peak is barely perceptible.

At the earliest counting times in these experiments (3–4 h from the end of irradiation) the region of the  $^{132}\text{I}$  coincidence photopeak contained about equal numbers of counts due to  $^{132}\text{I}$ ,  $^{134}\text{I}$ , and  $^{135}\text{I}$ . It was possible to determine the  $^{132}\text{I}$  contribution to this region from an analysis of decay data obtained in this region for each source. The sources were counted over a period of 10–15 h, and the counts in the photopeak region were resolved into 2.29-h  $^{132}\text{I}$ , 52.8-min  $^{134}\text{I}$ , and 6.75-h  $^{135}\text{I}$  by a computer program based on a method of multiple linear regression. Similarly, the other coincidence photopeaks shown in Fig. 2 were used to measure the  $^{134}\text{I}$  and  $^{135}\text{I}$  present in each source.

For measurement of the  $^{131}\text{I}$  and  $^{133}\text{I}$  in the sources, single-parameter pulse-height spectra were taken on a 3-by-3-in. NaI(Tl) well detector, and again repeated spectra were taken for each source. The counts in the region of the 0.36- and 0.54-MeV photopeaks of  $^{131}\text{I}$  and  $^{133}\text{I}$ , respectively, were used to assay these nuclides. The  $^{133}\text{I}$  photopeak was followed over a period extending from 2–10 days after each run and the  $^{131}\text{I}$  photopeak over a period extending from 1–4 weeks after each run. In each case, the counting was begun

this late because of interferences at early times from shorter-lived iodine nuclides.

Counting efficiencies for both the coincidence and singles measurements were obtained by calibrations against  $4\pi\beta$  counting. The  $4\pi\beta$  sources were prepared by reaction of iodine vapors in microgram quantities with silver which had been vacuum-deposited onto VYNS films.<sup>9</sup> Sources suitable for calibration of all the iodine isotopes of interest except  $^{135}\text{I}$  were prepared by isolation of tellurium fission products, followed by separations of their iodine daughters. By selection of the times of the tellurium separation and the subsequent iodine separation it is possible to enhance the amount of the isotope of interest and reduce the interference of other isotopes. In each of the calibration runs,  $^{125}\text{I}$  was added to the supply of iodine activities before the sources were prepared. This tracer was used to interrelate the different strengths of the  $4\pi\beta$  source and the coincidence and singles  $\gamma$  sources.  $^{125}\text{I}$  is a particularly useful tracer since its disintegration rate in a wide variety of sources can be easily determined by a simple  $\gamma$ -ray measurement.<sup>10</sup>

The counting efficiency was not determined for the  $^{135}\text{I}$  coincidence measurements because of the difficulty of doing  $4\pi\beta$  counting with this nuclide. Such counting is difficult because an unknown amount of the 15.6-min and 9.2-h  $^{135}\text{Xe}$  escapes in the gas used to flush the counter during the measurement. Because no counting efficiency was determined in these experiments it was possible only to establish the relative number of  $^{135}\text{I}$  atoms formed per fission as a function of the iodine separation time. As will be shown later, this information can be used to determine the half-life and fractional cumulative yield of  $^{135}\text{Te}$  and the fractional independent yield of  $^{135}\text{I}$ .

### III. RESULTS AND DISCUSSION

Table I shows the independent yields of  $^{131}\text{I}$ – $^{134}\text{I}$  calculated for each experimental run. In the determina-

<sup>9</sup> A. A. Delucchi and A. E. Greendale, *Radiochim. Acta* **6**, 47 (1966); Naval Radiological Defense Laboratory Report No. USNRDL-TR-948, 1965 (unpublished).

<sup>10</sup> J. S. Eldridge and P. Crowther, *Nucleonics* **22**, 56 (1964).

TABLE II. Fission yields in mass-131, -132 and -133 chains.

