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Roger C. Pettersen

(M. S. Thesis)

October 1963

PURIFICATION METHODS USED FOR THE MASS-SPECTROMETRIC STUDY OF FISSION OF U^{235} AND U^{238} AT MODERATE ENERGIES

Contents

PURIFICATION METHODS USED FOR THE MASS-SPECTROMETRIC STUDY OF FISSION OF U^{235} AND U^{238} AT MODERATE ENERGIES

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ABSTRACT

The sources of natural rare-earth contamination in U_3O_8 pressed-powder targets for cyclotron bombardments have been investigated further and studied more systematically. Mass-spectrometric analysis with an enriched Nd^{144} spiking solution is employed to follow the successive steps of target preparation, and to check the purity of the reagents and handling materials. Target purity is at the level of 2 parts per billion (ppb) for Nd^{144} when it is ready for cyclotron bombardment.

A solvent-extraction method for Ce(IV) is modified to remove cerium-fission products quickly and quantitatively from a bombarded target. We hoped to determine yields for cerium and praseodymium isotopes, but natural contamination in the extraction procedure prevented accurate measurements.

A previously known technique was used to determine independent fractional-chain yields for the mass-135 chain for fission of U^{238} and Th^{232} with 60-MeV helium ions. The independent fractional yields were found to fit a smooth Gaussian curve that was somewhat broader than that for lower-energy bombardments reported elsewhere. Relative independent yields for some of the cesium isotopes and relative cumulative yields for some cerium isotopes are also reported

I. INTRODUCTION

The mass spectrometer is very useful in the study of fission yields because of its high precision and its ability to measure the stable products as well as the radioactive species. Its high precision permits relative yields to be measured to within an accuracy of about 1%. However, natural contamination of stable fission products from external sources of the same isotopes may mask the true fission yield. One region of the fission-product range of interest for study in the mass spectrometer is that of the rare earth isotopes, because they are easily analyzed from a thermal ion source, and because of their relatively low abundance in nature. For a cyclotron bombardment with a total integrated beam of 50 to 60 microampere-hours (μA-h), about (1 to 5) $\times$ 10^{-9} g of each of the isotopes of interest is produced. In many cases, the amount of natural contamination is equal to or greater than the yield of the reaction, thus preventing measurements in some important regions.[1,2] The first part of this work is a systematic study to tie together the efforts of other workers to determine the source or sources of the contamination.[1,2] Each step in the target preparation and handling was checked by use of a Nd^{144} spiking solution, a method used in mass spectroscopy to determine quantitatively the amount of natural contamination in a sample. Neodymium was chosen as the contaminant because it is easily analyzed on a single iridium filament in the mass spectrometer and is representative of the abundance of the other rare earths.[3]

The second portion of this work is a report of a method used by Peppard et al. at Argonne National Laboratory to quickly and quantitatively extract Ce(IV) into an organic solvent, while quantitatively leaving the +3 rare earths behind.[4] The procedure was modified to make the Ce(IV) as "clean" of natural contaminants as possible. The purpose was to obtain the independent fission yields of Pr^{143} and Pr^{144} by removing the cerium as soon as possible after bombardment. (The independent fission yields are those produced as a direct result of fission before any decay has occurred). The praseodymium was

allowed to decay to neodymium, after which mass analysis of cerium and neodymium would show the yields for the praseodymium isotopes. Large amounts of natural neodymium and cerium contamination prevented measurements from being made. Relative cumulative yields for cerium were determined and are reported in the appendix.

The third portion of this work is an extension to higher energy of a study of the distribution of the independent yields for the isotopes of a given mass chain (all elements with the same atomic weight). The independent fractional chain yields (the independent yield divided by the total yield for the particular mass chain) are plotted as functions of their atomic number. The method was that used by McHugh to determine the independent fractional chain yields for isotopes of the mass-135 chain.[5] Charge-distribution curves were found for the helium-ion-induced fission of U^{238} and Th^{232} near 60 MeV.

II. TARGET PURIFICATION

Previous workers attempted to improve upon the purity of U_3O_8 pressed-powder targets used in cyclotron bombardments for the mass-spectrometric study of nuclear fission.[1,2] A thorough study of the reagents and handling steps is described here, and some changes are introduced to improve target purity. The contamination level was reduced to about a tenth that of previous targets.

