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Nuclear-Charge Distribution of Fission-Product Chains of Mass Numbers 131–133*

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New information on the nature of the nuclear charge distribution in thermal-neutron fission of ^{235}U has been obtained from measurements of fission yields of the principal constituents in each of three adjacent mass chains. A regular variation in the width of the charge-distribution curves with mass number has been shown, and values of the most probable nuclear charge (Z_p) have been determined. No effects attributable to the 50-proton or 82-neutron closed shells have been observed in the charge-distribution curves examined. Independent and cumulative fission yield values were determined for the principal constituents in each of the three mass chains 131, 132, and 133. Values measured are: ^{131}Sn, $(1.28\pm0.21)\%$ cumulative; ^{131}Sb, $(1.66\pm0.40)\%$ independent; ^{132}Sn, $(0.59\pm0.17)\%$ cumulative; ^{132}Sb, $(2.76\pm0.35)\%$ independent; ^{132}Te, $(0.86\pm0.10)\%$ independent; ^{133}Sn, $<0.015\%$ cumulative; ^{133}Sb, $(3.05\pm0.39)\%$ independent; ^{133m}Te, $(2.19\pm0.33)\%$ independent; ^{133g}Te, $(2.07\pm0.31)\%$ independent; and ^{133}I, $(0.17\pm0.06)\%$ independent. The charge distribution curve for each mass chain was established from the fission yield data. The data were fitted to the Gaussian relationship, $P(Z)=A\exp-(Z\text{-}Z_p)^2/c$. The widths of the curves as given by values of c for mass chains 131, 132, and 133 are, respectively: 1.10 ± 0.05, 0.74 ± 0.08, and 0.57 ± 0.05. The data show a monotonic decrease in the width of the charge distribution curves with increasing fission-product mass number. Implications of this observation are discussed in respect to previous assumptions of a single charge distribution curve for all fission-product masses in low-energy fission. Values of Z_p as given by the Gaussian curves for mass chains 131, 132, and 133 are, respectively: 50.72 ± 0.07, 51.07 ± 0.06, and 51.59 ± 0.04. These values are compared with published values derived from assumptions of a single charge distribution curve for all mass chains. Predicted effects of nuclear shells on charge distribution have been examined in the light of the present data. No effect was detected in mass chains 131, 132, and 133. New half-life values determined are: ^{131}Sn, 1.32 ± 0.23 min; ^{132}Sn, 1.00 ± 0.17 min; ^{132}Sb, 3.13 ± 0.33 min; and ^{133}Sb, 2.67 ± 0.33 min. The half-life of ^{133}Sn was estimated to be 55 sec.

I. INTRODUCTION

THE nature of nuclear charge division in fission is as yet only poorly characterized. Thus there is presently insufficient data (1) to clearly define the most probable nuclear charge division for all mass divisions; (2) to establish if, and to what extent, the dispersion of charge varies with mass number; and (3) to indicate the influence of nuclear shells on both of the preceding processes. Some information pertinent to these questions has been obtained in the studies reported here. In particular, we report on the determination of nuclear charge distributions for three mass chains (131, 132, 133) in the region of the 50-proton and 82-neutron nuclear shells for the case of thermal neutron-induced fission of ^{235}U. Studies of other mass chains will be reported in subsequent papers.

The results of charge distribution studies have been summarized by Hyde.[1] The residual fission products, resulting from prompt neutron emission from primary fission fragments, are produced in measurable quantities only in a narrow range of nuclear charge for any particular mass chain. A large fraction of those residual nuclides produced by low-energy fission have half-lives which are short relative to the time required for their radiochemical separation. Because of this difficulty, fission yields for more than one member of a mass chain have not often been determined. As a result, not much information is available on the dispersion of charge within individual mass chains.

Enough information has been accumulated, however, to characterize general features of the distribution of charge in fission. Thus, it is known that the most probable charge occurs on the average about 3 charge units from beta stability, and that the dispersion of yield about this most probable charge is quite narrow. It is also known that the distances from beta stability for the light and heavy fragments are, on the average, about the same. However, because of the paucity of yield data within individual mass chains, it has not been possible, except in only a very few chains,[2] to establish both the location of Z_p and the shape of the charge-distribution curve from the yield measurements. For this reason, the radiochemical yield data have at times[3] been used to define the shape of a charge distribution curve by assuming knowledge of the location of Z_p, and, more recently,[2] have been used to define the location of Z_p by assuming knowledge of the shape of the charge distribution curve. Both of these attacks on the problem of charge division have required that one make the assumption that the same charge distribution curve is applicable to all mass chains. This assumption will be examined later in this paper in light of the most recent data on charge dispersion.

The search for effects of closed nuclear shells on the

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* This work was supported in part by the U. S. Atomic Energy Commission.

[1] E. K. Hyde *et al.*, *The Nuclear Properties of the Heavy Elements.* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964) Vol. 3, pp. 141–156.

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

[3] 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), p. 489.

distribution of charge in fission is made difficult because of the lack of clear trends in the experimental data. There are presently available several theories and empirical rules which yield conflicting predictions for the value of Z_p as a function of mass. It can safely be said that no one of these has been shown to hold throughout the fission product region. Also, none of the theoretical approaches has given quantitative information on the shape of the charge distribution curves. As a consequence, the appearance of a shell effect within a mass chain will probably be detected only if the effect is a relatively large one. Several predictions have been made[4–6] of the effects of closed nuclear shells in the mass region studied in this work, and these will be discussed in light of the yield data that were obtained for these chains.

II. EXPERIMENTAL

Information on charge distribution is obtained most accurately by radiochemical means. In the following are described the procedures developed and utilized for very rapidly isolating purified short-lived fission-product radionuclides.

Irradiations were carried out in the 10-kW reactor of the Nuclear Technology Laboratory of Stanford University. Pneumatic sample-transfer (rabbit) systems, described in other reports,[7,8] were used for the transport of targets to and from the site of irradiation.

