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Publication Date

1951-05-28

UCRL- 1195

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R. L. Folger, P. C. Stevenson, and G. T. Seaborg

May 28, 1951

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Carbide and Carbon Chemicals Company (Y-12 Area)	17
Chicago Patent Group	18
duPont Company	19 - 23
General Electric Company, Richland	24 - 27
Hanford Operations Office	28
Idaho Operations Office	29 - 30
Iowa State College	31
Kellex Corporation	32
Knolls Atomic Power Laboratory	33 - 36
Los Alamos	37 - 39
Mound Laboratory	40 - 42
Naval Research Laboratory	43
New York Operations Office	44 - 45
North American Aviation, Inc.	46
Oak Ridge National Laboratory (X-10)	47 - 52
Patent Branch, Washington	53
Savannah River Operations Office	54
UCLA Medical Research Laboratory (Warren)	55
U. S. Naval Radiological Defense Laboratory	56
University of California Radiation Laboratory	57 - 60
University of Rochester	61 - 62
Wright-Patterson Air Force Base	63 - 64
Technical Information Service, Oak Ridge	65 - 79

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HIGH ENERGY PROTON SPALLATION - FISSION OF URANIUM

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May 28, 1951

ABSTRACT

The fission and spallation reactions caused in uranium by bombardment with high energy protons (340 to 350 Mev) were investigated. The reaction products were separated from the target by chemical processes and identified by their radioactive properties. The relative yields of the observed fission products were measured, and the results plotted as a function of mass number. Several of the spallation products were identified and their yields estimated.

An attempt was made to determine the most probable atomic number for a nuclide of given mass formed directly from fission. Studies were made of the relative yields along several isobaric chains as a function of atomic number. From these data predictions of the mass and charge of the fissioning nucleus are made.

Using the reaction $Al^{27}(p,3pn)Na^{24}$ of known cross section to monitor the bombarding beam, the formation cross sections for several fission product nuclides were measured. Values for these reference nuclides were used to transform the relative yields into formation cross sections. Integration over the range of mass numbers of the area under the curve of formation cross section as a function of mass number led to a value for the total fission cross section for uranium bombarded with high energy protons, assuming predominantly binary fission.

[*]Now at E. I. du Pont de Nemours and Co., Oak Ridge, Tennessee.

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I. INTRODUCTION

A. Summary of Previous Data on High Energy Fission

In recent years there has been considerable interest in the mechanism of fission induced by high energy particles (greater than a few Mev) as compared to that induced by low initial excitation. The greatest amount of work has been done with the target elements bismuth,[1] thorium,[2,3] and uranium.[3,4] High energy fission of other heavy elements from Z = 73 to Z = 82 has been observed.[5] Batzel and Seaborg[6] have announced the high energy fission of a number of medium weight elements.

The yield distribution of fission products has been studied by chemical separation techniques followed by identification of the nuclides through radioactivity measurements, and also by measurement of the kinetic energies of the fission fragments. The predominant characteristics of high energy fission which appear from these studies are:

(1) A single peak is found in the yield versus mass distribution curve, indicating symmetric fission.

(2) Evaporation of nucleons is evidenced by the mass number of the yield peak being less than one-half the mass of the target nucleus.

(3) The kinetic energies of the fission fragments seem to be practically independent of the initial target nucleus excitation.

Predominantly symmetric fission has been reported by Goeckermann and Perlman[1] (bismuth with 190 Mev deuterons), O'Connor and Seaborg[4]

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[*]Now at E. I. du Pont de Nemours and Co., Oak Ridge, Tennessee.

(uranium with 380 Mev helium ions), and Jungerman and Wright[3] (uranium with 90 Mev neutrons).* At intermediate energies, the contribution of asymmetric fission is shown by the appearance of two peaks with a shallow dip between them. This effect has been observed by Newton[2] (thorium plus 38 Mev helium ions), and Jungerman and Wright[3] (uranium plus 45 Mev neutrons).*

In the high energy deuteron fission of bismuth, Goeckermann and Perlman[1] observed symmetric binary fission in which the mass of the nuclei having the peak yield was less than one-half the mass of the target nucleus. The mass difference indicated the evaporation of 10 to 12 nucleons. The relative yields of the fission product isobars seemed to indicate that the most probable primary products were those having the same charge to mass ratio as the fissioning nucleus. Yamaguchi,[7] in a theoretical treatment of nuclear evaporation, has shown that for bismuth bombarded with 200 Mev deuterons, the half-width for neutron emission is greater than the half-width for fission until 10 to 11 neutrons have been emitted. In the case of uranium bombarded with 380 Mev helium ions,[4] the peak mass was not well enough defined to demonstrate nuclear evaporation although the curve showed a strong possibility that such particle evaporation had occurred.

Measurements of fission fragment kinetic energies have been made by Jungerman and Wright,[3] using ionization chamber techniques, and indirectly by Douthett,[8] measuring the ranges of the fragments in aluminum. In bombarding uranium with 45 and with 90 Mev neutrons, Jungermann[3] observed

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* Represents ionization chamber measurement of fragment kinetic energies.

the mean kinetic energies to be 79^{+}_{-} and 80^{+}_{-} 2 Mev, respectively. Within

the limits of experimental error, the ranges of fission fragments obtained

by radiochemical separation from absorber foils were respectively the same

for slow neutron fission[9,10] of U^{235}, and the fission of natural uranium

with 18 Mev deuterons.[8] In the case of uranium plus 335 Mev protons,[8]

there is a systematic lowering of the observed fission fragment energies

by about 6 to 8 Mev compared to slow neutron fission. Two explanations

have been advanced for this lowering of observed kinetic energy.[8] If

evaporation of nucleons takes place to a significant extent, the mass of

the fragment complementary to the observed fission fragment will be less

than that of the comparable fragment in the case of slow neutron fission.

The proportionately lighter fragment will carry off a relatively greater

amount of the available energy, leaving less for the observed fragment.

Another explanation is that evaporation of some charged particles leaves

less coulombic potential, hence less total available kinetic energy.

These results suggest that the actual fissioning nucleus in each

case is in a relatively unexcited state and that the high initial exci-

tation energy is not transmitted as increased kinetic energy of the frag-

ments.

B. Statement of the Problem

The purpose of this research was to investigate the fission and

spallation reactions caused in uranium by bombardment with high energy

protons (340 to 350 Mev). The reaction products were separated from the

target by chemical processes and identified by their radioactive properties.

Measurement was made of the relative yields of the observed fission products

and the results plotted as a function of mass number (Figure 1).

An attempt was made to determine the primary products (those formed directly from fission with highest probability for a given mass) for various mass numbers. An estimate of the location of primary products with respect to the stable nuclides may be made by observing the variation of independent yield (formed directly in fission rather than by growth from a radioactive "parent") with mass for a given element in the region of interest. A more accurate determination of primary products may be made by determining the independent yields of isobaric nuclides as a function of atomic number.

In contrast to the case of bismuth[1] the fission instability of the heavy elements in the region of uranium is such that fission is energetically possible at low excitation energies without the necessity of the previous evaporation of large numbers of neutrons to give a more favorable Z^2/A parameter. However, at the high excitations obtainable with 340 Mev protons, it is possible for many nucleons to be evaporated prior to fission.[1] It was of interest therefore to determine the most probable fissioning nucleus under these conditions. From the work of O'Connor and Seaborg,[4] the yield versus mass curve might be expected to show a distinct symmetry about a single peak. This would suggest that symmetric binary fission is the most probable mode of fission. When the most probable fission is symmetric, the mass of the peak of the yield versus mass curve should be one-half the mass of the fissioning nucleus. When the Z/A ratio is constant as in the case of bismuth fission,[1] a homogeneous fluid model of the fissioning nucleus may be chosen. The ratio Z/A of the primary fission products may then be used to determine the atomic number of the fissioning nucleus.

The excited nuclei formed in the target by the bombarding beam do not necessarily undergo fission. In some cases the evaporation of nucleons may proceed to give spallation products which do not fission. An attempt was

made to identify several spallation products and make a rough estimate of their yields in an effort to determine what fraction of the total reactions proceeded by this path.

The cross section for the reaction $Al^{27}(p,3pn)Na^{24}$ with 340 to 350 Mev protons was measured by comparing it to the cross section for the reaction $C^{12}(p,pn)C^{11}$ which has been measured by Aamodt, Peterson, and Phillips.[11] By monitoring the bombarding beam with aluminum foils it becomes possible to measure the formation cross sections for several reference mass nuclides. Using these values, the yield versus mass curve could be transformed into a "formation cross section versus mass" curve. Integration under this curve makes possible the determination of the fission cross section for uranium with 340 Mev protons, assuming predominantly binary fission.