Fission product	Independent yield (%) <sup>a</sup>	Fractional yield <sup>b</sup>	Fractional cumulative yield <sup>c</sup>
<sup>131</sup> Sn	1.28±0.21 <sup>d</sup>	0.437±0.072	0.437±0.072
<sup>131</sup> Sb	1.66±0.40	0.567±0.137	0.875±0.014
<sup>131m+g</sup> Te		0.124±0.014 <sup>e</sup>	0.99856±0.00012
<sup>131</sup> I	(4.2±0.4)×10 <sup>-3</sup>	(0.144±0.012)×10 <sup>-2</sup>	
<sup>132</sup> Sn	0.59±0.17 <sup>d</sup>	0.135±0.039	0.135±0.039
<sup>132</sup> Sb	2.76±0.35	0.630±0.080	0.800±0.023
<sup>132</sup> Te	0.86±0.10	0.196±0.023	0.99603±0.00025
<sup>132</sup> I	(1.74±0.11)×10 <sup>-2</sup>	(0.397±0.025)×10 <sup>-2</sup>	
<sup>133</sup> Sb	3.05±0.39 <sup>d</sup>	0.461±0.059	0.404±0.045 <sup>f</sup>
<sup>133m+g</sup> Te	4.26±0.45	0.644±0.068	0.9734±0.0014
<sup>133</sup> I	0.176±0.009	(0.266±0.014)×10 <sup>-1</sup>	

<sup>a</sup> Except for the iodine yields, all yields are from Ref. 1.

<sup>b</sup> The fractional yields are based on the total chain yields as given by S. Katcoff, *Nucleonics* **18**, 201 (1960). Values are fractional independent yields except for those cases in which only the cumulative yield is given; then the value is the fractional cumulative yield.

<sup>c</sup> For these chains it is assumed that the independent yields of chain members beyond iodine can be neglected in computing the fractional cumulative yields.

<sup>d</sup> Cumulative yield.

<sup>e</sup> From D. G. Sarantites, G. E. Gordon, and C. D. Coryell, *Phys. Rev.* **138**, B353 (1965).

<sup>f</sup> Since the estimated fractional cumulative yield of <sup>133</sup>Sb derived from the tellurium and iodine fractional independent yields (1-0.644-0.0266=0.329±0.068) is of comparable precision to the measured antimony fractional cumulative yield (0.461±0.059), the estimate shown in the table is based on a weighted average of these two values. Each value was weighted by the reciprocal of its estimated variance.

tion of these yields, factors such as (1) counting efficiency, (2) number of fissions, (3) iodine chemical yield, and (4) iodine growth from  $\beta$  decay of precursors both during irradiation and before iodine separation were considered. The corrections for growth of <sup>131</sup>I, <sup>132</sup>I, and <sup>133</sup>I were based on the branching ratios and half-lives given by Meek and Rider<sup>11</sup> and the fission yields given by Strom *et al.*<sup>1</sup> For <sup>134</sup>I, calculations were based on an <sup>134</sup>Sb half-life and cumulative yield of 11.1 sec and 0.32%,<sup>12</sup> respectively, and a <sup>134</sup>Te half-life and cumulative yield of 42 min<sup>13</sup> and 6.48%,<sup>14</sup> respectively. In all runs used for calculation of independent fission yields the contribution of precursors to the observed yield was small. For obtaining the values shown in Table I, a correction was also made for iodine fission fragments which are trapped in the carbon powder used in the reaction mixture and which do not exchange with the carrier iodine. It was found that 16% of the iodine fission fragments were lost by this mechanism. This correction is discussed in the Appendix. The weighted averages given in Table I were obtained by weighting of each individual value by the reciprocal of its estimated variance.

To the authors' knowledge, the <sup>131</sup>I and <sup>132</sup>I independent fission yields have not been reported previously. The values obtained in this work were compared with the other yield data available in the mass-131 and -132 chains under the assumption that

all of the data in these chains can be represented by Gaussian distributions.<sup>1,15</sup> In the mass-132 chain, the Gaussian curve that best fits the present data was found to be essentially unchanged from the curve that was previously reported.<sup>1</sup> Also the yield data were found to be in excellent agreement with a Gaussian distribution. In the mass-131 chain, the Gaussian curve that best fits the data was found to be somewhat narrower than the curve that was previously reported.<sup>1</sup> A Gaussian curve does not fit the yield data well in this chain; however the curve is reasonable, considering the precision of the measurements.

The <sup>133</sup>I independent yield measured in these experiments is in good agreement with the value previously reported from this laboratory<sup>1</sup> of (1.7±0.6)×10<sup>-3</sup>. However, the latest measurement is of considerably improved precision, primarily because earlier iodine-separation times were achieved.