A Nd^{144} spiking solution was employed throughout as a measure of the neodymium present as natural contamination. (This solution was prepared and checked by Dr. Y. Y. Chu.) Neodymium was considered representative of the rare earths.[3] The spiking solution was employed in this manner. This entire procedure was carried out in a dust-free box. A sample of HCl to be analyzed was spiked with a known amount of the Nd^{144} spike, then placed in a dust-free box and evaporated to dryness under an infrared lamp. The sample was taken up with a drop of water in a pipet and evaporated on an iridium filament that had been cleaned and checked for neodymium in the mass spectrometer. The ratio of Nd^{144} to Nd^{143} was measured and used with the known spike ratio to calculate the amount of natural Nd^{144} contamination. The equation was

$$x = \frac{M[\, 1 - (R_m/R_s)]}{(R_m/2) - 1} ,$$

where x is the amount in grams of Nd^{144} contamination, M is the amount in grams of Nd^{144} added as the spike, R_m is the measured or observed ratio of Nd^{144} to Nd^{143}, and R_s is the known spike ratio for Nd^{144} to Nd^{143}.

All reagents and materials, and each step of the target-making procedure, have been carefully checked by use of the Nd^{144} spike to determine if there is any one major source of rare earth contamination. Table I shows the results of the analyses.

The quartz-distilled water was prepared by first passing water from the house distilled-water line through a polytetrafluoroethylene (Teflon) column packed with mixed-bed resin (AG 501-X8) obtained

Table I. Results of Nd^{144}-spiking analysis for U_3O_8 purification.

	Substances analyzed	Natural Nd^{144} contamination
1.	Quartz-distilled water	6.2×10^{-13} g/ml
2.	12 $\underline{M}$ HCl evaporated in a quartz tube	5.5×10^{-12} g/ml
3.	12 $\underline{M}$ HCl evaporated in a Teflon tube	6.2×10^{-13} g/ml
4.	10 $\underline{M}$ HNO_3	1.5×10^{-12} g/ml
5.	12 $\underline{M}$ HCl through quartz ion-exchange column	6.4×10^{-10} g/ml
6.	12 $\underline{M}$ HCl through Teflon ion-exchange column	3.4×10^{-12} g/ml
7.	Purified uranium solution	5.0×10^{-11} g/100 mg
8.	U_3O_8 powder	1.3×10^{-10} g/100 mg
9.	U_3O_8 powder after pressing. Purified by N. Souka	1.1×10^{-9} g/100 mg
10.	U_3O_8 powder after pressing. Purified by R. Pettersen	1.2×10^{-10} g/100 mg
11.	Platinum foil	2.9×10^{-10} g/100 mg
12.	Copper foil	6×10^{-11} g/100 mg
13.	Gold foil	3×10^{-10} g/100 mg

from Bio-Rad Laboratories. The resin was held in the column by Teflon shavings packed in the bottom. This deionized water was then doubly distilled from a quartz still and collected in a polyethylene container.

The HCl was prepared by passing the gas through quartz-wool filters and bubbling it into the quartz-distilled water in a polyethylene container. The HNO_3 was chemically pure stock reagent (Baker and Adamson.)

As seen from lines 2 and 3 in Table I, strong acids evaporated in quartz increase the neodymium contamination by a factor of ten or more. Apparently strong acids at elevated temperatures (such as those used in distillation) dissolve a small portion of glass or quartz that contains rare earths, whereas concentrated HNO_3 stored in a glass bottle at room temperature does not. Thus all subsequent samples were evaporated to dryness in Teflon cones. Teflon ion-exchange columns were machined in the chemistry mechanical shops and used in place of quartz.