II. FISSION PRODUCT YIELDS

A. General Considerations

The quantity $IN\sigma$ (where N represents the number of atoms exposed to a bombarding beam of intensity I), hereinafter referred to as the independent rate of formation, R, (independent of the duration of the bombardment) is proportional to the cross section for formation of the radioactive species under consideration. Most of the earlier determinations in this work were made relative to the independent rate of formation of Ba^{140} and its precursors since no absolute formation cross sections had been measured at that time. The initial yield results are therefore expressed relative to the independent rate of formation of Ba^{140} and its precursors, defined as 1.00. The "relative yield" is defined to be the ratio of the cross section of formation of the nuclide in question to the cross section of formation for Ba^{140} and its precursors.

Chemical Techniques.-- The final sample for measuring the disinte-
gration activity of a radioactive nuclide does not in general consist of
100 percent of the amount of this species produced in the bombardment
(allowing for decay). To correct for the amount of active species lost
during the chemical separation procedure, it is customary to mix with
the active species a known amount (usually 1 to 10 mg) of the inactive
element, the carrier, whose isotopes are being considered. When due pre-
cautions have been taken to ensure equilibrium exchange, the percent
recovery of the active species is assumed to be identical with the percent
recovery of the carrier material.

In designing a chemical separation procedure it is necessary to
consider the estimated radioactive yield of the desired nuclide in re-
lation to the estimated yields of the most likely contaminants.

Detecting Instruments.-- Alpha emitting samples were measured either
in a proportional counter or in an ionization chamber apparatus. Alpha
energies were determined by means of an alpha pulse analyzer.[12]

Samples emitting beta and gamma radiation were counted by means of
end-window Geiger-Müller tubes. When this work was begun, the tubes
available were of the argon-alcohol type. For the more recent work,
end-window tubes filled with a mixture of argon and chlorine (Amperex,
Type 100-C) were used. These tubes show greater stability of counting
characteristics than the argon-alcohol tubes, and are capable of measur-
ing counting rates a factor of ten higher than the argon-alcohol tubes
could measure.

A crude magnetic beta ray spectrometer of low resolving power was
used to differentiate between positive and negative electrons, and to

measure maximum beta energies in some cases.

Backscattering.-- Correction factors for the scattering of beta parti-
cles into the counter from the backing material on which the sample is
mounted have been empirically determined by several workers.[13-15] The
amount of backscattering depends primarily upon the thickness and atomic
number of the backing material. The scattering is relatively insensitive
to beta energy in the range 0.6 to 3 Mev.[14,15] In most of our work,
the sample considered and the Ba^{140} monitor were mounted on similar
backings so that for relative yields the backscattering correction can-
celled out.

Self-Scattering and Self-Absorption Corrections.-- Since the radio-
active samples in many cases were of finite mass, it was necessary to
determine the loss of gain of apparent activity in passing through the
mass of the sample. There is a loss of apparent activity due to ab-
sorption in the material of the sample. Scattering of beta particles
and electrons by the material of the sample causes an apparent increase
in the activity. Some preliminary experiments were performed to deter-
mine empirically the combined self-absorption and self-scattering cor-
rections.[16,17] The absorbing power of sample material for particles
changes only slowly with the atomic number, but the scattering power
increases markedly. In some cases the scattering effects were so great
that a net increase in apparent counting rate was obtained even for
several milligrams per square centimeter of sample thickness.

Air Window Absorption.-- The loss of activity by absorption in the
air between the sample and the counter window, and in the window itself
was determined by straight line extrapolation of aluminum and/or

beryllium absorption curves whose initial portions were carefully measured

using thin absorbers.

Counting Efficiencies.-- In determining yields of various nuclides

it was necessary to make some assumptions regarding relative counting

efficiencies. For beta particles and electrons a counting efficiency

of 100 percent was assumed with the argon-alcohol filled tubes. The

calculated counting efficiencies used for electromagnetic radiation,

based on absorption in the gas of the counter tube, were 0.5 percent for

energies from 20 kev to 0.5 Mev, and 1 percent per Mev thereafter for

argon-alcohol filled tubes.[18] The ratio of the over-all efficiency of

the argon-chlorine (Amperex) tubes to the argon-alcohol tubes for the

same geometry was determined to be 45 percent by counting a number of

samples having beta and electromagnetic radiations of various energies.[19]

Effective Geometry.-- The effective geometry is related to the solid

angle subtended by the sensitive volume of the counter tube at various

distances from the sample. The sample holders used in the Geiger count-

ing had five positions varying from 0.3 to

6.7 cm from the tube window. An attempt was made to count the monitor

sample in the same position as the sample of the nuclide whose relative

yield was being determined. Where such an arrangement was not practical

initially, due to too great a difference in the counting rates of the

samples, the "geometry" ratio would be determined empirically when the

counting rates were more nearly equal. This ratio was not a true geo-

metric ratio because of increased air absorption at greater distances

from the counter window. For cases in which the absorption characteristics

of a particular beta spectrum were rather well known so that air absorption

corrections could be applied, measurements of the true geometric ratios between various positions were made.

B. Observed Fission Product Nuclides

The chemical procedures used to isolate the various elements are discussed in Appendix I.

Sodium.-- From a uranium target prepared with extreme care to eliminate any light element surface contamination, the isolated sodium fraction showed a beta activity of 15 hour half-life in amount larger than could be accounted for by impurities in the uranium. The decay was followed for four half-lives without observing any deviation due to radioactive impurities. Aluminum absorption measurements of the beta radiation showed the 1.4 Mev beta energy of Na^{24}.[*]

Manganese.-- In the manganese fraction separated after an eight hour bombardment of a uranium target, a 5.8 day activity was observed. The absorption characteristics and half-life corresponded to those observed for Mn^{52} formed by spallation of iron with 340 Mev protons.[20] The amount of Mn^{52} produced could be accounted for by the known amounts of iron and nickel impurities in the uranium target.

Iron.-- The iron decay measurements showed two major components. One was the expected 46 day Fe^{59}. The shorter component, having a half-life of about 8.4 hours, seems to be a new activity. Decay measured through 222 mg/cm^2 of aluminum (to block the particulate radiation of Fe^{59}) showed the 8.4 hour activity. No growth was observed (limit detectable, 20 minutes). This new activity is probably due to Fe^{60}.

Aluminum absorption measurements of the 46 day activity showed the soft beta radiation of Fe^{59}. Aluminum absorption measurements on the

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[*]Unless otherwise indicated, values of the half-life and beta energy for the various nuclides were obtained from either the "Table of Isotopes"[21] or the "Chart of the Nuclides."[22]

8.4 hour activity showed a beta particle of about 1.5 Mev energy. The sample was measured on a crude magnetic beta ray spectrometer. The 8.4 hour activity showed negative beta radiation. No positrons were observed.

Cobalt.-- Decay measurements of the cobalt fraction showed a component of 1.7 hours, presumably due to Co^{61}. Aluminum absorption measurements of the 1.7 hour activity showed a beta energy of about 1.3 Mev.

Nickel.-- The decay of nickel showed components of 2.6 hours and 56 hours, presumably Ni^{65} and Ni^{66}, respectively. Aluminum absorption measurements of the 56 hour Ni^{66} showed the 2.9 Mev beta particle of 4.3 minute Cu^{66} in equilibrium.

Copper.-- Three isotopes of copper were identified. The 3.4 hour Cu^{61} was observed by following the decay of its positrons on a crude beta ray spectrometer. The 12.9 hour Cu^{64} was also seen in this manner as well as by resolution of the decay curve obtained with a conventional Geiger counter. Sixty hour Cu^{67} was clearly seen in the beta decay curve.

Fermi-Kurie plots of the crude beta spectrometer positron data showed the 1.2 Mev β^{+} of Cu^{61} and the 0.66 β^{+} of Cu^{64}. Aluminum absorption measurements, taken after the 12.9 hour activity had decayed out, showed the 0.6 Mev β^{-} of Cu^{67}.

Zinc.-- Observation of the decay of the zinc samples showed a half-life of 49 hours. A growth corresponding to a daughter half-life of 14 hours was observed, indicating the presence of 49 hour Zn^{72} with 14 hour Ga^{72} daughter.

Arsenic.-- Decay of the arsenic samples observed through 350 mg/cm^{2} of aluminum (to block the particulate radiation of As^{77}) showed an

activity of 26.8 hours half-life. Decay without absorber showed the 40 hour activity of As^{77}.

Selenium.-- The predominant activity in the selenium samples was the 59 minute Se^{81m} with the 17 minute Se^{81} in equilibrium. It was possible to detect the 25 minute Se^{83} both by resolution of the decay curve and by observing the 2.4 hour Br^{83} daughter in the decay measurements. Twenty-four hours after bombardment, the decay curve showed essentially no activity other than the 2.4 hour Br^{83}.

Aluminum absorption measurements on the 59 minute activity showed the 1.5 Mev beta particle of Se^{81}.

Bromine.-- The bromine decay curves showed mainly 2.4 hour and 35 hour components when measured without absorber. Decay measured through 490 mg/cm^2 of aluminum showed components of 4.4 hour and of 34 minute half-life after subtraction of a 35 hour component due to the penetrating radiation of Br^{82}. Decay measured through 750 mg/cm^2 of aluminum showed a component of 34 minute half-life and a residium of 4.4 hour and 35 hour activities. The relative counting efficiency of the 4.4 hour activity was considerably reduced by the increased absorber. Aluminum absorption measurements of the 2.4 hour activity showed a 1 Mev beta particle.