The <sup>134</sup>I independent yield reported here is in good agreement with the previously reported value<sup>16</sup> of 0.94±0.13%. Based on a mass-134 chain yield<sup>11</sup> of 7.92%, the <sup>134</sup>I independent yield measured in these experiments corresponds to a fractional independent yield of 0.11±0.01. This fractional independent yield is in good agreement with the value reported by Wahl<sup>17</sup> of 0.11±0.02.

Table II shows the fission-yield data presently available in the mass-131, -132, and -133 chains. The fractional cumulative yields from this table were

<sup>11</sup> M. E. Meek and B. F. Rider, General Electric Co. Report No. APED-5352, 1967 (unpublished), revision 1.

<sup>12</sup> A. A. Delucchi and A. E. Greendale, *Phys. Rev.* **173**, 1159 (1968); Naval Radiological Defense Laboratory Report No. USNRDL-TR-67-140, 1967 (unpublished).

<sup>13</sup> J. M. Ferguson, D. L. Love, and D. Sam, *J. Inorg. Nucl. Chem.* **24**, 1 (1962); Naval Radiological Defense Laboratory Report No. USNRDL-TR-458, 1960 (unpublished).

<sup>14</sup> L. Yaffe, A. E. Day, and B. A. Greer, *Can. J. Chem.* **31**, 48 (1953).

<sup>15</sup> A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, *Phys. Rev.* **126**, 1112 (1962).

<sup>16</sup> A. C. Pappas, Laboratory for Nuclear Science, Massachusetts Institute of Technology, Technical Report No. 63, 1953 (unpublished); recalculated values by A. C. Pappas of <sup>134</sup>I yield determined by L. E. Glendenin, Laboratory of Nuclear Science, Massachusetts Institute of Technology, Technical Report No. 35, 1949 (unpublished).

<sup>17</sup> A. C. Wahl, *Phys. Rev.* **99**, 730 (1955).

plotted on a probability-scale plot with the atomic number  $Z$  as the abscissa. On such a plot,<sup>15</sup> a Gaussian distribution is given by a straight line and the mean of the distribution,  $Z_p$  ("most probable charge"), and standard deviation of the distribution,  $\sigma$ , can be read directly from the plot. Values of  $Z_p$  and  $\sigma$  were determined for each chain by fitting of a Gaussian curve to the yield data on such a plot.

Once the  $Z_p$  values are known, the data for all three chains can be conveniently shown on one probability-scale plot, as in Fig. 3, with the quantity  $Z - Z_p$  as the abscissa. The straight lines in this figure represent the Gaussian curves that best fit the yield data in each chain, and the values of  $Z_p$  and  $\sigma$  for each curve are shown in the figure. The trend of decreasing distribution widths with increasing mass number previously reported<sup>1</sup> for these three chains is supported by the yield data shown in Fig. 3. However, it should be noted that the strongest evidence for this trend is the narrow width of the yield distribution in the mass-133 chain, and that the yield distribution in this chain is defined by only two yield measurements. Measurement of at least one more independent yield in the mass-133 chain would add greatly to the knowledge of the shape of the yield distribution in this chain.

The counting efficiency was not determined for the  $^{135}\text{I}$  measurements; therefore, the absolute fission yield

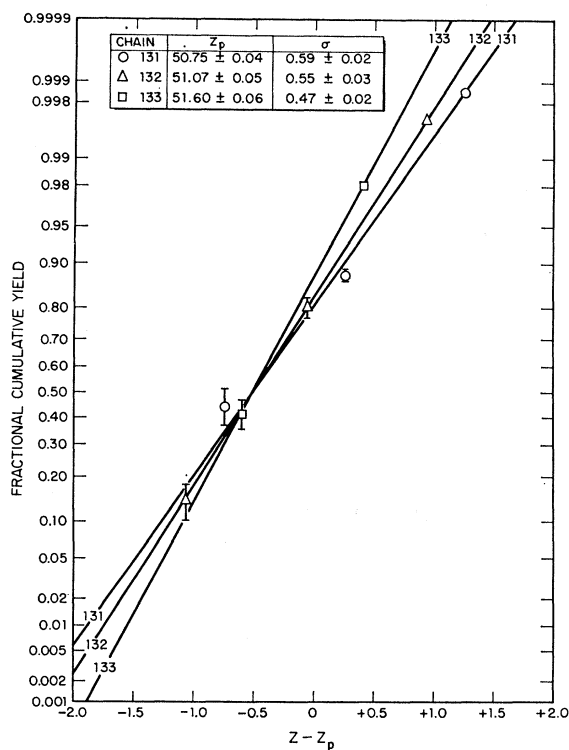


Fig. 3. Probability-scale plot of fractional-cumulative-yield data for mass-131, -132, and -133 chains.