During the runs, the reactor remained at an operating power level of 10 kW. The neutron flux in the area of bombardment was approximately 10^{10} n/cm^2-sec and the cadmium ratio for gold was determined to be 12 ± 2 under the conditions of the experiments. A gold foil was present on each rabbit as a flux monitor in each experimental run. The ^{198}Au activity resulting from these bombardments was related to the number of fissions occurring in the target samples by a series of radiochemical determinations of ^{99}Mo, ^{139}Ba, and ^{140}Ba found in identical samples. The cumulative fission yields of these nuclides resulting from the thermal neutron fission of ^{235}U were taken to be 6.25, 6.42, and 6.25%,[9] respectively.

Following irradiation, rapid radiochemical separations were carried out to separate tin, antimony, tellurium, or iodine (depending upon the nuclide under investigation) from the fission products and, particularly, from other chain members. The radiochemical procedures by which tin was separated as stannane, antimony as stibine, and iodine as the element have been reported elsewhere.[10,11,8]

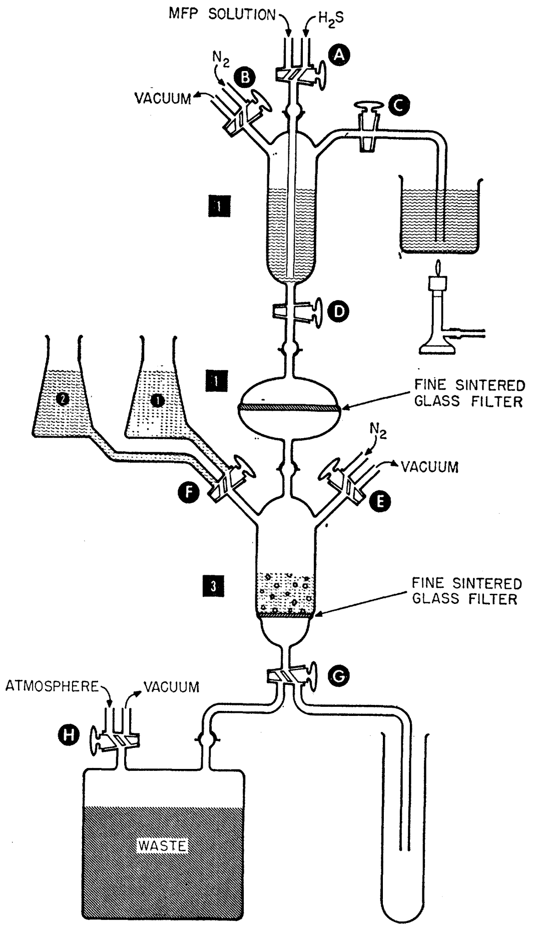

FIG. 1. Apparatus for separation of tin.

Some of the earlier separations of tin performed in this work were based on sulfide precipitations. These separations were carried out in an apparatus designed to allow rapid manipulations. The apparatus is shown in Fig. 1.

Tin separations were performed in the following way. To unit 1, in Fig. 1, a solution containing 1 ml of concentrated HCl, 10 mg of Sb(V), 10 mg of Sn(IV), ^{113}Sn tracer (for chemical yield determination), and approximately 0.5 mg of iodine as iodate was added. The solution was maintained at between 40 and 60°C. Following irradiation and immediately prior to the return of the rabbit, stopcock A was opened for the admittance of the irradiated solution, and stopcock B was turned to the vacuum position. The return of the rabbit from the reactor caused it to be impaled onto two hypodermic needles by means of which the solution and its rinse were transferred from the rabbit

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[4] P. Armbruster, in *Symposium on the Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965).

[5] H. Freisner and K. Wildermuth, Phys. Letters **2**, 212 (1962).

[6] D. M. Wiles, B. W. Smith, R. Horsley, and H. G. Thode, Can. J. Phys. **31**, 419 (1953).

[7] A. E. Greendale and D. L. Love, Nucl. Instr. Methods **23**, 209 (1963).

[8] A. E. Greendale, D. L. Love, and A. A. Delucchi, Anal. Chim. Acta **34**, 32 (1966).

[9] H. Farrar and R. H. Tomlinson, Nucl. Phys. **34**, 367 (1962).

[10] A. E. Greendale and D. L. Love, Anal. Chem. **35**, 1712 (1963).

[11] A. E. Greendale and D. L. Love, Anal. Chem. **35**, 632 (1963).

into unit 1. The contents of the rabbit included 120 mg of uranium as UO_2Cl_2, with the uranium having a ^{235}U enrichment of 93.2%.

Hot water was drawn into unit 1 through stopcock C, which was then closed. Stopcock A was then turned to permit entry of H_2S, which was allowed to saturate the solution. Stopcock A was then turned off and stopcock B was turned to allow N_2 gas to exert about 5 psig pressure over the solution and the sulfide precipitate. Stopcock D was then opened and the antimony and tellurium sulfides were filtered onto the fine sintered glass frit in unit 2.

The solution, in which fission product tin and carrier remained, proceeded into unit 3. Supported above the glass frit in unit 3 were 35 ml of an $(NH_4)_2S$ solution made up shortly before the experiment by saturating 0.43 *N* NH_4OH with H_2S at 0°C. In this environment, tin sulfide was formed. The reagents were well mixed by an air stream coming through the frit via stopcock G. Stopcocks G and H were then positioned such that the supernatant solution was drawn off to the waste container, leaving tin sulfide on the frit. Stopcock H was then opened to the atmosphere, E was turned to vacuum position, and F was turned such that 25 mliter of 0.36 *N* NaOH was allowed to drain into unit 3. The tin sulfide dissolved in the base and was again mixed well by passage of air up through the frit of unit 3. Stopcock F was then opened, allowing 3 mliter of concentrated HCl into unit 3, and tin sulfide was again precipitated. This precipitate was filtered in the same manner as was the first tin sulfide. Stopcock F was then opened to allow 10 ml of base into unit 3, the tin sulfide was redissolved, and the solution was drained into a test tube by the appropriate opening of stopcock G. These steps were sufficient to insure adequate decontamination from other chain members.[12]