From the evidence we conclude that Br^{80m}, Br^{82}, Br^{83}, and Br^{84} are formed as fission products with Br^{83} being produced in highest yield.

Rubidium.-- The rubidium sample showed an activity of 19.5 day half-life, presumably Rb^{86}. The sample was not isolated soon enough to detect any of the shorter lived rubidium isotopes. Aluminum absorption measurements on the rubidium activity showed a 1.8 Mev beta particle.

Strontium.-- From decay measurements a 9.7 hour strontium activity
was observed. A 60 day yttrium activity was chemically separated from
the strontium sample, indicating the presence of Sr^{91}. Another strontium
sample, from which yttrium had been removed a week after bombardment of
the target, showed an activity of 54 day half-life. This was thought
to be Sr^{89}.

Aluminum absorption measurements on the 54 day activity showed
the characteristic 1.5 Mev beta particle of Sr^{89}. The background due
to electromagnetic radiation was negligible.

Zirconium.-- The zirconium decay showed a 17 hour activity which
grew a 69 minute daughter, presumably due to Zr^{97} and Nb^{97}. After
decay of the Zr^{97}, a decay line of predominantly 65 day half-life (Zr^{95})
was seen.

Niobium.-- Four major species were identified in the niobium decay:
68 minute Nb^{97}; an activity of approximately 23 hours, probably Nb^{96};
90 hour Nb^{95m} obtained by analytical treatment of the counting data;
and 37 day Nb^{95}.

Molybdenum.-- The only activity observed in the molybdenum fraction
was the 67 hour Mo^{99}.

Ruthenium.-- Decay measurement showed a 4.4 hour activity and
longer components. After allowing one year for intermediate activities
to die out, decay for a period of one year showed a one year activity.
When the activity due to this component was subtracted from the earlier
data, an activity of approximately 43 day half-life was observed. Sub-
traction of this in turn showed a 37 hour activity, indicating the
presence of 4.4 hour Ru^{105} decaying to 37 hour Rh^{105}. The 43 day activity

was presumed to be Ru^{103}.

Aluminum absorption measurements on the one year activity showed the characteristic hard beta particles of the Rh^{106} daughter in equilibrium with 1.0 year Ru^{106}.

Palladium.-- Two palladium activities were observed directly, 21 hour Pd^{112} and 14 hour Pd^{109}. The formation of 26 minute Pd^{111} was demonstrated by the separation of 7.5 day Ag^{111} from a portion of the target immediately after bombardment and from a second portion after an interval of three to four hours. The specific activity of Ag^{111} was significantly higher in the second portion than in the first, indicating its formation by the decay of Pd^{111}.

Decay was measured without absorber, through 80 mg/cm^2 of aluminum (to block the beta particles of Pd^{112}), and through 490 mg/cm^2 of aluminum (blocking the beta particles of Pd^{112} and Pd^{109}, but allowing the radiation of 3.2 hour Ag^{112} in equilibrium with Pd^{112} to be counted). Growth of the 3.2 hour Ag^{112} was observed.

Silver.-- Decay through 1380 mg/cm^2 aluminum (to block all particulate radiation from silver isotopes except that of Ag^{112}) showed the 3.2 hour Ag^{112}. Decay without absorber on a sample separated very soon after bombardment (to minimize growth of 3.2 hour Ag^{112} from 21 hour Pd^{112}) emphasized the 5.3 hour Ag^{113}. The 7.5 day Ag^{111} was clearly seen, and shown to be formed (see above) as a primary fission product, its independent yield being 18-20 percent of the total observed β^- chain. Decay followed over a period of ten months showed an activity of 250-270 days, presumably Ag^{110}.

The formation of 20 minute Ag^{115} was shown by separating 53 hour Cd^{115} from a portion of the target immediately after bombardment and from a second portion after an interval of about four hours. The specific activity of Cd^{115} separated from the second portion was significantly higher than that from the first portion, indicating its formation by decay of Ag^{115}.

A sample of silver was initially separated from cadmium, then allowed to decay. Cadmium separated from this sample after four hours showed a 53 hour half-life with no indication of a 43 day component. This indicates that Ag^{115} decays predominantly to the Cd^{115} ground state rather than to Cd^{115m}.

Aluminum absorption measurements on the 3.2 hour activity showed the characteristic 3.6 Mev beta particle of Ag^{112}. Measurements on the 7.5 day activity showed the 1 Mev beta energy of Ag^{111}. The background due to electromagnetic radiation was less than 0.05 percent of the initial activity. Correcting for the difference in counting efficiencies, this means that any 8.6 day Ag^{106} must be present in an amount less than 5 percent of that of Ag^{111}.

Cadmium.-- Decay through 946 mg/cm^2 of beryllium (to block all particles of energy less than 1.9 Mev) showed evidence of the 49 minute Cd^{111m} and indicated the possible presence of 6.7 hour Cd^{107}. Decay of the beta radiation showed 2.8 hour Cd^{117}, 53 hour Cd^{115}, and 43 day Cd^{115m}. There was a very long lived component which suggested the presence of 470 day Cd^{109}. Aluminum absorption measurements showed the 1.1 Mev beta particle of the 53 hour Cd^{115}.

Tin.-- Decay measurements showed components of about 27 hour and
of 9.5 day half-life and a longer component, presumably of 130 day
half-life. These seem to be Sn^{121}, Sn^{125}, and Sn^{123}, respectively.
The other isomer of each mass number was not observed.

Tellurium.-- Of the many components in the tellurium decay curve,
it was possible to resolve out only the 30 hour Te^{131m} and its 8 day
I^{131} granddaughter.

Cesium.-- The predominant activity in the cesium fraction was the
13 day Cs^{136}. Aluminum absorption measurements of this activity showed
the characteristic 0.3 Mev beta energy of Cs^{136}. Comparison of the
activity observed through aluminum absorbers and through beryllium
absorbers indicated soft electromagnetic radiation (of the order of
K x-rays in this region) present to the extent of about 15 percent of
the particulate radiation. This indicates relatively good yields of
neutron deficient cesium isotopes decaying by orbital electron capture.

Barium.-- Most of the bombardments were interrelated by measuring
the amount of 12.8 day Ba^{140} produced, and using this as an internal
monitor. In most cases the barium was not separated from radium. In
these cases, the sample was counted through enough aluminum absorber
to prevent any alpha particles emitted by radium or its daughters from
reaching the counter tube. In addition to observing its half-life,
Ba^{140} was identified by observing the growth of its 40 hour La^{140} daughter.

A sample of the radioactive barium was analyzed in the mass spectro-
graph.* The emulsion side of the collector plate was placed against the
emulsion of a second plate and the radioactive species allowed to decay .

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*Mass spectrographic analysis by F. L. Reynolds of this laboratory.

for a period of about two weeks. Lines on the second or "transfer" plate
indicated radioactive barium isotopes of masses 131, 133, 135, and 140
in comparable yields. An approximately 10 day activity was observed in
a cesium sample separated from a purified barium sample some time after
bombardment.

Although Ba^{131} has approximately the same half-life and may be present
in yields of the same order of magnitude as Ba^{140}, the activity of
$Ba^{131}-Cs^{131}$ detected by the Geiger tube is less than 1 percent of the
corresponding activity of $Ba^{140}-La^{140}$ because of the large difference
in counting efficiencies.

Components of 29 to 39 hours having soft particle radiation were
observed in the decay curves. These probably represent a mixture of
Ba^{133m} and Ba^{135m}. An 85 minute component, presumably Ba^{139}, was also
observed. In a sample of barium separated from radium on an ion-exchange
column, the 85 minute, 29 to 39 hour, and 12.8 day components were ob-
served. The positrons of 2 hour Ba^{129} were not observed on a crude beta
ray spectrometer measurement of the column separated barium.

Lanthanum.-- Decay curves showed essentially a single component,
the 40 hour La^{140}, over a period of 8-9 half-lives. Decay of the
radiation which passed through 946 mg/cm^2 of beryllium (to block the
particles and give other possible electromagnetic radiation a more
favorable counting efficiency relative to La^{140}) gave a curve essentially
parallel to the curve taken without absorber. Since the final separation
of lanthanum was not accomplished until 56 hours after bombardment, the
possibility of formation of nuclides of shorter half-life in comparable
yield is not excluded.

Aluminum absorption curves of the 40 hour activity compared closely

with absorption curves of the radiations from the 40 hour β^- daughter

of Ba^{140}. From the absorption curves, the counting rate was corrected

for air and window absorption, and for the contribution of the electro-

magnetic radiation. The disintegration rate was calculated from this

corrected counting rate.

Calculations based on the relative yields of Ba^{140}, La^{140}, and Nd^{140}

(explained in Section III) show that approximately 30 percent of the

total mass number 140 yield is formed as La^{140}.