TABLE III.  $^{135}\text{I}$  counting data.

Run No.	Iodine separation time (sec) <sup>a</sup>	Counts/min $^{135}\text{I}$ $10^{11}$ fissions
4	6.0	8797
5	10.0	8538
6	11	8845
7	11	9379
9	15.8	10 249
10	20.6	10 429
11	29.8	10 871
13	160	11 592
14	783	11 232
15	2413	10 633

<sup>a</sup> From end of 10-sec irradiation.

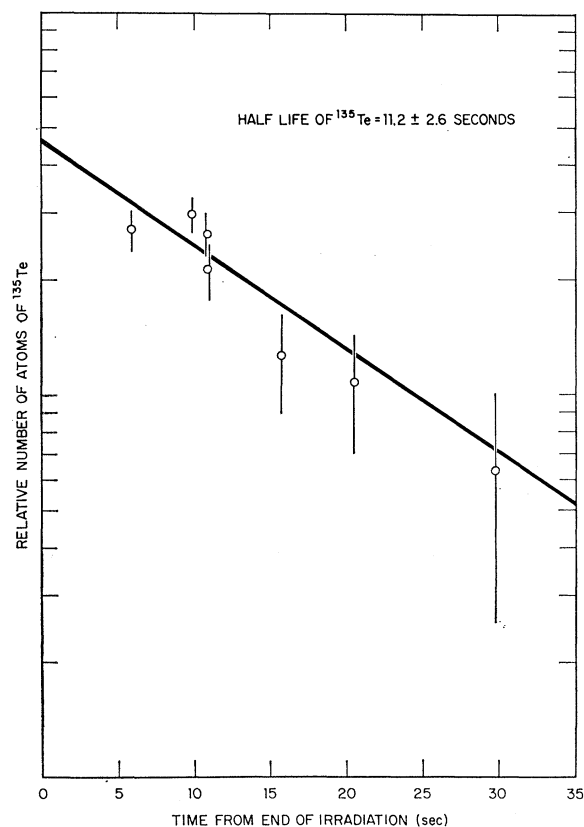
of  $^{135}\text{I}$  could not be obtained. However, from the counting data that were obtained it was possible to determine the fractional cumulative yield of  $^{135}\text{Te}$  and the fractional independent yield of  $^{135}\text{I}$ , as well as the half-life of  $^{135}\text{Te}$ . This was done as follows: The relative activity of  $^{135}\text{I}$  separated at times ranging from 6 sec to 40 min from the end of a 10-sec irradiation was measured. The counting data were normalized, to correspond to the  $^{135}\text{I}$  activity that would result at the times of the iodine separations from  $10^{11}$  fissions, by correction of the data for the iodine chemical yield and the number of fissions obtained in each run. The normalized data are shown in Table III. These data were analyzed under the assumption that they represent the growth and decay of 6.75-h  $^{135}\text{I}$  from a single tellurium precursor. Because of the short half-life<sup>18</sup> and the expected low cumulative fission yield of  $^{135}\text{Sb}$ , its presence was neglected in analysis of the data. The data were assumed to be described by the standard equations for a parent-daughter mixture. The count rate observed in the  $^{135}\text{I}$  coincidence photopeak can be expressed as  $\epsilon N_2 \lambda_2$ , where  $\epsilon$  is the counting efficiency of  $^{135}\text{I}$ ,  $N_2$  is the number of  $^{135}\text{I}$  atoms present in the sample, and  $\lambda_2$  is the decay constant for  $^{135}\text{I}$ . This count rate at any iodine separation time  $t$  from the end of a fixed irradiation interval is given by the expression