The separation of tellurium was accomplished in a manner similar to that required for the tin sulfide procedure. The apparatus used for the rapid separation of tellurium is pictured in Fig. 2. A 20 ml solution, 8 *N* in HCl, containing 0.01 mg of antimonous and 0.01 mg antimonic carriers, and 5 drops of a 0.1% solution of a surface-active agent (Aerosol OT) was introduced into the reaction vessel of the tellurium apparatus. This solution was heated to boiling by heating elements which surrounded the vessel. The heating elements were made of nichrome wire cemented in place by a porcelain cement (Sauereisen). Hydrogen sulfide gas was bubbled through the solution prior to the insertion of the rabbit into the reactor. The solution in the rabbits to be irradiated contained the following components: 0.5 ml of 1 *N* HCl solution containing 120 mg of 93.2% enriched ^{235}U as UO_2Cl_2, 0.5 ml of 8 *N* HCl containing 20 mg of tellurous and 20 mg of telluric carriers, and 0.2 ml of a 6 *N* HCl solution containing a known amount of ^{125m}Te tracer (for chemical yield determinations).

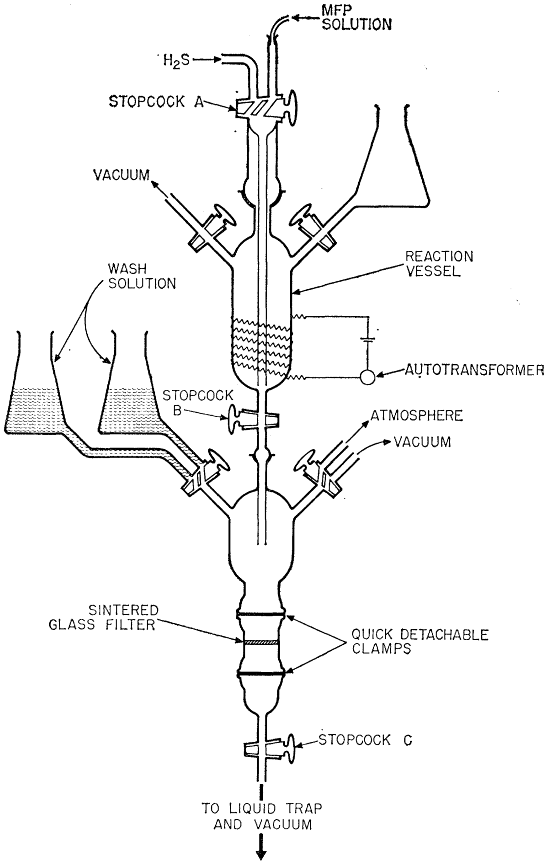

FIG. 2. Apparatus for separation of tellurium.

Following irradiation, the contents of the rabbit were removed with the hypodermic-needle arrangement (described earlier) into the reaction vessel. While the solution in the reaction vessel was maintained close to boiling, the addition of H_2S gas readily precipitated the tellurium. Stopcock A regulating the admission of H_2S was then closed, while stopcock B at the bottom of the reaction flask was opened. The precipitate that was formed was filtered onto the fritted glass, and the supernatant solution was discarded into a liquid trap by opening stopcock C. The precipitate was then washed twice with 10 ml portions of hot 8 *N* HCl

TABLE I. Decontamination factors in tellurium separation.

Tracer material	Decontamination factor
$^{124}Sb(III)$ (antimonous carrier, 0.01 mg/20 ml)	160
$^{124}Sb(V)$ (antimonic carrier, 0.01 mg/20 ml)	82
$^{113}Sn(II)$ (stannous carrier, 10 mg/sample)	6×10^2
$^{113}Sn(IV)$ (stannic carrier, 10 mg/sample)	1.5×10^3
$^{131}I^-$ (carrier-free)	91

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[12] W. E. Nervik, U. S. At. Energy Comm. NAS-NS-3023, 1960 (unpublished).

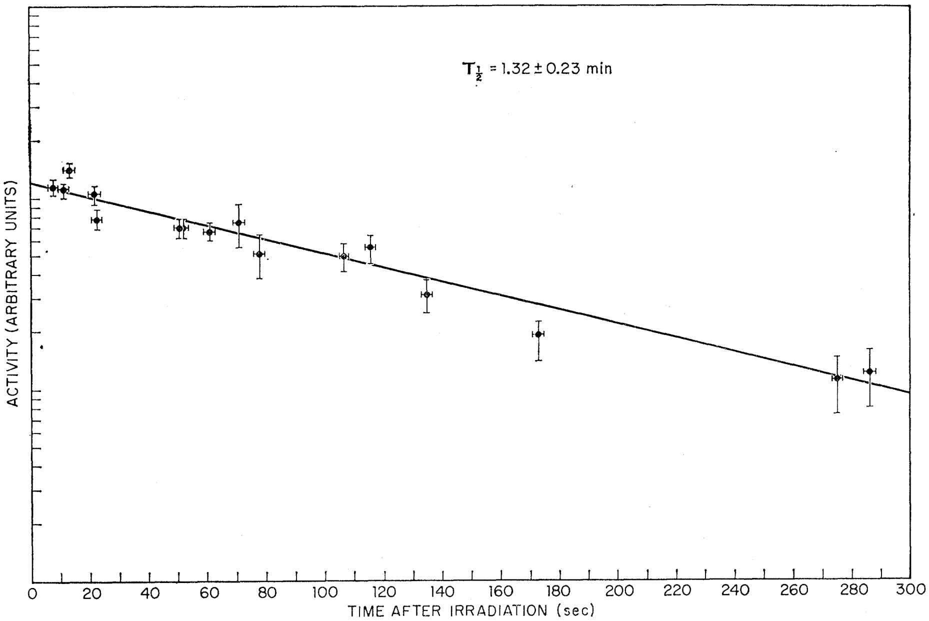

FIG. 3. Decay of ^{131}Sn.

solutions containing 0.01 mg of antimonous and 0.01 mg of antimonic carriers and 5 drops of 0.1% Aerosol OT solution. The tellurium precipitate, containing coprecipitated fission product selenium, was then reserved for separation of iodine daughters.