Cerium.-- Decay curves showed components of approximately 35 hours

and approximately 30 days with a component of intermediate half-life.

Analysis of the data indicated Ce^{143} with Pr^{143} daughter, and Ce^{141}.

Decay over a period of about six months indicated the presence of 275 day

Ce^{144}.

Comparison of the number of x-rays emitted by the long lived activity

with the number from Ce^{144} tracer obtained from slow neutron fission of

uranium indicated the presence of 140 day Ce^{139} in our sample. Aluminum

absorption measurements on the long lived component showed two beta com-

ponents which could be attributed to Ce^{144} and the 17 minute Pr^{144} in

equilibrium.

Praseodymium.-- Observation of the decay through 406 mg/cm^2 of

aluminum showed a 19 hour half-life. Observation of the decay without

absorber showed two components, one of 13.5 day half-life, the second

(by subtraction) of half-life 16 to 19 hours. Aluminum absorption

measurements on the 19 hour activity showed hard beta radiation of 2.3 Mev

energy and indicated a soft component of about 0.6 Mev energy in about

20 percent abundance. After decay of the 19 hour activity, aluminum

absorption measurements of the 13.5 day activity showed a particle of

0.92 Mev maximum energy. On the basis of the decay and absorption measure-

ments, it was concluded that Pr^{142} and Pr^{143} were both present in good

yield.

Pr^{142} is a shielded nuclide; consequently, its independent yield was

obtained directly. The independent yield of Pr^{143} was obtained by removal

of the 33 hour Ce^{143} parent from the rare earth group within two hours

after bombardment. From the yield curve (Figure 1) it is expected that

the total yield of products of mass number 143 will be less than the

total yield of products of mass number 142. Since in the case of these

two isotopes the ratio of the yield of Pr^{143} to that of Pr^{142} was about

1.7, the Pr^{143} represents a greater fraction of the total mass number

143 yield than the Pr^{142} does of its total mass number yield. This indi-

cates that whereas the most probable primary charge for a given mass is

on the neutron excess side for isotopes of the lighter elements, the

trend is for the most probable primary product to be much closer to

stability for the higher mass chains.

Neodymium.-- Observation of the decay through 407 mg/cm^2 of beryllium

(sufficient to block the beta particles of Nd^{147}) showed two components.

After subtraction of the 11 day tail due to the gamma ray of Nd^{147}, a

3.3 day decay was found. This is due to the activity assigned to Nd^{140}

by Wilkinson and Hicks.[23] Decay measurements without absorber showed

the 11 day half-life of Nd^{147} for over three half-lives. Aluminum ab-

sorption curves showed the characteristic beta particle energy of Nd^{147}.

Since less than 2 percent of the initial total activity could be attributed

to Nd^{140}, this activity did not interfere and was not characterized by absorption. Its counting efficiency was calculated from the data of Wilkinson and Hicks.[23]

Calculations based on the relative yields of Ba^{140}, La^{140}, and Nd^{140} show that approximately 7.5 percent of the total mass 140 yield is formed as Nd^{140}.

Promethium.-- Decay was observed both without absorber and through 485 mg/cm^2 of aluminum (sufficient to block the beta particles of Pm^{149}). The decay without absorber showed components of approximately 13 hours, of 47 hours, and of about 43 days. Decay of the radiations transmitted through the absorber showed components of 47 hours (the gamma ray of Pm^{149}), 5.3 days, and 43 days. Of the activities, only the 47 hour Pm^{149} and the 5.3 day Pm^{148} have been assigned. A 12.5 hour promethium activity has previously been reported, but no mass assignment was given. The 43 day activity represents a previously unreported nuclide.

Absorption of the radiations of the 47 hour activity showed a single beta component of about 1.1 Mev energy. This is the reported maximum energy for the beta particles of Pm^{149}. After decay of the 47 hour and 5.3 day components, absorption measurements of the 43 day activity were made. Aluminum absorption showed two beta components of energies about 0.5 Mev and about 2.3 Mev. The hard component was present to only 5 to 10 percent abundance. The sample was also measured on a crude beta ray spectrometer which showed (from a Fermi-Kurie plot of the data) β^- components of 0.6 and 2.4 Mev. The electromagnetic background of the aluminum absorption curve was of the order of 3 percent. After blocking the particulate radiation with beryllium absorber, the electromagnetic

component was absorbed in copper. No K x-rays of neodymium (which have a half-thickness of 165 mg/cm^2 in copper) were observed. Absorption of the electromagnetic radiation in lead showed a half-thickness of 7.5 g/cm^2 which corresponds to a gamma ray of approximately 0.9 Mev energy.

The yield of the shielded Pm^{148} is only about 1 percent of that for the Pm^{149} which represents the total β^- chain for its mass. The yield of the 43 day promethium is about 10 percent of that for Pm^{149}. On the basis of yield and the fact that this activity was not reported formed in slow neutron fission or by Nd^{150}(n,γ) or (p,n) reaction, a tentative assignment of the 43 day activity to Pm^{146} is made.

Samarium.-- The samarium decay showed components of about 10 hours and of 47 hours, the 47 hour component being quite prominent. After decay of this activity, a 15-16 day line was observed. Aluminum absorption measurements of the 47 hour activity indicated a beta particle of about 0.8 Mev energy. In the absorption measurements the 2.4 Mev β^- of Eu^{156} was also observed.

The principal activities found in the samarium fraction were thus the 47 hour Sm^{153} and the 10 hour Sm^{156} with its 15.4 day Eu^{156} daughter. The yield of Sm^{156} relative to Sm^{153} was only 27 percent. The yield of Sm^{153} represents the entire β^- chain for mass 153 whereas Eu^{156} is expected to be a large contributor to the mass 156 yield.

Europium.-- The principal europium activity seen was the 15.4 day Eu^{156}. Aluminum absorption showed the characteristic 2.4 Mev β^- of Eu^{156}.

Terbium.-- The terbium decay curve resolved roughly into two components, the longer of which had a half-life of 74 days. The shorter component was a mixture of activities with half-lives between 5 and 7 days

and a shorter one. Decay was observed without absorber and also through
183 mg/cm^2 of beryllium (sufficient to block the 0.5 Mev β^- particles of
Tb^{161}). After subtraction of the 74 day residium, the decay through
beryllium showed strong indication of a 5.1 day activity. The decay
observed without absorber showed a mixed activity with a 6.8 day activity
predominating after subtraction of the 74 day component. These observations
indicate the formation of Tb^{160} and Tb^{161}, and the probable formation of
Tb^{153} and Tb^{156}.

In the terbium fractions separated 12 to 14 hours after bombardment,
traces of alpha activity were observed. A sample submitted to Mr. J. O.
Rasmussen for alpha pulse analysis showed alpha energy and decay corre-
sponding to 4.5 hour Tb^{149}.

Dysprosium.-- The chief component observed in the dysprosium decay
was the 80 hour Dy^{166}. A long lived activity was also observed, but it
was not possible to determine its half-life. Aluminum absorption measure-
ments on the equilibrium mixture showed the 0.4 Mev beta particle of
Dy^{166} and the 1.9 Mev beta particle of the Ho^{166} daughter.

Calculations based on the relative yields of Dy^{166}, Ho^{166}, and Yb^{166}
(see Section III) indicate that 2.54 percent of the total mass 166 yield
is formed as Dy^{166}.

Holmium.-- The major component observed in the holmium decay was the
27 hour Ho^{166}. The decay was followed through eight half-lives. The
activity remaining as long lived component was less than 0.02 percent
of the Ho^{166} activity present at end of bombardment. Aluminum absorption
measurements showed the approximately 1.9 Mev beta energy of Ho^{166}.
The 7 day activity presently assigned to Ho^{163} was not observed in our

holmium sample in a resolvable amount. This may indicate that the 7 day

holmium is shielded, and hence should be assigned to Ho^{162}.

Calculations based on the relative yields of Dy^{166}, Ho^{166}, and Yb^{166}

indicate that 4.7 percent of the total mass 166 yield is formed as Ho^{166}.

Erbium.-- The erbium sample first showed growth, which was identified

as a holmium activity from the plot of the radioassay data (see Appendix I,

Recovery and Chemical Yield of Rare Earths). The samples of column elutriant

containing erbium carrier showed a growth of activity while those samples

taken after elution of the erbium but before elution of the holmium carrier

showed rapid decay. This indicated a new erbium activity decaying by

orbital electron capture and/or positron emission to a short lived holmium

activity. Decay was observed without absorber and also through 99.7 mg/cm^2

of beryllium (to block the beta particles of Er^{169}). Two new activities

were observed in the decay of activity transmitted through absorber; one

of about 17 hour half-life and the other of 65 hour half-life. The decay

without absorber showed a mixture of these activities and a component

of 9.4 day, the Er^{169}.

The growth observed corresponded to a 4.5 hour half-life for the

daughter activity. The holmium activity[24] of this half-life is assigned

to Ho^{161}. The new erbium activities are probably $Er^{160,161}$ and Er^{163}.