Briefly, the method for uranium purification and target preparation was as follows: 12 M HCl containing a few drops of 10 M HNO_3 was used to dissolve U^{235} foil, and the solution was boiled for a short period to expel any excess HNO_3.[1,2] The yellow uranium solution was adsorbed on an anion column that had been thoroughly washed with various concentrations of HCl. Several column volumes of 12 M HCl were passed through to remove the rare earths from the uranium, and the uranium was then stripped off with 0.1 M HCl. The uranium solution was made 12 M in HCl again by bubbling HCl gas through it, and the uranium was readsorbed and washed on another identical column. This was repeated a third time, after which the uranium solution was finally collected and stored in a polyethylene bottle.

A portion of the uranium solution was treated with ammonia gas, which precipitated the hydroxide. The precipitate was transferred with a clean polyethylene pipet to a clean platinum crucible and carefully ignited in an oven at 850°C, which converted the precipitate to

black U_3O_8 powder. This powder was distributed with a stainless steel spatula on a copper backing plate that had grooves 0.003 inch deep machined into it. Since stainless steel is known to be highly contaminated with natural rare earths,[6] we tried using gold-plated spatulas, but these did not decrease the contamination. The powder was covered with a clean platinum foil 0.0005 inch thick, and then pressed at 8000 to 9000 pounds with a hydraulic press. This assured nearly uniform target thickness and good thermal contact with the backing plate. The target was then mounted on a target block for the 88 inch cyclotron and covered with 0.001-inch platinum foil securely fastened with a cover plate. It was then ready for bombardment.

Line 6 of Table I gives the amount of natural neodymium contamination resulting from passing a spiked sample of "clean" 12 $\underline{M}$ HCl through a Teflon ion-exchange column. Line 7 is the same thing for a purified 12 $\underline{M}$ HCl solution of uranium, and shows a greater than tenfold increase of the natural contamination. Although we assume that rare earths pass right through an ion-exchange column in strong HCl, we performed a double spiking experiment to make sure some of the neodymium wasn't being held up on the column and coming off when the uranium was stripped with 0.1 $\underline{M}$ HCl.

A Nd^{148} spike solution was prepared with an enriched isotope from Oak Ridge, and cross-checked against the Nd^{144} spike. A weighed amount of U_3O_8 powder was dissolved with 12 $\underline{M}$ HCl and a few drops of 10 $\underline{M}$ HNO_3. The uranium solution was boiled slightly to remove the HNO_3 and then spiked with Nd^{144}. The spiked solution was transferred to an ion-exchange column. The uranium was adsorbed and the liquid that passed through collected and divided into two equal portions. One was analyzed for neodymium as it came off the column, and the other was spiked, with an amount of Nd^{148} roughly equivalent to half the original Nd^{144} spike, and then analyzed. The second analysis furnished direct proof of the percent recovery of neodymium from the column, and was found as expected, to be very close to 100% of the contamination found in the first analysis. Both analyses

determined the amount of neodymium contamination originally in the uranium oxide powder, and were found to agree. Both gave a value consistent with that of line 8 of Table I, a separate analysis of the U_3O_8 powder. On the basis of this evidence, one concludes that re-peated washings of uranium adsorbed on an ion-exchange column should remove most of the rare earth contamination.

The same uranium adsorbed on the ion-exchange column was thoroughly washed with "clean" 12 M HCl and stripped with 0.1 M HCl. The ion-exchange column was washed with various concentrations of "clean" HCl and the uranium solution respiked with Nd^{148}, then re-adsorbed on the column in 12 M HCl. Analysis of the neodymium that passed through the column showed that it contained only about one-tenth as much contamination; this is also consistent with line 7 as com-pared with line 8 of Table I. No enrichment of the Nd^{144} was noted, strongly suggesting that the contamination is coming directly from the ion-exchange column. Apparently uranium stripped off a column may bring with it from the resin some neodymium, which normally is not re-leased while the uranium is being washed. This is a possibility, since it has been found that the resin does contain about 10^{-8} gram of neo-dymium per gram of resin, and the column has about 2 to 3 grams of resin packed into it.[6] Thus the resin could be a large reservoir of contamination from which small amounts ($\approx 10^{-10}$ gram) are released each time the uranium is stripped from the column.

Lines 7 and 8 of Table I show that the powder is slightly more contaminated than the purified uranium solution, the contamination being probably introduced during the handling and ignition procedures. The metal foils were checked to make sure they weren't contributing rare earths during the pressing operation. As shown in Table I, it would be impossible to account for the observed contamination from any of the contact metals in the pressing operation, as they are relatively free of rare earths.