Table I gives decontamination factors of elements contained in the mass chains under investigation for the above tellurium procedure.

III. HALF-LIVES, MASS ASSIGNMENTS, AND YIELDS

^{131}Sn

Tin separations were carried out by both the sulfide precipitation method and the stannane evolution method on a number of irradiated, enriched ^{235}U solutions. These were performed at a number of intervals which were timed from the end of thermal-neutron irradiations of predetermined duration.

Iodine was removed from these tin fractions at convenient times for the measurement of ^{131}I (usually 1 day). The resulting iodine sources were generally not counted until sufficient time had elapsed such that the contribution of 2.3-h ^{132}I to the total activity was insignificant. Spectra were then taken of the radioiodine on one or more calibrated gamma-ray, multichannel, pulse-height analyzers. The number of atoms of ^{131}I was determined by measurements of its 364-keV photopeak. This information and a knowledge of the genetic relationships and half-lives of ^{131}I precursors allowed the number of atoms of ^{131}Sn to be calculated for each tin separation time. The resulting decay curve for ^{131}Sn is presented in Fig. 3, normalized to a constant number of fissions occurring in each irradiated solution.

It is evident from the plot that any precursors are quite short-lived (or more unlikely, long-lived) or are produced in low yield. Also, there is no evidence of an isomer of appreciable cumulative yield. The decay-curve data was therefore treated as being of cumulative ^{131}Sn. On this basis, a least-squares analysis was made of the data and the value is substantially shorter than that reported by Pohm *et al.*[13]

With the above information, the cumulative fission yield was determined for ^{131}Sn. The spread of the data was found to be substantially greater than the determinate error of any individual measurement; therefore the standard error of the mean is reported as the error. The values obtained are presented in Table II. The average value for the cumulative yield of ^{131}Sn is $(1.28\pm0.21)\%$.

^{132}Sn

In a number of instances, tin separations were carried out in the manner described for ^{131}Sn. In these cases, of course, the iodine was isolated and gamma pulse height analyzed sufficiently early to accurately measure the contribution from ^{132}I. The 673-keV gamma photopeak was used for determining the disintegration rates. Alternatively, tellurium separations were made on the tin fractions approximately one day following tin isolation. Spectra of ^{132}Te (including daughter ^{132}I) were then recorded on instruments of known efficiencies. In either case, the number of atoms of ^{132}Sn was obtained (normalized to a constant number of total fissions) as a function of time after solution irradiation. This is shown in the plot given in Fig. 4.

As with ^{131}Sn, there is no evidence for precursors of

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[13] A. V. Pohm, R. C. Waddell, and E. N. Jensen, Phys. Rev. **101**, 1315 (1956).

TABLE II. Independent and cumulative fission yields (in %).

^{131}Sn[a]	^{131}Sb	^{132}Sn[a]	^{132}Sb	^{132}Te	^{133}Sn[a]	^{133}Sb[a]	^{133m}Te	^{133g}Te	^{133}I
0.91[b]	1.85	0.63[b]	2.93	0.96	<0.015	3.84	2.21	2.00	0.26±0.08
1.30[b]	1.58	0.52[b]	3.20	1.18		2.80	2.20	2.21	0.15±0.03
1.15[b]	2.13	0.63[b]	3.14	0.99		3.15	2.24	2.07	0.49±0.14
1.66[b]	2.31	0.57[b]	3.08	0.92		3.24	1.96	1.99	
1.34[b]	1.95	0.47[c]	3.09	0.62		3.25	1.65		
1.37[b]	1.39	0.67[c]	2.76	0.68		2.93	2.84		
1.06[b]	1.16	0.64[c]	2.05	0.71		2.92	2.13		
1.24[b]	1.00	0.59[c]	3.62	0.83		3.25	2.64		
1.06[c]	1.89		3.02			2.96	2.10		
1.05[c]	1.77		3.17			3.09	1.95		
1.18[c]	1.48		2.48			1.88	2.17		
1.55[c]	1.36		1.66			3.40			
1.30[c]			0.95			2.76			
1.31[c]			3.44			3.13			
1.57[c]						3.00			
1.45[c]						3.05			
						3.22			
Average values									
1.28±0.21	1.66±0.40	0.59±0.17	2.76±0.35	0.86±0.10	<0.015	3.05±0.39	2.19±0.33	2.07±0.31	0.17±0.06

[a] Cumulative fission yields; values for remaining radionuclides are independent fission yields.
[b] Indicates tin procedure involving sulfide precipitation.
[c] Indicates tin procedure involving stannane production.

comparable half-lives and/or substantial yield relative to ^{132}Sn. In the determinations of half-life and cumulative yield, therefore, the data are considered to represent the cumulative yield of tin. The half-life of 1.00 ±0.17 min. was obtained from a least-squares fit of the data, as contrasted to the value of 2.2 min reported by Pohm *et al.*[13]

In addition, studies of antimony in the 132 chain indicate that a perceptible growth occurs from precursor tin. The value for the tin half-life that is compatible with the antimony data is found to be 50±20 sec.

Calculations of the cumulative yield of the 132 chain through tin were made using the 1.00±0.17 minute half-life figure. The average cumulative fission yield through tin for mass-chain 132 was found to be (0.59±0.17)%. Values obtained in separate runs are given in Table II.