The data are not sufficient to match the activities to definite mass

numbers.

Thulium.-- Decay measurements without absorber showed an approximately

10 day activity prominently. Observation of the decay through approxi-

mately 40 mg/cm^2 of aluminum and through approximately 60 mg/cm^2 of

beryllium emphasized an activity of two to three days half-life. These

absorbers blocked the conversion electrons of 9.6 day Tm^{167}. The 9.6 day

activity was masked to such an extent by the absorbers that it was not

resolvable graphically from the shorter activity. Since an activity on

the neutron deficient side of stability in this region would be expected

to have about the same counting efficiency as the 9.6 day Tm^{167}, it is

felt that the two to three day activity represents a negative beta tran-

sition, possible Tm^{172}.

Ytterbium.-- The ytterbium decay curve showed components of 32 days

and 62 hours. The sample showed growth corresponding to a 7.7 hour

daughter activity. The samples of column elutriant containing ytterbium

carrier showed this growth of activity while those samples taken after

elution of the ytterbium but before elution of the main body of thulium

showed a decay of about eight hours half-life. The new ytterbium activity

is thus shown to decay to a thulium activity. From the observed growth

and an estimate of the time of separation from thulium, the counting

efficiency of the ytterbium parent was found to be small compared to

the thulium daughter. On the basis of the assignment of the 7.7 hour

thulium activity[25] to Tm^{166}, the new 62 hour ytterbium activity is

assigned to Yb^{166}.

Aluminum absorption measurements on the 62 hour activity showed

the presence of conversion electrons, a hard beta particle, and electro-

magnetic radiation background. The relative abundance of electrons and

hard beta particles (2 Mev) compared favorably with those reported for

the particles[25] from Tm^{166}. Beryllium and copper-beryllium absorption

measurements of the 32 day activity showed electrons, x-rays, and gamma

rays comparable to some of those reported for 32.5 day Yb^{169}. Ninety-nine

hour Yb^{175} was not detected either by decay or absorption measurements.

Isobaric chain yield calculations indicated that 13.5 percent of the total yield of mass 166 was formed as Yb^{166}. The most probable primary charge for the nuclide of this mass is $Z = 71$.

C. Tabulation of Observed Yields

The yields of selected fission products measured relative to the yield of Ba^{140} (and its precursors) are given in Table I below. By definition, the relative yield, Y_R, of Ba^{140} (including its precursors) is 1.00.

Formation cross sections were measured for a number of nuclides including their precursors (i.e., total β^- chains). By means of these, the relative yield values ($Y_R = 1.00$ corresponds to $\sigma = 23.5$ mb) given in Table I below were converted to formation cross section values. Integration of the yield versus mass curve (Figure 1) in terms of the formation cross sections indicates a total fission cross section of 2.0 barns for uranium with 340 to 350 Mev protons.

Table I

Relative Yield of Fission Products ($Ba^{140} = 1.00$)

Mass Number	Nuclide Measured	Class*	Y_R
24	Na^{24}	2	0.0021
59	Fe^{59}	2	0.0078
65	Ni^{65}	2	0.024
66	Ni^{66}	2	0.027
67	Cu^{67}	2	0.091
72	Zn^{72}	2	0.090
76	As^{76}	1	0.0091
80	Br^{80m}	1	0.015
81	Se^{81m}	3	0.34
82	Br^{82}	1	0.067
83	Se^{83}	3	0.22
	Br^{83}	3	0.16
84	Br^{84}	3	0.14
86	Rb^{86}	1	0.59
89	Sr^{89}	3	1.4
91	Sr^{91}	3	1.6
92	Sr^{92}	3	1.7
103	Ru^{103}	3	1.8
106	Ru^{106}	3	2.2
109	Pd^{109}	3	0.19
111	Pd^{111}	3	1.76
	Ag^{111}	3	0.44
112	Pd^{112}	3	0.22
115	Cd^{115}	3	1.46
	Cd^{115m}	3	0.51
131	Te^{131m}	3	0.25
136	Cs^{136}	1	0.25
139	Ba^{139}	3	1.8
140	Ba^{140}	3	1.00
	La^{140}	1	0.48
	Nd^{140}	3	0.18
142	Pr^{142}	1	0.33
143	Pr^{143}	1	0.55
147	Nd^{147}	3	1.4
153	Sm^{153}	3	0.19
156	Sm^{156}	3	0.05
	Eu^{156}	3	0.12
166	Dy^{166}	3	0.0019
	Ho^{166}	1	0.0020
	Yb^{166}	3	0.030
193	Os^{193}	3	0.00043
~198	$Au^{198,199}$	3	0.00094
224	Ra^{224}	3	0.12
236	(p,3n) reaction**		0.51

*Class 1 Independent yield or shielded nuclide
 Class 2 Approximately total yield of mass number
 Class 3 Partial yield of mass number
**W. W. Meinke, G. C. Wick, and G. T. Seaborg, University
of California Radiation Laboratory Report UCRL-868 (Sept. 26, 1950).

III. ISOBARIC YIELD STUDIES

A. Introduction

As mentioned in the introduction (cf., page 4) identification of
the charge of the fissioning nucleus depends upon the determination of
the primary fission products. An experimental attempt has been made
here to measure the independent yields of various members of each of
several reference mass chains. The nuclide whose yield corresponds to
the maximum of the curve obtained by plotting the independent yield as
a function of atomic number for a given mass number is defined to be
the primary product for that mass number. In practice it is usually
quite difficult experimentally to measure directly the independent yields
for all of the members of a given mass chain. Those independent yields
which have been measured experimentally are listed in Table II below.
The yields of the measured members furthest from stability for any
chain include the yields of their precursors, except in the case of a
shielded nuclide.

B. Experimental Techniques

Of the several techniques used in measuring independent yields,
the choice depends upon the half-lives and the relative difficulty of
the separation chemistry for the species sought. For example, consider
the chain of mass 111:

$$Pd^{111} \xrightarrow[26 \text{ min}]{\beta^-} Ag^{111} \xrightarrow[7.5 \text{ day}]{\beta^-} Cd^{111} \ .$$
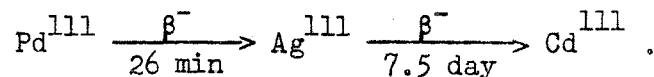
The uranyl nitrate target was bombarded for 2.0 minutes, fused, poured
into a dissolver solution containing a known quantity of silver carrier

and some palladium "holdback" agent, and the solution divided into two
portions. From one portion, silver chloride was immediately precipi-
tated to separate from palladium.

Table II

Independent and Isobaric Yield Data

Mass Number	Nuclide Measured	Relative Yield	Estimated Fraction of Total Chain
76	As^{76}	0.009	0.05
83	Se^{83}	0.22	0.6
86	Rb^{86}	0.59	0.6
111	Pd^{111}	1.76	0.5
	Ag^{111}	0.44	0.12
136	Cs^{136}	0.25	0.12
140	Ba^{140}	1.00	0.43
	La^{140}	0.48	0.21
	Nd^{140}	0.18	0.075
166	Dy^{166}	0.0019	0.044
	Ho^{166}	0.0020	0.047
	Yb^{166}	0.030	0.72

This separation was accomplished within three minutes after the end
of the bombardment. The other portion was allowed to stand until the
26 minute Pd^{111} had all decayed to Ag^{111}. The silver fractions were
then purified separately. From the initial specific activity of the
silver obtained in the two cases and the known half-lives of the two
species, it was possible to calculate from the integrated decay equations
the independent yield of Ag^{111} and the yield of Pd^{111} plus its precursors.
An example of another type of treatment is afforded by the chain of
mass 140:

$$Ba^{140} \xrightarrow[\text{12.8 day}]{\beta^-} La^{140} \xrightarrow[\text{40 hour}]{\beta^-} Ce^{140} .$$
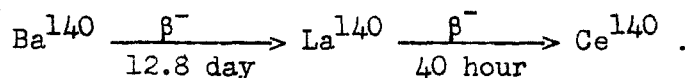
Since the activities involved are relatively long, a one hour bombardment

could be used. Separation of the two species was accomplished within

one to two hours after bombardment. After purification of the two frac-

tions and counting of the radioactive samples, the observed yield of

Ba^{140} was used to correct the La^{140} activity for that portion which had

grown from barium during bombardment and before separation of the species.

In this way the independent yield of La^{140} was obtained.

In cases where the independent yield of daughter activity is small

relative to the parent, precautions must be taken to minimize the amount

of contribution from the parent in the initially separated sample of

daughter activity. This may be done by reducing the bombardment time,

minimizing the time elapsed from end of bombardment to initial separation

of the desired activity, and developing a specific separation procedure

which is rapid and essentially complete in one operation.

Independent yields of "shielded" nuclides may be measured directly

since the activity could not grow from the neighboring isobars which

are stable.