The uranium and metal-foil samples shown in Table I were pre-pared by dissolving the material in 12 M HCl with a portion of 10 M

HNO_3 added. (The gold and platinum foil were obtained as stock from the chemistry store room, and the copper from the laboratory metal rack.) The solution was boiled and converted to the chloride. A measured quantity of the Nd^{144} spike was added and the solution transferred to a clean ion-exchange column. The metal ions were adsorbed and the neodymium passed through and analyzed.

In summary, it has been found that through the use of Teflon instead of quartz for the ion-exchange columns and in the evaporation step, rare-earth contamination was decreased to about one tenth. Each step in the target preparation seemed to introduce small amounts of rare earths. It is suggested that the anion-exchange resin in some way introduces rare earths as the uranium is stripped from the column. Most other materials and reagents were found to be relatively free of rare earths.

III. CERIUM EXTRACTION

It has been reported that 0.75 $\underline{M}$ di-2-ethyl hexyl orthophosphoric acid (HDEHP) in heptane will extract Ce(IV) from 10 $\underline{M}$ HNO$_3$ with a distribution coefficient of greater than 8×10^3.[4] The distribution coefficient for the remaining +3 rare earths is about 10^{-3}, thus giving a separation factor of about 10^7. We wanted to make use of this selective extraction to measure the independent yields of Pr^{143} and Pr^{144} by separating them from their cerium parents soon after bombardment and letting the praseodymium decay to neodymium. Relative measurements of the cerium- and neodymium-isotope ratios would be sufficient to determine the praseodymium independent yields and fractional-chain yields.

Peppard et al. used 1 $\underline{M}$ NaBrO$_3$ to oxidize Ce(III) to Ce(IV) at tracer-level concentrations.[4] HDEHP and $\underline{n}$-heptane (1:3 by vol.) were shaken with the acid phase to extract the Ce(IV). The Ce(IV) was back-extracted with 3% H$_2$O$_2$ in 10 $\underline{M}$ HNO$_3$.

Since sample preparation of any type is often found to introduce a certain amount of natural contamination, this procedure was checked with Nd^{144} spiking experiments, and some modifications were made before using it on a target run. An ozone generator was patterned after one found in the literature,[7] as the use of ozone would be a "clean" and efficient method for oxidizing Ce(III) to Ce(IV). The generator was a discharge type consisting of an inner soft-glass tube for a mercury electrode, surrounded by a glass jacket through which oxygen entered at the top and left from the bottom. This was immersed in 7.7 $\underline{M}$ ZnCl$_2$ solution, which acted as the second electrode. A high-voltage transformer supplied from a variable auto-transformer gave an 11,000-volt discharge in the tube. The ozone content was roughly determined at 2% by means of displacing a known volume of water in an inverted flask with the gas. A solution of KI was quickly added, the flask stoppered, and then shaken. The iodine formed was titrated with 0.05 $\underline{M}$ Na$_2$S$_2$O$_3$.

A trial run was made on a Ce^{144} tracer whose concentration was about 10^{-9} g/ml, but oxidation did not occur. Although oxidation with the ozone was attempted on the tracer at temperatures ranging from

0°C to 60°C, and also at varying acid concentrations, the attempts were unsuccessful. When tried with macro quantities (10^{-3} g/ml) of tracer, however, oxidation occurred. But since the fission-yield level is about 10^{-9} to 10^{-10} g per target, there was no need of trying it on intermediate concentrations. A higher ozone concentration may have helped to oxidize the tracer, but there was no other source available.

The oxidation by 1 $\underline{M}$ NaBrO$_3$ in 10 $\underline{M}$ HNO$_3$ was tried and verified, but this introduced large amounts of contamination by natural cerium and other rare earths. A series of extractions was used to purify the reagents. The +3 rare earths were removed from the HDEHP by purified HNO$_3$, into which they are extracted with a distribution ratio of 10^3. It was found that the HDEHP will extract +3 rare earths quantitatively from a slightly basic solution, so that the purified HDEHP was used to remove the +3 rare earths from a slightly basic 2 $\underline{M}$ NaBrO$_3$ solution. This solution was then diluted with the 10 $\underline{M}$ HNO$_3$ to the desired concentration for the oxidation step. The 0.1 $\underline{M}$ NaBrO$_3$ in 10 $\underline{M}$ HNO$_3$ was sufficient to oxidize the cerium, but 0.05 $\underline{M}$ NaBrO$_3$ was not strong enough.