^{133}Sn

The experimental method used to obtain the ^{133}Sn half-life will not be described in detail in this paper. A similar technique has been used by others to measure the half-lives of fission-product rare-gases.[14]

The half-life of ^{133}Sn was determined by gas sweeping of stannane from an in-pile fission product solution, and the subsequent collection of the antimony daughter product on a charged wire. The distribution along the wire of the ^{133}I daughter activity was measured and used to determine the half-life of the tin precursor. The solution used to produce the stannane contained K_2CO_3(1M), 93.2% enriched ^{235}U as a uranyl carbonate complex, and KBH_4 (0.5M). The solution was adjusted to a pH of 11, and under these conditions it was found that the production of stibine (SbH_3) is greatly inhibited. However, the data obtained using this technique were not consistent, since the apparent half-lives obtained for ^{131}Sn, ^{132}Sn, and ^{133}Sn varied from run to run within wide limits. These erratic results are believed to be due to antimony contamination arising from stibine formation. The value here reported for the half-life of ^{133}Sn is that found when the observed half-lives of ^{131}Sn and ^{132}Sn from the charged wire experiments were in agreement with the values obtained by the methods described previously. The half-life is thus estimated to be 55 sec.

Yield values were obtained by tin sulfide separations followed by iodine determinations. The amount of ^{133}I found by gamma-ray spectrometry was not sufficient to firmly establish the cumulative yield of the 133 chain through tin. Because of this uncertainty and that of the half-life, the value is reported as a probable upper limit: 0.015%.

^{131}Sb, ^{132}Sb, and ^{133}Sb

The chemistry performed to determine the number of antimony atoms was similar for the three nuclides ^{131}Sb, ^{132}Sb, and ^{133}Sb. Antimony was separated from irradiated uranium solutions by its formation as stibine and recovered by the subsequent thermal decomposition of stibine.

Antimony separations were carried out at intervals following irradiations of predetermined duration; the separations were spaced such that the growth and decay of radioantimony could be observed. The number of atoms of radioantimony which were present at each

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[14] C. R. Dillard, R. M. Adams, H. Finston, and A. Turkevich, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), pp. 616–620 and 624–634.

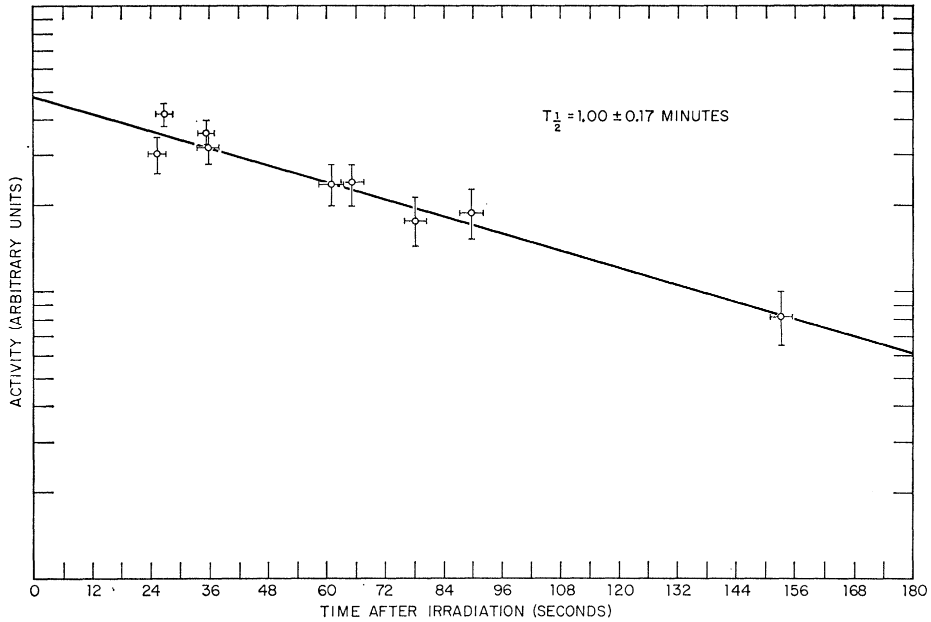

FIG. 4. Decay of ^{132}Sn.

separation time was determined by gamma-ray spectrometry of the radioiodine descendents associated with the appropriate antimony nuclide. With these data and the genetics of decay of antimony descendents, the growth and decay curves of the radioantimonies were calculated.

In the case of ^{131}Sb a half-life of 19.4±2.3 min was found. As the points of the decay curve were established by separate irradiations and separations, rather than the observation of direct decay, this was felt to be in reasonable agreement with the value of 23 min reported by Pappas.[15] The 23-min value was used in the yield calculations.

The average value of the independent fission yield for ^{131}Sb was determined to be (1.66±0.40)%. Yields calculated from individual runs are found in Table II.

The curve of ^{132}Sb growth and decay is presented in Fig. 5. The growth from the tin precursor is evident. The

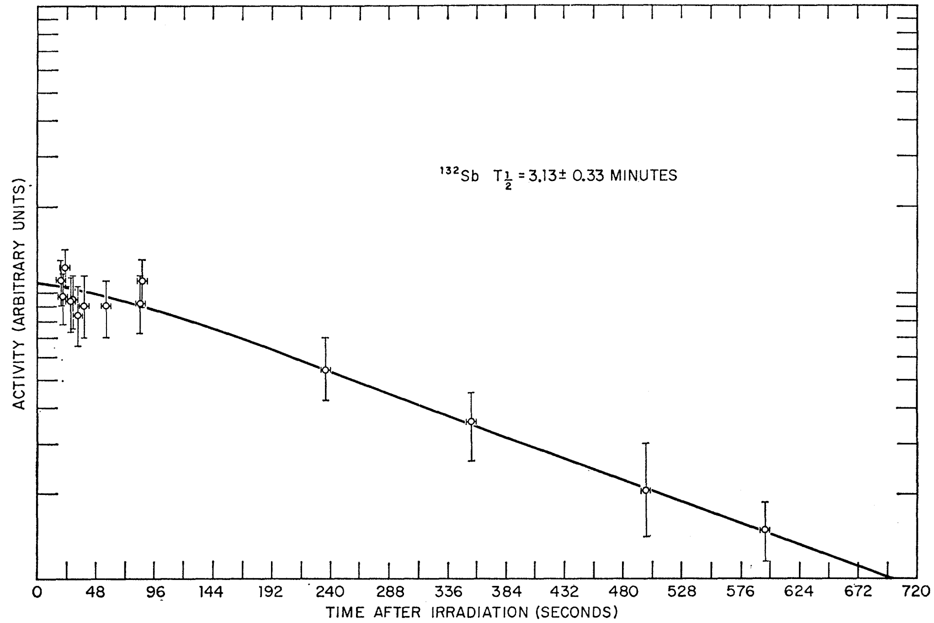

FIG. 5. Growth and decay of ^{132}Sb.