C. Interpretation of the Data

In order to interpret the data obtained for independent yields,

the assumption is made that the independent yields of the various isobars

of a given decay chain are distributed symmetrically about some particu-

lar Z in a manner which can be well approximated by the normal curve

of error,* using suitable parameters to adjust the half-width and peak

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*A similar assumption was used by Glendenin, Coryell, and Edwards[26]
in the development of their hypothesis of Equal Charge Displacement for
primary fission fragments from slow neutron fission of U^{235} and Pu^{239}.
However, the approximation proposed above is not dependent upon the Equal
Charge Displacement hypothesis (which, from the experimental data, does
not appear to hold for the type of fission discussed in this paper).

height of such a distribution curve to the experimental results obtained.
A description of the mathematical method of fitting the normal curve of
error to such data is given in Appendix II.

It will be seen from Appendix II that three experimental data are
necessary and sufficient to establish the constants of the curve. We
can therefore obtain some idea of the validity of the basic assumption
by studying the independent yields in chains containing more than three
members. Potentially suitable chains are those of masses 166, 141, 111,
and 72, each chain containing four measurable members.

Initial sets of three experimental data have been obtained for
the chains of mass 140 and mass 166. The decay schemes of these chains
are shown in Figure 2. Figures 3 and 4 show the standard-curve-of-error
histograms for the mass 140 and mass 166 chains, respectively. The
parameters for these curves are tabulated in Table III.

Table III

Parameters for Standard Curve of Error for Isobaric Yield Data

	Z_c	Standard Deviation	Z_c/A
mass 140	57	1.9	0.41
mass 166	71	2.7	0.43

It is apparent that if one could predict the parameters of these
charge-distribution histograms as functions of the chain mass number,
it would be possible to derive correction factors for each mass number
to give the total chain yield, including stable nuclides, in terms of

the yield of any nuclide of the chain. The rough estimates of total chain

yields given in Table II were made on the basis of the few data presently

available.

IV. SPALLATION PRODUCTS

A. Introduction

The work of O'Connor and Seaborg[4] on the bombardment of uranium

with 380 Mev helium ions showed that there was apparently a continuous

yield of radioactive products for the entire range of elements from the

uranium region down to the vicinity of atomic number 25. Above mass 120

the yield of fission products decreased with increasing mass number.

In the region above mass 200 the yield curve was observed to increase

with increasing mass. This increase was attributed to products formed

by high energy spallation of uranium. The yield of products found in

the region of the minimum of the curve might be attributed to a combi-

nation of fission and spallation reactions. That fission can occur

in such a manner as to give products of such high mass was shown by

the observation of complementary product nuclei of low mass.

In the present work it was observed that the primary fission

products tend toward the neutron deficient side of stability with in-

creasing mass number as was found for bismuth fission.[1] However, in

the region of mass 180 to 200, several β^- emitting nuclides have been

observed. These are attributed tentatively to spallation reactions.

B. Results

Hafnium.-- The purified hafnium fraction showed a negative beta

activity having an energy of about 0.4 Mev and long life. This activity

has been attributed to Hf^{181}.

Osmium.-- Decay of the osmium fraction showed a component of about 32 hours half-life which was attributed to Os^{193}.

Gold.-- The decay of the gold sample gave indication of the 14 hour and 5.6 day isomers of Au^{196}. An intermediate activity was observed which seemed to be a mixture of Au^{198} and Au^{199} (2.7 day and 3.3 day, respectively). Aluminum absorption measurement on this activity showed a beta energy of about 1 Mev and a softer component whose energy was not determined.

Radium.-- Decay of the radium samples was followed by both Geiger counter measurements and alpha particle counting. From the decay curves and pulse analysis of the alpha activity, radium isotopes of mass 223, 224, and 225 were identified. A new Geiger counter activity, having a half-life of the order one to two hours, was observed in the separated radium fraction. This activity might be from Ra^{227}, Ra^{230}, or a mixture of both.

Actinium.-- The formation of Ac^{226} was indicated by the presence of a short-lived thorium alpha activity eluted from an ion exchange resin column some 20 to 30 hours after bombardment. The activity was identified as thorium by its elution characteristics.

V. DISCUSSION

From examination of the data herein presented, a few marked trends are observed. Almost all of the low mass nuclides identified were of the neutron excess type. As higher mass numbers were approached, neutron deficient nuclides increased in relative abundance of formation.

However, in the spallation region, which merges with the region of heavy

fission fragments in the vicinity of mass number 180, neutron-excess

nuclides again predominate.

Let us consider a nuclide formed by high energy spallation of a

U^{238} nucleus, this nuclide having an amount of internal energy much

in excess of the fission threshold. This energy may be dissipated in

one of three different ways: it may in part be used to cause fission,

any excess being carried off by the fission fragments, by particle

emission, or by radiation; it may be dissipated as radiation, the nuclide

in question descending to a nonfissioning ground state; or it may be

carried off, in part or entirely, by an emitted nucleon or composite

particle, such as an alpha particle or a deuteron, leading to a dif-

ferent nuclide which again may undergo one of these three changes. The

mechanism involved probably compares more closely to the case for "fast

fission" of bismuth[1] than to the so-called "slow fission" of U^{235} induced

by thermal neutrons.

If we represent all the possible nuclides as points on a plane

with coordinate axes Z and A-Z, then the various spallation and fission

products possible from U^{238} with protons will all fall within that

portion of the first quadrant of this plane having $Z \leq 93$, $A-Z \leq 146$.

Normalizing the total probability of fission to unity, the various

partial fission probabilities of the nuclei lying within the fissioning

region can be described by a contour map. The contour lines are the

loci of points of equal partial fission probability. The fission

product yield curve may be plotted in a similar manner on the same

chart (see Figure 5).* The dotted lines of slope -1 represent isobaric
chains. The independent yield of the members of these chains as a
function of Z is given by the value of the contour line intersected for
each Z.

The contour representation of fission product yields offers a
criterion for establishing the charge to mass ratio of the primary
fission products. If the charge to mass ratio is constant, as in the
case for bismuth fission,[1] the curve connecting the points of maximum
relative yield for a given mass number (i.e., the so-called "ridge line")
will be a straight line. Although the present data are not conclusive,
there is a definite suggestion (cf. Figure 5) that the "ridge line"
is not straight, i.e., Z/A is not constant for the fission products.
(Analysis of the isobaric charge distribution curves, cf. Section III,
indicates the same result.) If this phenomenon is real, one would
.expect a distribution of fissioning nuclei rather than a single dominant
fissioning species.

We may assume that the shape of the fissioning-nucleus contour map
is markedly energy-sensitive. Let us further assume that the fission-
fragment yield distribution is uniquely determined by the fissioning-
nucleus distribution. Some qualitative predictions of the behavior of
the fission-fragment yield curve with variation of bombarding energy
may then be made.

The most obvious effect of increasing the bombardment energy will
be to increase the number of intermediate spallation products leading

*In the figure, the bombardment product fission probability contours
(upper right corner of Figure 5) are hypothetical; the fission product
yield contours are expressed relative to Ba^{140} (including its precursors) = 1.0.

to fission, since Douthett[8] has shown that excess bombarding energy does

not appear as kinetic energy of the fragments. This will lead to a

lower average mass for the "fissioning nucleus," with its concurrent

effect of lowering the mass of the fission fragments which have the

maximum yield. Also, should neutrons be evaporated from the excited

nuclides in preference to protons or other charged particles, as seems

likely, the charge-to-mass ratio of the average fissioning nucleus, and

hence of its fragments, should be increased. It is thus expected that

at high energies the nuclides closer to stability, or even on the

neutron deficient side, should appear in greater relative abundance,

as is actually observed.

The formation of Cu^{61} (neutron deficient) and Cu^{64} (shielded) as

fission products with yield of the order of magnitude of Cu^{67}, in the

region where the formation of neutron excess nuclides is expected to

predominate, suggests that there is an effective mode of fission dif-

fering from the simple symmetric binary fission discussed above. If a

large number of neutrons were evaporated from the excited target nucleus,

an energetically favorable ternary fission mechanism could lead to the

observed copper isotopes. The high yield of Na^{24} relative to the

extrapolated smooth curve (Figure 1) may also point in this direction.