A Nd^{144} spike was added to the purified and slightly basic 0.1 $\underline{M}$ NaBrO$_3$ and extracted with pure HDEHP, and then back-extracted with 10 $\underline{M}$ HNO$_3$. Mass analysis showed 2×10^{-10} g of contaminating Nd^{144} per ml, which is below the fission-yield level. Teflon containers were used throughout for this experiment.

A 50-MeV helium-ion bombardment of a U$_3$O$_8$ pressed-powder target was scheduled on the 88-inch cyclotron. A few hours after the bombardment, the target was dissolved and the cerium separated from the other rare earths. The samples were highly contaminated with rare earths, so only relative cumulative yields for some of the cerium isotopes could be measured. These are listed in the appendices. To give an estimate of the amount of contamination, the following procedure was used:

The helium-ion beam was degraded by an aluminum isolation foil 0.002 inch thick and a platinum cover foil 0.001 inch thick, both of

which were accurately checked to find the weight per unit area. The tables of Williamson et al. were used to estimate that the 50-MeV particles were reduced to an average energy of 39.8 MeV in the U_3O_8.[8] For this energy, a cross section for the chain with mass 142 was taken from the literature.[9] This along with the total integrated beam current allows an estimate of the total absolute yield of all elements with mass 142 formed in the reaction. The fraction of this chain that is found as Ce^{142} can be estimated by predicting a value for the most-probable charge, Z_p for this mass chain. This can be done using the data given by McHugh.[5] The difference between the cerium atomic number and the Z_p is then fitted to the known Gaussian curve, which describes the fractional chain yield as a function of charge. This gives the fractional chain yield for the Ce^{142}. In this case, because of the short-lived predecessors, and the very low yield for isotopes of the same mass of higher Z, most of the mass-142 chain yield is Ce^{142}, when analyzed in the mass spectrometer. This estimate showed that 1×10^{-9} g of Ce^{142} was formed in the bombardment. By using the 142:140 ratio (see appendices) measured in the mass spectrometer, we can determine the relative amount of contaminant and reaction-formed Ce^{142}, and thus the absolute amount of contaminating Ce^{142}. This was estimated to be 7.8×10^{-9} g. A second bombardment furnished roughly the same amount of contamination; this possibly indicated a systematic source of rare earths.

It is rather difficult to explain this amount of contamination on the basis of presently known evidence. The target itself can most certainly be ruled out, as it has been thoroughly studied. There is some evidence at this time that the solvent-extraction separation is fairly "clean, " but it is difficult to trace the source to any one step. Yet some step or steps in the separation are the most likely suspect, as they were not as thoroughly studied as the target preparation. The HDEHP or the peroxide may be contributors, or it may be that at each step in the handling and transferring of solutions small amounts are added, thus accounting for the large contamination. But further study is needed on the chemical-separation procedure before any definite conclusions can be drawn.
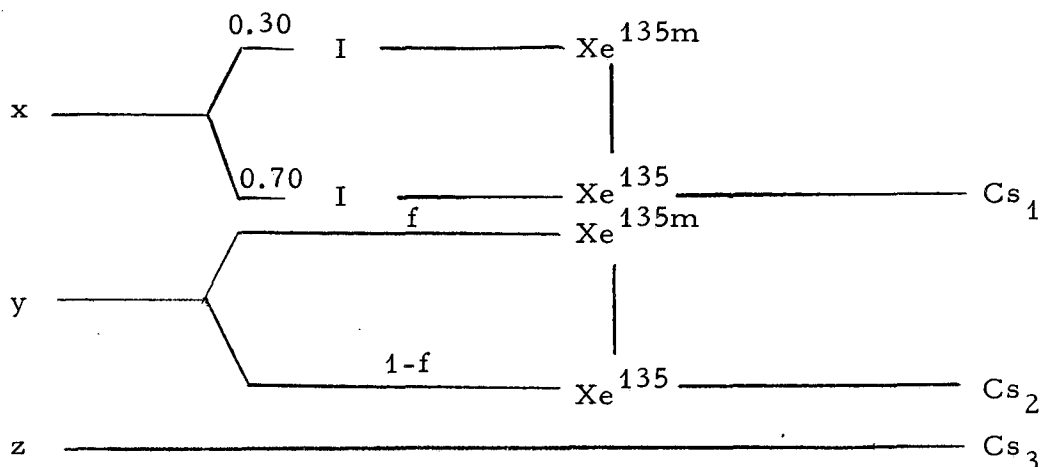
IV. CHARGE DISTRIBUTION FOR THE MASS-135 CHAIN