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[15] A. C. Pappas, MIT, Technical Report No. 63, 1953 (unpublished); U. S. Atomic Energy Commission Report No. AECU-2806 (unpublished).

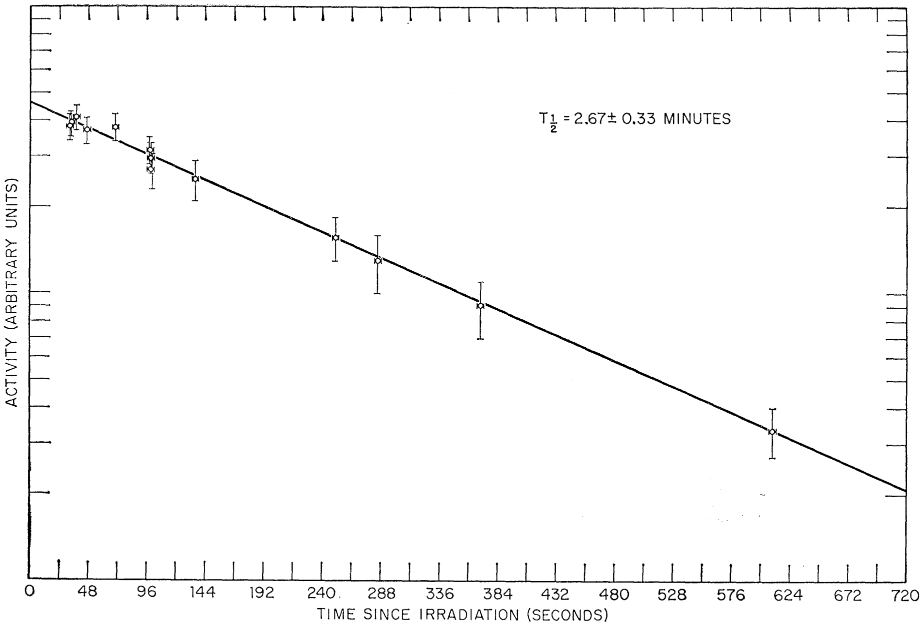

FIG. 6. Decay of ^{133}Sb.

line indicates the theoretical growth decay for ^{132}Sb with the tin and antimony half-lives and yields as given in this report. The half-life was determined to be 3.13 ±0.33 min for ^{132}Sb. This is longer than that reported by Pohm *et al.*[13] and by Pappas.[15] The average, independent fission yield was calculated to be (2.76±0.35)%; the individual yields are presented in Table II.

For ^{133}Sb, the decay curve given by Fig. 6 indicates that the yield of an ~55-sec tin precursor must be low. No contribution from precursors is evident. The half-life obtained was found to be 2.67±0.33 min. Previously reported values are 4.1 min by Pappas[15] and 4.2 min by Cook.[16] The average, cumulative fission yield of (3.05±0.39)% is derived from the values given in Table II.

^{132}Te, ^{133g}Te, and ^{133m}Te

Tellurium was chemically separated by sulfide precipitations from irradiated uranium solutions. Following tellurium isolation, iodine was separated to enable mass assignments to be made and to quantitatively determine radioiodine. Precursor yields and chain genetics enabled calculations to be made of tellurium independent fission yields. The independent yield of ^{132}Te was found to be (0.86±0.10)% which was obtained from the data of Table II.

In attempts to define more precisely the half-life of ^{133g}Te, which was determined by Pappas[15] to be approximately 2 min and by Prussin and Meinke[17] to be 12.45 min, beta-decay studies were undertaken on freshly separated tellurium in the presence of coprecipitated selenium. Upon resolution of the decay curves, a component with a half-life between 12 and 17 min was found. Due primarily to the presence of shorter lived selenium fission products, this was the limit of the resolution. However, the additional consideration of activity levels and half-lives expected from ^{84}Se and ^{85}Se relative to ^{133g}Te indicated that the activity was that of ^{133g}Te. More conclusive experiments (discussed later) were carried out by following the growth of ^{133}I which support the half-life value given by Prussin and Meinke.

As ^{133}Te is indicated to exist as two isomers with half-lives of 50 min[18] for the upper state and 12.45 min[17] for the ground state, the method for independent fission-yield determinations was as follows: Tellurium separations were made as rapidly following irradiations as possible. The separated tellurium samples were allowed to stand for about 1 h after which iodine was quantitatively extracted from them. The tellurium samples were retained and iodine was again separated, usually about 1 day later. The quantities of ^{133}I found in these extractions as determined by gamma-ray spectrometry allowed the calculation of independent fission yields for both isomers, if the chain relationships given in Fig. 7 for the ^{133}Te isomer pair and their precursors are assumed. The independent yields found were (2.07±0.31)% and (2.19±0.33)% for ^{133g}Te and ^{133m}Te, respectively, as given in Table II.

^{133}I

Iodine was extracted from irradiated uranium solutions through the use of the periodate chemistry de-

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[16] G. B. Cook, Atomic Energy Research Establishment Report No. AERE C/R-424, 1949 (unpublished).

[17] S. G. Prussin and W. W. Meinke, Radiochim. Acta 4, 79 (1965).

[18] J. M. Ferguson, D. L. Love, and D. Sam, J. Inorg. Nucl. Chem. 24, 1 (1962).