VI. ACKNOWLEDGMENTS

APPENDIX I

General Chemical Procedures[*]

Sodium.-- The uranium target was dissolved in nitric acid containing
a known amount of sodium ion as carrier. The uranium was extracted into
a solution of tributyl phosphate in benzene. After boiling the aqueous
phase to reduce its volume, strontium and barium nitrates (carrying
radium) were precipitated from fuming nitric acid. The excess nitric
acid was removed and silver chloride and manganese dioxide precipitated.
The supernatant was boiled to dryness with hydrochloric acid. The
residue was taken up in water and barium sulfate precipitated. From
the supernatant, sodium zinc uranyl acetate was precipitated. This
precipitate was dissolved in 6 $\underline{N}$ hydrochloric acid and sodium chloride
precipitated from isopropyl alcohol saturated with hydrogen chloride
gas. The precipitate was dissolved in 0.2 $\underline{N}$ hydrochloric acid and the
members of the acid sulfide group precipitated by passing in hydrogen
sulfide gas. The solution was made alkaline with ammonium hydroxide
and hydroxides and sulfides precipitated. The supernatant was evapo-
rated to near dryness and the residue taken up in 0.2 $\underline{N}$ hydrochloric
acid. Sodium zinc uranyl acetate was precipitated and then sodium
chloride as above. This precipitate was taken up in perchloric acid,
potassium, rubidium, cesium and ruthenium carriers added, and the
solution fumed to small volume. The solution was chilled and isopropyl
alcohol added. After centrifuging and discarding the insoluble per-
chlorates, the supernatant was saturated with hydrogen chloride gas

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[*] Many of these procedures are modifications of procedures developed
on the Plutonium Project[27] and by the chemistry group of the Radiation
Laboratory at Berkeley.[28]

while kept at ice temperature to precipitate sodium chloride. The
sodium chloride was reprecipitated, washed, dried at $110^{\circ}C$, and sodium
determined by weighing as NaCl.

Manganese.-- From a nitric acid solution of the uranium target
containing manganese carrier, manganese dioxide was precipitated by
the addition of potassium bromate. The precipitate was dissolved in
dilute nitric acid containing hydrogen peroxide. Concentrated nitric
acid was added and the manganese oxidized to permanganate by the
addition of sodium bismuthate. After centrifuging to bring down the
excess bismuthate (insoluble), the supernatant was decanted and the
permanganate reduced by the addition of oxalic acid. Silver chloride
was precipitated to remove any acid insoluble compounds not brought
down on the excess sodium bismuthate. The supernatant was diluted
and members of the acid sulfide group were precipitated by the addition
of hydrogen sulfide gas. The supernatant was made basic and manganous
sulfide precipitated. The sulfide was dissolved in nitric acid and
the foregoing procedure repeated for additional purification. After
dissolving the sulfide from this second cycle, manganese dioxide was
precipitated and the manganese determined by weighing as Mn_3O_4 after
ignition.

Iron.-- Iron carrier in an aqua regia solution of the target
(containing the minimum amount of nitric acid) was boiled with concen-
trated hydrochloric acid to remove most of the nitrate. The material
was taken up in 9 $\underline{N}$ hydrochloric acid and the iron extracted into
di-isopropyl ether. After thoroughly washing the ether layer with

7.5 $\underline{N}$ hydrochloric acid, the iron was re-extracted into water.[*] The

aqueous solution was acidified to 1 $\underline{N}$ hydrochloric acid and antimony

and tellurium sulfides precipitated. Ferric hydroxide was precipitated

with ammonia gas using barium and strontium "holdback" agents. The

precipitate was dissolved in hydrochloric acid, ruthenium carrier added,

and the solution fumed with perchloric acid. Ferric hydroxide was

reprecipitated with ammonia and dissolved in nitric acid. Niobium

oxalate complex was added and niobium pentoxide precipitated. Barium

sulfate and silver iodide were also precipitated to remove radioactive

impurities. Ferric hydroxide was reprecipitated with ammonia and dis-

solved in 3 $\underline{N}$ nitric acid. Hydrofluoric acid was added and barium

fluozirconate precipitated. Ferric hydroxide was precipitated from the

supernatant and dissolved in 9 $\underline{N}$ hydrochloric acid for extraction with

di-isopropyl ether. Upon re-extraction into water, the iron was

precipitated with ammonia. The hydroxide was washed and ignited, and

the iron determined by weighing as Fe_2O_3.

Cobalt.-- The acidic target solution containing cobalt carrier

was made strongly ammoniacal. The supernatant was removed and the

precipitate washed twice with hot aqueous ammonia. From the combined

supernatant and washings, ferric hydroxide and barium and strontium

carbonates were precipitated. The supernatant was acidified with

hydrochloric acid and the acid sulfide group contaminants removed by

precipitation with hydrogen sulfide gas. The solution was made basic

and cobalt sulfide precipitated. The sulfide was dissolved in

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[*]It was recently found that the yield of iron was materially in-
creased by re-extraction into an acetic acid-sodium acetate buffer
solution instead of into water.

concentrated nitric acid which was then diluted for precipitation of silver chloride. The supernatant was made basic with potassium hydroxide to precipitate cobalt hydroxide. This was dissolved in 2 to 3 N acetic acid. Nickel "holdback" agent was added and potassium cobaltinitrite precipitated. The precipitate was dissolved in concentrated hydrochloric acid and the procedure repeated beginning with the ferric hydroxide - alkaline earth carbonate precipitations. The second cobaltinitrite precipitate was dissolved as before and potassium cobaltinitrite re-precipitated. This final precipitate was washed, dried at $110^{o}C$ and cobalt determined by weighing as $K_3Co(NO_2)_6 \cdot H_2O$.

Nickel.-- The acidic target solution containing nickel carrier was made strongly ammoniacal. The supernatant was removed and the precipitate washed twice with hot aqueous ammonia. From the combined supernatant and washings, ferric hydroxide and barium and strontium carbonates were precipitated. The solution was boiled gently to remove some of the excess ammonia, and dimethylglyoxime added to precipitate the nickel. The precipitate was dissolved in concentrated nitric acid which was then diluted to approximately 1 N. Copper, palladium, and antimony sulfides were precipitated several times. The supernatant was boiled to remove excess hydrogen sulfide, then made basic with ammonia gas. Ferric hydroxide was precipitated again. The supernatant was made 0.5 N in hydrochloric acid and palladium dimethylglyoxime was precipitated. The supernatant was neutralized and nickel dimethylglyoxime precipitated. After reprecipitation and washing, the nickel dimethylglyoxime was dried at $110^{o}C$ and weighed as such.

Copper.-- The acidic target solution containing copper carrier was
made strongly ammoniacal. The supernatant was removed and the precipi-
tate washed twice with hot aqueous ammonia. From the combined super-
natant and washings, ferric hydroxide was precipitated. A 5 percent
solution of sodium thiocyanate and sodium sulfite was added to the
supernatant and it was acidified with glacial acetic acid, after
"holdback" agents of the ammonia complex group elements had been added.
The cuprous thiocyanate precipitate was dissolved in a minimum amount
of concentrated nitric acid and the solution made 2 $\underline{N}$ in hydrochloric
acid. Cadmium, yttrium, and strontium "holdback" agents were added,
and copper precipitated as the sulfide. The cupric sulfide was dissolved
in concentrated nitric acid which was diluted to approximately 4 $\underline{N}$ for
the precipitation of silver chloride. The supernatant was buffered
with ammonium hydroxide - sodium acetate, and copper thiocyanate again
precipitated. The precipitate was washed, dried at $110^{\circ}C$, and copper
determined by weighing as CuCNS.

Zinc.-- The nitric acid solution of the target containing zinc
carrier was diluted to approximately 1 $\underline{N}$ and zinc mercurithiocyanate
precipitated in the presence of oxalic acid. The precipitate was dis-
solved in 4 $\underline{N}$ nitric acid, the solution was diluted and the zinc mer-
curithiocyanate reprecipitated twice. The precipitate was redissolved,
the solution diluted, and copper, silver, tin, antimony, and bismuth
sulfides precipitated. The supernatant was buffered with ammonium
acetate, and zinc sulfide precipitated. The precipitate was dissolved
in concentrated hydrobromic acid and evaporated to dryness several
times with bromine-hydrobromic acid to remove selenium. The residue

was taken up in 1 $\underline{N}$ sodium hydroxide to which sodium carbonate had been added. Ferric hydroxide and barium and strontium carbonates were precipitated from this solution. The supernatant was acidified to 1 $\underline{N}$ in hydrochloric acid. Zinc mercuric thiocyanate was precipitated, washed, dried at 110^oC and weighed as such. The gravimetric factor was determined empirically. [N.B. A better sample for counting purposes may be obtained by dissolving the final zinc mercurithiocyanate precipitate, removing the mercury by precipitating it as mercuric sulfide, neutralizing to methyl orange endpoint, and precipitating zinc ammonium phosphate at about 80^oC.[29] The precipitate is dried at 105^oC and weighed as $Zn(NH_4)PO_4$.]

Arsenic.-- The nitric acid solution of the target containing arsenic carrier was boiled nearly to dryness. (N.B. A more recent method removes the nitrate by boiling with formic acid.)[30] The residue was taken up in 6 $\underline{N}$ hydrochloric acid, a crystal of ammonium iodide added, and arsenic and germanium sulfides precipitated at ice temperature. The sulfides were washed with 6 $\underline{N}$ sulfuric acid saturated with hydrogen sulfide. The sulfides were dissolved in concentrated ammonium hydroxide. The solution was washed into a glass still, and tellurium, antimony and tin "holdback" agents added. Concentrated hydrochloric acid was added and germanium tetrachloride was distilled from the solution in a stream of chlorine gas. Hydrogen chloride gas was passed into the residue to remove the chlorine. Cuprous chloride was added to reduce the arsenic to arsenic(III), and arsenic trichloride was distilled from the solution in a stream of hydrogen chloride. The arsenic trichloride was trapped in concentrated hydrochloric acid at

ice temperature. Arsenic(III) sulfide was precipitated from the distillate by the addition of hydrogen sulfide gas. The sulfide was dissolved in concentrated ammonium hydroxide, tellurium, antimony, and tin carriers added, and the arsenic trichloride distilled from hydrochloric acid as above. Arsenic(III) sulfide was again precipitated from the distillate and dissolved in concentrated ammonium hydroxide. The solution was acidified with concentrated hydrochloric acid and arsenic(III) sulfide precipitated with hydrogen sulfide. The precipitate was washed, dried at $110^{\circ}C$, and arsenic determined by weighing as As_2S_3.