Previous workers have used an experimental procedure to measure the independent fractional-chain yields of Cs^{135} and Xe^{135}, and the cumulative fractional-chain yield of I^{135} formed from the helium-ion-induced fission of uranium isotopes and Th^{232}.[2,5] A mass-chain yield is the sum of the yields of all elements with the same mass number formed from the fissioning species. The independent fractional-chain yield is then the yield of one of these elements, formed as a direct result of fission, divided by the total yield for that mass chain. The cumulative fractional-chain yield is the yield of that element formed as a result of fission plus the sum of all the predecessor beta-decaying elements, divided by the total chain yield. We wanted to determine the independent fractional-chain yields of the fission products for a given mass chain as a function of their charge, or in other words, the relative probability of forming nuclides of various charges for a given mass chain.

This distribution with charge has been found[5] to fit a smooth Gaussian curve whose equation can be written as

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

where y_i is the independent fractional-chain yield of an element with charge Z, c is the width constant of the distribution, and Z_p is the most probable charge (not necessarily integral) for the fission products of the given mass number. By fitting a smooth Gaussian curve to the points found from the experiment to be described, width constants for the curve can be found. McHugh has found that a value of $c = 0.95(\pm 0.05)$ best fits the curve for all energies up to 39 MeV excitation for helium-ion-induced fission of Th^{232}.[5] Further assumptions made are that the distribution curve is constant for all mass chains and does not vary with different fissioning species. The Z_p changes, of course, but it is found that the width constant of the curve does not change at energies below 45 MeV. The width constant of this curve can be determined directly by calculating the independent fractional-chain yields for Cs^{135} and Xe^{135}, and the cumulative fractional-chain yield for I^{135}. The method and decay equations have been adequately described elsewhere and will be only briefly touched upon here.[2,5]

Consider the decay scheme for the 135 chain,

```
        0.30                              135m
       /──── I ──────────── Xe
  x ──┤                           │
       \                          │
        0.70  I ──────── Xe^135 ──────────────── Cs_1
              f          Xe^135m
              ┌──────────
  y ──┤                          │
       \                          │
        1-f ────────── Xe^135 ──────────── Cs_2

  z ────────────────────────────────── Cs_3
```

where x, y, and z are the independent production rates of I^{135}, Xe^{135}, and Cs^{135} respectively, and f is the fraction of the independent yield populating the Xe^{135m} isomer. Three equations give the contribution to the Cs^{135} from each of the independent production rates. The contributions from each are a function of the length of the cyclotron bombardment, T, and the time from the beginning of bombardment to the time, t, when the target is dissolved and the decay chain is broken. Three different times are used to cut off the decay to cesium— ≈ 2 h, ≈ 13 h, and several hundred hours after the start of bombardment. For each case the Cs^{135} is measured relative to the Cs^{137}. The Cs^{137} is a long-lived cumulative-chain yield, so it is constant for all three measurements. These three ratios, along with an estimated correction for the Ba^{135} fractional-chain yield, are sufficient data for a computer program to give the independent fractional chain yields. This computer program was written by Dr. J. A. McHugh.

For this experiment, circular 0.001-inch-thick metal foils of U^{238} and Th^{232} were each cut into three equal sectors and bombarded with helium ions in the 88-inch cyclotron. Each piece was dissolved at a measured time following the bombardment, and boiled slightly to expel the xenon.