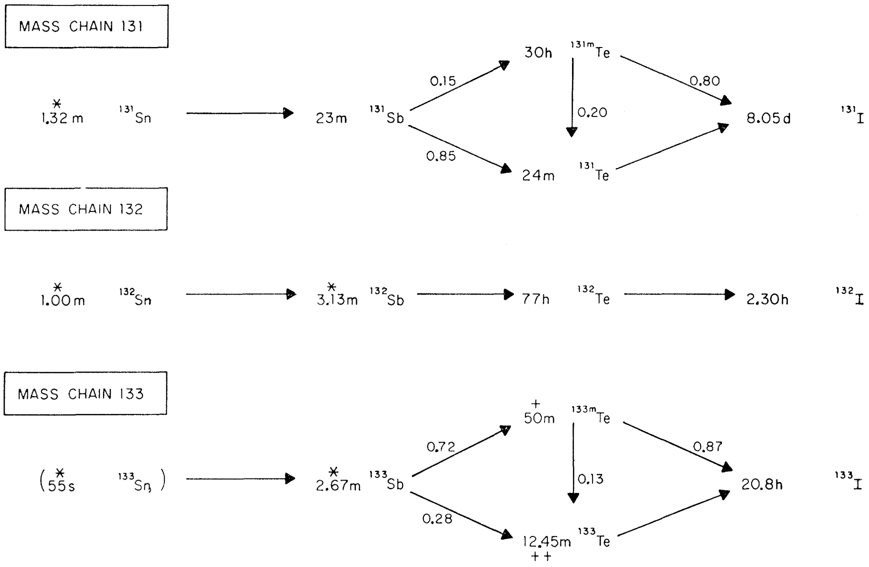


FIG. 7. Genetic relationships for the mass chains 131, 132 and 133. All branching and half-life values are from Katcoff's compilation [S. Katcoff, Nucleonics **18**, 201 (1960)] except: *—this work; +—Ref. 18; ++—Ref. 17.

scribed elsewhere.[8] The separations were carried out at time intervals chosen to establish the independent yield of ^{133}I and, in addition, to provide additional information on the half-life of ^{133g}Te.

The ^{133}I was quantitatively determined by gamma-ray spectrometry. Curve B is the expected growth curve for ^{133}I were ^{133g}Te to have a 12.45 min half-life and is seen to conform well with the experimental data. Curve A is the growth curve were the ground state of tellurium to have a half-life of approximately 2 min. The magnitude of the determinate error is sufficiently low that the 12.45 min half-life is strongly suggested. This information as well as data obtained from beta-decay studies of tellurium-selenium appears to confirm the value obtained by Prussin and Meinke.[17] Their value was therefore used in the calculations which determined the mean independent fission yield of ^{133}I to be (0.17±0.06)%. This yield was obtained from the data reported in Table II.

Figure 8 indicates that the growth of ^{133}I from precursors is rapid. Because of this, the most accurate determinations of the independent yield of ^{133}I are those obtained from separations carried out shortly after irradiation. The values listed in Table II are those corresponding to the three earliest iodine separations. Because of the large differences in errors associated with these determinations each value was weighted inversely by the square of its error in establishing the average value.

IV. DISCUSSION AND CONCLUSIONS

The charge distributions of the mass chains studied may be determined from the yield data given in Table III. The second column of Table III gives the independent and cumulative yields determined in this work. Included in the 131-mass-chain data is the fractional independent yield of the ^{131}Te isomer pair

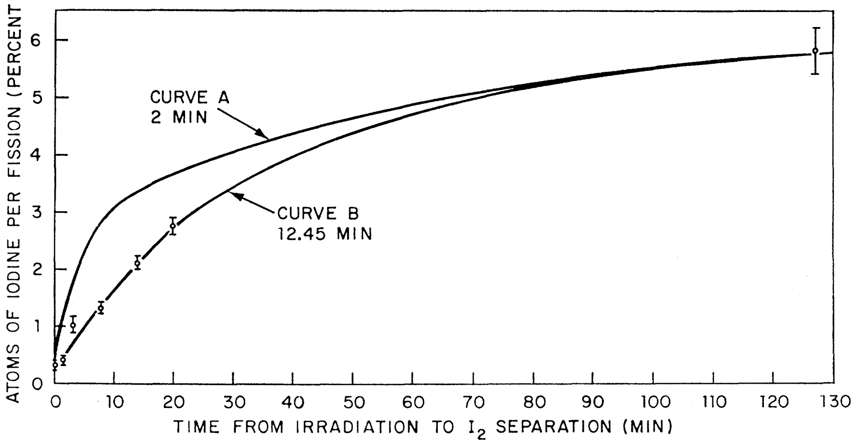
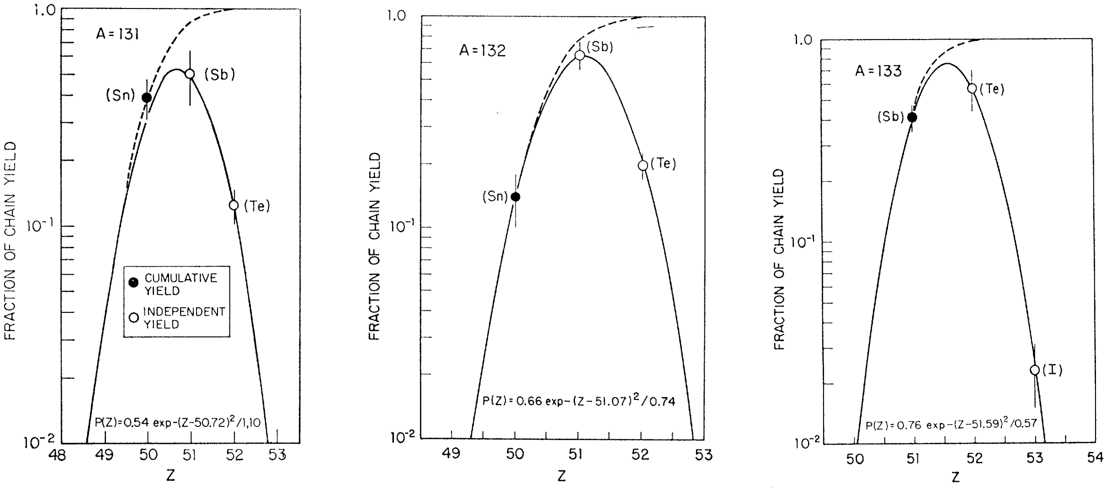


FIG. 8. Growth curve of ^{133}I.