$$\epsilon N_2 \lambda_2 = \epsilon N_2^0 \lambda_2 \exp(-\lambda_2 t) + [\epsilon N_1^0 \lambda_1 \lambda_2 / (\lambda_1 - \lambda_2)] [\exp(-\lambda_2 t) - \exp(-\lambda_1 t)], \quad (1)$$

where  $N_1^0$  is the number of  $^{135}\text{Te}$  atoms present at the end of the irradiation,  $N_2^0$  is the number of  $^{135}\text{I}$  atoms present at the end of the irradiation, and  $\lambda_1$  is the  $^{135}\text{Te}$  decay constant. Rearranging Eq. (1) we obtain

$$\epsilon N_2 \lambda_2 = \left( \epsilon N_2^0 \lambda_2 + \frac{\epsilon N_1^0 \lambda_1 \lambda_2}{\lambda_1 - \lambda_2} \right) \times \exp(-\lambda_2 t) - \frac{\epsilon N_1^0 \lambda_1 \lambda_2}{\lambda_1 - \lambda_2} \exp(-\lambda_1 t). \quad (2)$$

<sup>18</sup> C. E. Bemis, G. E. Gordon, and C. D. Coryell, J. Inorg. Nucl. Chem. **26**, 213 (1964).

FIG. 4. Decay of  $^{135}\text{Te}$ .

It was assumed that for runs 13–15

$$\frac{\epsilon N_1^0 \lambda_1 \lambda_2}{\lambda_1 - \lambda_2} \exp(-\lambda_1 t) \ll \left( \epsilon N_2^0 \lambda_2 + \frac{\epsilon N_1^0 \lambda_1 \lambda_2}{\lambda_1 - \lambda_2} \right) \exp(-\lambda_2 t), \quad (3)$$

and that, therefore, the count rate of  $^{135}\text{I}$  at these times could be given by

$$\epsilon N_2 \lambda_2 = \langle \epsilon N_2^0 \lambda_2 + [\epsilon N_1^0 \lambda_1 \lambda_2 / (\lambda_1 - \lambda_2)] \rangle \exp(-\lambda_2 t). \quad (4)$$

The data from runs 13–15 were used to determine the coefficient of the exponential term in Eq. (4). The right-hand side of Eq. (4) was then computed for each of the earlier iodine separation times of runs 4–7 and runs 9–11. The count rates observed at these earlier times were subtracted from the values computed by Eq. (4). The resulting data are shown in Fig. 4 and correspond to the first term in Eq. (2), i.e.,

$$[\epsilon N_1^0 \lambda_1 \lambda_2 / (\lambda_1 - \lambda_2)] \exp(-\lambda_1 t).$$

The straight line shown through the data was obtained by a nonlinear least-squares procedure with each datum weighted by the reciprocal of its estimated variance. In this manner, the half-life of  $^{135}\text{Te}$  was

found to be  $11.2 \pm 2.6$  sec. This half-life is somewhat lower than the value of  $18 \pm 2$  sec recently obtained by Denschlag.<sup>19</sup>

At this point, the values of both coefficients of the exponential terms in Eq. (2) were known, as well as the value of  $\lambda_1$ , and it was possible to determine the ratio  $N_1^0/N_2^0$ . After a correction was made to this ratio for saturation effects during the 10-sec irradiations, the atom ratio of  $^{135}\text{Te}$  (cumulative) to  $^{135}\text{I}$  (independent) formed in thermal-neutron-induced fission of  $^{235}\text{U}$  was found to be 1.19. Based on this information and the reported fractional independent yield of  $^{135}\text{Xe}$  of  $0.037 \pm 0.012$ ,<sup>15</sup> the fractional cumulative yield of  $^{135}\text{Te}$  and the fractional independent yield of  $^{135}\text{I}$  were computed to be  $0.52 \pm 0.07$  and  $0.44 \pm 0.07$ , respectively. These values agree within the precision of the measurements with the values reported by Wunderlich<sup>20</sup> of  $0.41 \pm 0.12$  and  $0.56 \pm 0.09$  for the  $^{135}\text{Te}$  fractional cumulative yield and the  $^{135}\text{I}$  fractional independent yield, respectively. Our  $^{135}\text{I}$  yield is lower than the value of  $0.72 \pm 0.17$  reported by Fröhner,<sup>21</sup> but is in excellent agreement with the value of  $0.47 \pm 0.02$  recently obtained by Denschlag.<sup>19</sup> If the isobaric-yield distribution in this chain is assumed to be Gaussian, the yields we measured for  $^{135}\text{Te}$  and  $^{135}\text{I}$  lead to computed  $Z_p$  and  $\sigma$  values for the mass-135 chain of  $52.46 \pm 0.10$  and  $0.59 \pm 0.06$ , respectively.