McHugh reports that the iodine as well as the xenon is expelled by the boiling, but two attempts to verify this with I^{131} tracer in both HNO_3 and HCl solutions were not successful.[5] It was found that the anion-exchange column used to separate the cesium from the uranium adsorbs the iodine, whereas the cesium is allowed to pass through. McHugh ran the ion column immediately after the boiling step, and thus his data were not affected. The boiled solutions from the first run of this work were first stored and later run on the ion-exchange column just before mass analysis. Hence high independent yields were obtained for the iodine, and low independent yields for the xenon. A second experiment was tried, and the ion-exchange columns were run immediately following the boiling step to adsorb the iodine and remove it from the cesium. The results are given in Table II and Figs. 1 and 2 and are more compatible with a somewhat widened Gaussian curve than the results of the first run.

The distribution-width constants were found to be c = 1.25 for U^{238} + 58.1-MeV helium ions; and c = 1.10 for Th^{232} + 59.4-MeV helium ions. These results agree qualitatively with those found by Pate et al. for proton-induced fission of Th^{232} for proton bombarding energies between 8 and 87 MeV.[10] Pate et al. report a widening of the charge-distribution curve at about 25 MeV, which shows evidence of gradually increasing at higher energies. They explain this as being due to increased neutron emission before fission takes place.

Better data are needed on this problem before any definite statements can be made as to what is taking place. A recheck of the experiment is needed to be sure of the observed widening. It would be desirable to follow the widening for a given isotope for a wide range of bombarding energies up to about 120 MeV.
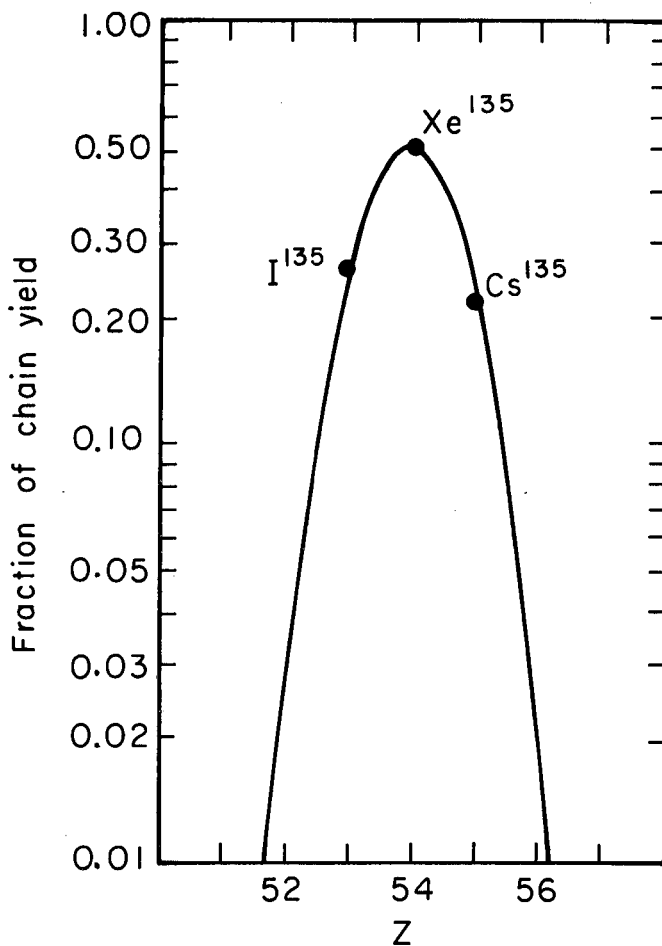
MU-32835