(a) (b) (c)

FIG. 9. Charge distribution functions of mass chains 131, 132, and 133. The fractional cumulative yields at integral values of Z may be obtained from the dashed-line curves. [The ^{131}Te yield is that of Sarantites *et al.* (Ref. 19)].

determined by Sarantites and co-workers.[19] The total yield of each mass chain, obtained by taking the sum of the individual yields listed, is given in the third column of the table. Since in each chain the nuclides indicated in Table III (plus precursors) are believed to account for more than 99% of the total chain yield, the use of the sum of these yields to determine the fractional yield distributions is felt to be justified. These chain yields agree, within the error limits quoted, with the more precise mass spectrometric data, such as those of Farrar and Tomlinson.[9] Despite the greater precision of the mass spectrometric data, the chain-yield values obtained in this investigation were used for fractional-yield determinations to eliminate, to some degree, the effect of systematic errors which might appear in the data obtained here.

TABLE III. Experimentally determined and derived quantities relating to charge distribution.

Fission product	Independent yield (%)	Chain yield (%)	Fractional yield	Z_p	c
^{131}Sn	1.28±0.21[a]	3.30±0.45	0.39±0.08	50.72± 0.07	1.10± 0.05
^{131}Sb	1.66±0.40		0.50±0.14		
^{131g}Te ^{131m}Te	0.36±0.04[b]		0.124±0.014[c]		
^{132}Sn	0.59±0.17[a]	4.21±0.40	0.14±.04	51.07± 0.06	0.74 0.08
^{132}Sb	2.76±0.35		0.66±0.10		
^{132}Te	0.86±0.10		0.20±0.03		
^{133}Sb	3.05±0.39[a]	7.48±0.60	0.41±0.06	51.59± 0.04	0.57± 0.05
^{133g}Te ^{133m}Te	2.07±0.31 2.19±0.33		0.57 ±0.13		
^{133}I	0.17±0.06		0.023±0.008		

[a] Cumulative yield.
[b] Derived from Sarantites' fractional independent yield and the total chain yield as given by Katcoff [S. Katcoff, Nucleonics **18**, 201 (1960).]
[c] D. G. Sarantites *et al.*, Ref. 19.

Figure 9 shows the fractional-yield data for each of the mass chains plotted against atomic number. Superimposed upon these data are Gaussian curves, determined in the same manner as described by Nethaway,[20] which provide the best fit to the yields. The equations of best fit for each mass chain are given in the figure and are of the form

$$P(Z)=A\ \exp-(Z-Z_p)^2/c$$

where $P(Z)$ is the fractional independent yield of the radionuclide with charge Z, A is the maximum value of the function, Z_p is the most probable charge (mean of distribution), and c is a measure of the width of the distribution.

The values of c and Z_p obtained by this treatment of the data are given in Table III. The values of c are seen to decrease monotonically with increasing chain mass number. Recent data on mass chain 134 show a smooth continuation of this behavior.[21] The experimental errors associated with the data presented here are small in comparison to the magnitude of the variation in the width of the charge distribution curves. It is thus concluded that no single charge distribution function is applicable to all residual mass chains. Gordon and Aras[22] arrive at the same conclusion from calculations of neutron evaporation in fission, under the assumption that prompt fragments are produced with a single charge distribution applicable to all mass chains. The absence of a single charge-distribution curve for all mass chains for low-energy fission will

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[19] D. G. Sarantites, G. E. Gordon, and C. D. Coryell, Phys. Rev. **138**, B353 (1965).

[20] D. R. Nethaway, Ph.D. thesis, Washington University, 1959, (University Microfilms, Ann Arbor, Michigan, L. C. Card No. MIC, 60–63) (unpublished).

[21] P. O. Strom, A. E. Greendale, and A. A. Delucchi (to be published).

[22] G. E. Gordon and N. K. Aras, in *Symposium on the Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965).

clearly require modifications of previously estimated quantities derived on the basis of a unique charge-distribution curve and of interpretations derived therefrom. Thus, estimated values of Z_p and conclusions based on such values, e.g., influence of closed nuclear shells on charge distribution, will need to be re-evaluated.

The Z_p values reported here compare reasonably well with those given by the Z_p function of Wahl *et al.*[2] and by the ECD rule[3] despite the rather large deviation in *c* for the three mass chains.[23] An examination of the yield data which were available to Wahl *et al.* to define the Z_p function in this mass region shows that the agreement with the values of Z_p reported here is somewhat fortuitous. The data available were for nuclides not far removed from Z_p and thus variations in the width as observed here would have only small influence on the values estimated for Z_p.

Shell effects have been predicted[4–6] which could perturb charge distribution. In particular, ^{132}Sn was expected to be produced in relatively high yield. Such an enhancement would have caused Z_p for the 132 chain to take a value close to 50. A result has been presented in the literature indicating this to be the case. Konecny *et al.*[24] have determined a value of 49.98 for Z_p of the 132 mass chain by mass separator methods. However, each of the values of tin, antimony, and tellurium measured in the 132 chain reported here contradicts the existence of any such enhancement. Similarly, a prediction[6] of an enhanced yield of ^{133}Sb, based on a preference for the 82 neutron shell configuration, was not borne out by the measurements on the 133 chain. The measurements reported here thus do not indicate any detectable effect of nuclear shells on charge distribution in residual mass chains 131, 132 and 133.

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[23] Recent work on charge distribution within the 134 mass chain indicates that there exists a trend away from the Z_p function of Wahl *et al.* in the mass region 131–136; P. O. Strom *et al.* (unpublished).

[24] E. Konecny, H. Opower, H. Gunter, and H. Gobel, in *Symposium on the Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965).