#### APPENDIX: EXCHANGE OF IODINE FISSION PRODUCTS WITH CARRIER IODINE

When measurements of the independent yield of  $^{134}\text{I}$  were first made with the radiochemical procedure described in this paper, it was noted that the values obtained averaged 15–20% below the published<sup>16,17</sup> values. To determine whether our disagreement with the published value was real and was possibly due to the radiochemical procedure we had employed, we remeasured the  $^{134}\text{I}$  yield in two independent ways.

In three experiments the iodine fission products were separated by a modification of a radiochemical procedure given by Glendenin and Metcalf<sup>22</sup> and in three other experiments by our faster procedure. The iodine activities from all runs were counted on a 3×3-in. NaI(Tl) well counter with the baseline on the counter set to record only  $\gamma$  rays above 750 keV. The counting data were normalized for the iodine chemical yield and number of fissions in each run, and corrections were made for growth from  $^{134}\text{Te}$ . It was found that the procedure based on the thermal de-

<sup>19</sup> H. O. Denschlag (private communication).

<sup>20</sup> F. Wunderlich, *Radiochim. Acta* **7**, 105 (1967).

<sup>21</sup> F. H. Fröhner, *Z. Physik* **170**, 62 (1962).

<sup>22</sup> L. E. Glendenin and R. P. Metcalf, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman, (McGraw-Hill Book Co., Inc., New York, 1951) National Nuclear Energy Series, Div. IV, Vol. 9, p. 1625.

composition of the uranium-peroxide-periodic-acid-carbon (U-I-C) mixture gave a relative independent yield for  $^{134}\text{I}$  that was 16.4% lower than that obtained with the procedure of Glendenin and Metcalf.

A possible explanation for this lower yield was discovered during analyses for  $^{99}\text{Mo}$  in capsules containing the U-I-C mixture. After irradiation, samples that were to be used for  $^{99}\text{Mo}$  analysis were treated with an aqueous solution containing molybdenum carrier, and the insoluble carbon fraction was separated by filtration. It was found that 18.1% of the  $^{99}\text{Mo}$  was associated with the carbon fraction. Since the percentage loss of  $^{134}\text{I}$  in the fast procedure was very nearly the same as the percentage of  $^{99}\text{Mo}$  found on the carbon, it was suspected that the lost  $^{134}\text{I}$  was somehow trapped on the carbon. One possible explanation was that both effects were due to the stopping of some of the fission fragments in the carbon particles. A calculation of the range of the molybdenum and iodine-fission fragments in periodic acid indicated that a large fraction of these would leave the original uranium-peroxide-periodic-acid granules in which fission had occurred. Some of these could be expected to be stopped within the carbon particles.

To determine the fraction of iodine fission fragments trapped in the carbon, the following experiments were conducted: Two capsules were irradiated. The first capsule contained the same U-I-C mixture used in the fast iodine procedure, while the second capsule contained uranium peroxide-periodic acid powder and no carbon. After irradiation the ingredients of the first capsule were dissolved in water and the carbon fraction was separated on Whatman 42 filter paper. A measure of the amount of 6.75-h  $^{135}\text{I}$  in the carbon was obtained by counting of the filter paper and carbon on a  $\gamma$ - $\gamma$  coincidence spectrometer. Coincidences between two  $\gamma$  rays, each having energies between 1.0 and 1.25 MeV, were recorded. The sample was counted over a period of 3 days, and the  $^{135}\text{I}$  contribution was determined by resolution of the decay data.<sup>23</sup> The  $^{135}\text{I}$  in the filtrate was purified before counting by the

radiochemical procedure of Glendenin and Metcalf,<sup>22</sup> and a  $\gamma$  source with the same counting geometry as the source containing carbon was prepared from this iodine by precipitation of silver iodide and collection on Whatman 42 filter paper. The amount of  $^{135}\text{I}$  in this source also was determined by coincidence spectrometry. In this manner it was found that 17.7% of the total  $^{135}\text{I}$  in the sample was associated with the carbon fraction.