Fig. 1. Gaussian distribution curve of width c = 1.25 fitted to fractional-yield data for the mass-135 chain from U^{238} + 58.1-MeV He^4. The I^{135} point is a cumulative yield.
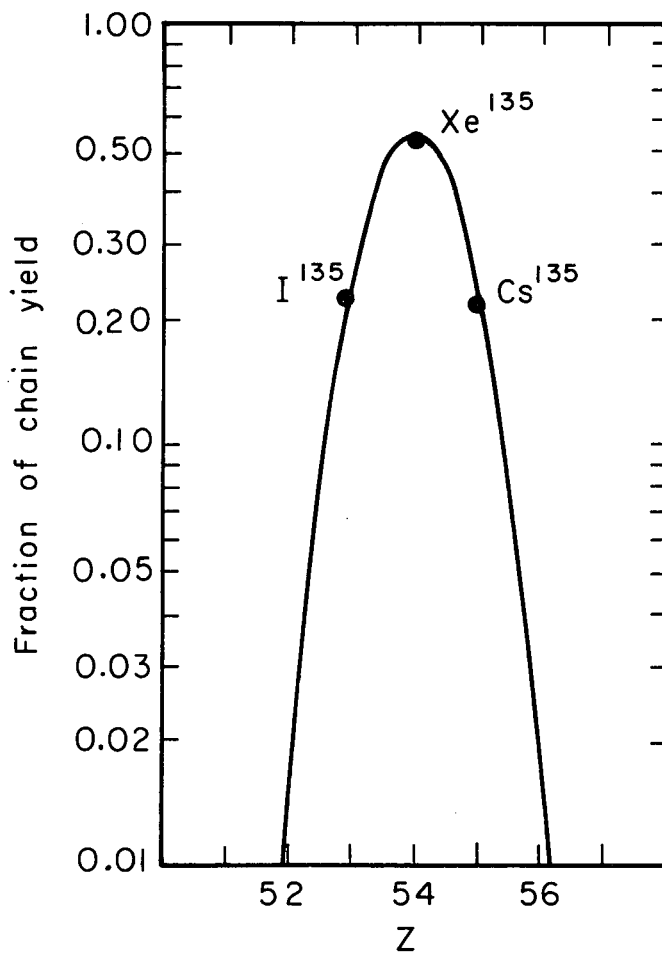
Fig. 2. Gaussian distribution curve of width c = 1.10 fitted to fractional-yield data for the mass-135 chain from the Th^{232} + 59.4 MeV He^4. The I^{135} point is a cumulative yield.

Table II. Independent fractional chain yields for mass 135 chain.

Yield ratio of Cs^{135}/Cs^{137}	$t(h)$[a]	$T(h)$[b]	$x(I^{135})$	$y(Xe^{135})$	$z(Cs^{135})$	Ba^{135}[c]
U^{238} + 58.1-MeV He^4						
0.3712±0.0023	1.88	0.800				
0.6830±0.0062	8.75	0.800				
1.351±0.0059	1040	0.850	0.2545	0.5013	0.2191	0.025
Th^{232} + 59.4-MeV He^4						
0.3857±0.0036	1.93	1.00				
0.8701±0.0073	13.57	1.00				
1.3532±0.0173	873.5	2.57	0.2221	0.5316	0.2313	0.0150

a. t is the elapsed time in hours from the start of bombardment to the time of dissolving the target

b. T is the length of bombardment in hours

c. Estimated fractional chain yield for Ba^{135}

ACKNOWLEDGMENTS

APPENDIX

A. Cesium yields measured relative to Cs^{137}

	Cs^{131}	Cs^{132}	Cs^{134}	Cs^{136}
U^{235} + 58.8 MeV He^4	0.03149±0.00055	0.1086±0.0014	0.5014±0.0088	0.7774±0.0201
U^{238} + 58.1 MeV He^4	0.1360±0.0014			
Th^{232} + 59.9 MeV He^4		0.1161±0.0055	0.1401±0.0013	0.4912±0.0033
U^{235} + 74.9 MeV He^4	0.0889±0.0025	0.2375±0.0131	0.7141±0.0415	0.7965±0.0060

B. Cerium yields measured relative to Ce^{142}

	Ce^{141}	Ce^{143}	Ce^{144}
U^{235} + 39.8 MeV He^4	0.9030±0.0248	0.9554±0.0565	0.6171±0.0190

Note: All measurements have been corrected for decay during and after bombardment.

The Ce^{142}/Ce^{140} ratio measured was 0.1418±0.0020. This was used to estimate the amount of natural cerium contamination.

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