The second capsule was used to determine the amount of  $^{135}\text{I}$  adsorbed onto the carbon during the dissolution and filtration steps. Carbon was added to the second capsule after the irradiation and intimately mixed with the uranium-peroxide-periodic-acid powder. The mixture was then processed in exactly the same way as the mixture from the first capsule. In this way it was found that 2.0% of the  $^{135}\text{I}$  was adsorbed onto the carbon during the dissolution and filtration steps. Correcting for this amount of adsorption, we concluded that 15.7% (17.7–2.0%) of the  $^{135}\text{I}$  fission fragments were stopped in the carbon during the irradiation of the U-I-C mixture.

The above conclusion is not altered by the fact that over one-half of the fission product  $^{135}\text{I}$  at the time of our measurements resulted from  $\beta$  decay of  $^{135}\text{Te}$ . One would expect the  $^{135}\text{Te}$  and  $^{135}\text{I}$  fission fragments to have very nearly the same ranges and therefore to be trapped essentially to the same extent in the carbon. The same fraction of the  $^{135}\text{I}$  will be found, therefore, in the carbon after all of the  $^{135}\text{Te}$  has decayed (several minutes) as would have been found if all of the  $^{135}\text{I}$  had been formed independently in fission.

On the basis of the above results, a correction of 16% was applied to the fission yields obtained for  $^{131}\text{I}$ ,  $^{132}\text{I}$ ,  $^{133}\text{I}$ , and  $^{134}\text{I}$  by the fast iodine radiochemical procedure. The  $^{135}\text{Te}$  and  $^{135}\text{I}$  fractional yields measured by this procedure did not require a correction for losses in the carbon. Those measurements were derived from a study of the relative amounts of  $^{135}\text{Te}$  and  $^{135}\text{I}$  found at various times after fission and therefore would be unaffected by trapping in the carbon, provided the fractional loss of each nuclide in the carbon is the same.

<sup>23</sup> The only other activity detected in the coincidence window was a small tail due to  $^{132}\text{I}$  in equilibrium with 78 h  $^{132}\text{Te}$ .

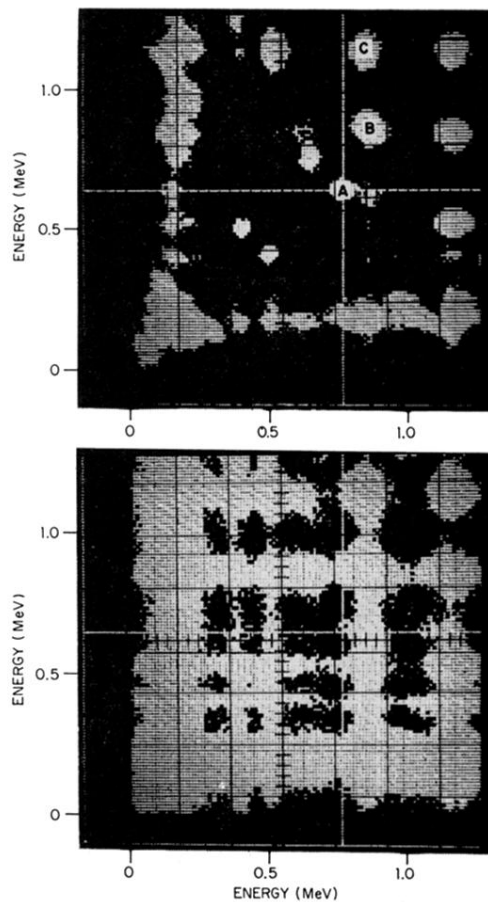


FIG. 2. Coincidence pulse-height spectra: Upper—approximately equal activities of  $^{132}\text{I}$ ,  $^{134}\text{I}$ , and  $^{135}\text{I}$ ; Lower—spectrum of iodine-fission products separated at early times after fission. Coincidence photopeaks: *A*— $^{132}\text{I}$ , 0.67 and 0.77 MeV, *B*— $^{134}\text{I}$ , 0.85 and 0.89 MeV, *C*— $^{135}\text{I}$ , 1.2 and 0.85 MeV.