Selenium.-- The uranium target was dissolved under reflux in hydrobromic acid containing bromine, and selenium and tellurium carriers. After solution was complete, the reflux column was replaced by a still head and selenium tetrabromide distilled into bromine water kept at ice temperature. After completion of the distillation, an equal volume of concentrated hydrochloric acid was added to the bromine water at ice temperature, and sodium bisulfite added to reduce the selenium to the red allotropic modification of the zero oxidation state. After washing, the selenium was dissolved in hydrobromic acid containing bromine and tellurium "holdback" agent, and redistilled. The reduction and distillation were repeated. The selenium from the final reduction was washed with water and dried with ethanol and diethyl ether. The precipitate was then dried at $110^{\circ}C$ and selenium determined by weighing as Se.

Bromine.-- The uranium target was dissolved under reflux in nitric acid containing a known amount of freshly precipitated silver bromide

(to prevent loss of bromine by volatilization). Exchange of the radio-
active bromine or bromide ions with the inert bromide of silver bromide
is presumed to be rapid and complete.[31] After separation from the target
solution, the silver bromide was washed well and metathesized to silver
sulfide in ammoniacal solution. The supernatant was carefully acidified
to 1 $\underline{N}$ in nitric acid, allowing the excess hydrogen sulfide to escape.
Iodide carrier was added, oxidized to iodine with 0.1 $\underline{M}$ sodium nitrite,
and extracted into carbon tetrachloride. The iodine extraction was
repeated twice. The bromide was then oxidized to bromine by potassium
permanganate and extracted into carbon tetrachloride. The organic
phase was washed with 1 $\underline{N}$ nitric acid containing permanganate. The
carbon tetrachloride was stirred with water and sodium bisulfite solu-
tion added until the bromine had been reduced to bromide (and conse-
quently gone into the aqueous phase). The bromide was then precipi-
tated by the addition of silver nitrate, washed, dried at $110^\circ C$, and
weighed as AgBr.

Rubidium.-- An aliquot of the nitric acid solution of the target
containing cesium and rubidium carriers was evaporated to fuming with
perchloric acid. After cooling, absolute ethanol was added and the
solution stirred at ice temperature. The cesium and rubidium per-
chlorates were washed with absolute ethanol and dissolved in 0.2 $\underline{N}$
hydrochloric acid. Silver chloride was precipitated, then tellurium,
ruthenium, tin, antimony, and silver sulfides. The solution was made
basic with sodium hydroxide, and lanthanum, cerium, yttrium, iron,
zirconium, and niobium precipitated. Barium and strontium carbonates
were then precipitated. The series of sulfide, hydroxide and carbonate

precipitations was repeated several times. The final supernatant
was evaporated to fuming with perchloric acid. The perchlorates were
washed with absolute ethanol, dissolved, and reprecipitated. The
perchlorates were then dissolved in 6 $\underline{N}$ hydrochloric acid and the cesium
precipitated with silicowolframic acid. (For further purification of
cesium see below.) More cesium carrier was added to the supernatant
and cesium silicowolframate again precipitated. The supernatant was
evaporated to near dryness. Chloroplatinic acid and ethanol were added
to precipitate rubidium chloroplatinate. The precipitate was washed
with ethanol, dried at 110°C and rubidium determined by weighing as
Rb_2PtCl_6.

Strontium.-- The target was dissolved in a minimum volume of con-
centrated nitric acid containing carriers of the elements to be isolated.
After solution was complete, red fuming nitric acid was added and the
tube chilled in an ice bath. The precipitate, containing the nitrates
of strontium, barium, and coprecipitated radium, was centrifuged and
the supernatant decanted. The nitrates were dissolved in a minimum
amount of warm water and reprecipitated with fuming nitric acid. The
nitrates were redissolved and the solution made 4 $\underline{N}$ in nitric acid.
Silver chloride was precipitated to remove acid insoluble materials
which might have been carried by the alkaline earth nitrates. The
supernatant was made basic with ammonia gas and ferric hydroxide pre-
cipitated to remove traces of ammonium hydroxide group impurities and
any insoluble material which might have remained in the solution. The
supernatant was buffered to pH 5 with acetic acid - ammonium acetate.
Barium chromate (carrying radium) was separated from the strontium

by precipitation at $95^{o}C$. Barium chromate was again precipitated from the supernatant in order to remove any traces of radioactive barium or radium. Strontium was precipitated as the carbonate, washed, dissolved in hydrochloric acid to remove carbon dioxide and reprecipitated as the oxalate for weighing.

Zirconium.-- The nitric acid solution of the target containing zirconium carrier was made about 4-5 $\underline{N}$ in nitric acid and about 8 $\underline{N}$ in hydrofluoric acid. Lanthanum and yttrium fluorides were precipitated twice to remove contaminants of the rare earth and actinide series. Barium was added to the supernatant to precipitate barium fluozirconate. The precipitate was dissolved in 8 $\underline{N}$ nitric acid saturated with boric acid. The foregoing precipitations were repeated for additional purification. The final $BaZrF_6$ precipitate was dissolved in approximately 3 $\underline{N}$ hydrochloric acid with boric acid added to complex the fluoride. The barium was removed by precipitation as barium sulfate. The supernatant was diluted to approximately 2 $\underline{N}$ in acid and the zirconium precipitated by 6 percent cupferron solution in the cold (ice bath). The precipitate was washed with the reagent, transferred to a porcelain crucible, ignited and weighed as ZrO_2. This procedure probably effects no separation from hafnium. However, the yield of hafnium isotopes is less than 10^{-3} that of the zirconium.

Niobium.-- Niobium carrier, complexed with oxalate, was added to a measured fraction of the target solution. Niobium pentoxide was precipitated by digesting in an 8 $\underline{N}$ nitric acid solution containing potassium bromate. The oxide was dissolved in saturated oxalic acid. The solution was taken up in 6 $\underline{N}$ hydrochloric acid and extracted with cupferron into

chloroform at $5^{\circ}C$. The aqueous phase was re-extracted and the combined

chloroform layers washed with cold 6 $\underline{N}$ hydrochloric acid containing

cupferron. The chloroform layer was boiled with concentrated nitric

acid and potassium bromate added to precipitate niobium pentoxide. The

oxide was dissolved in dilute nitric acid containing hydrofluoric acid.

Barium fluozirconate precipitated to remove zirconium, yttrium, and

lanthanide contaminants. Boric acid was added to complex the fluoride,

and niobium pentoxide was precipitated from strongly ammoniacal solution.

After washing, the oxide was dissolved in saturated oxalic acid, then

reprecipitated from 8 $\underline{N}$ nitric acid containing potassium bromate. After

washing, the precipitate was ignited and weighed as Nb_2O_5.

Molybdenum.-- The uranium target was dissolved in hydrochloric acid

containing molybdenum carrier and the solution completed by the addition

of minimum amounts of nitric acid. The solution was boiled to reduce

the amount of nitrate and bring the concentration of hydrochloric acid

to about 6 $\underline{N}$. Molybdenum was extracted with several portions of diethyl

ether. (A minimum amount of liquid bromine was added to the aqueous

phase to ensure that molybdenum was present in the VI oxidation state.)

The ether layers were combined, washed with 6 $\underline{N}$ hydrochloric acid, and

evaporated over water. Ferric ion and niobium oxalate complex were

added to the solution, and their hydrated oxides precipitated by addition

of ammonia. The supernatant (containing molybdenum as molybdate) was

acidified to about 0.5 $\underline{N}$ in hydrochloric acid. Molybdenum alpha-benzoin

oxime was precipitated from the chilled solution. After washing with

0.5 $\underline{N}$ sulfuric acid, the oxime was dissolved in concentrated nitric acid

and destroyed by boiling carefully to fuming with perchloric acid.

After the solution had been cooled and diluted, ferric hydroxide was
precipitated using sodium hydroxide. The supernatant was acidified and
molybdenum alpha-benzoin oxime reprecipitated and destroyed. After
another precipitation of ferric hydroxide, the supernatant was made just
acid and buffered with sodium acetate. Molybdenum was precipitated as
silver molybdate, washed, dried at $110^{\circ}C$ and weighed as Ag_2MoO_4.

Ruthenium.-- Ruthenium and osmium carriers were added to the nitric
acid solution of the target which was then boiled to remove osmium
tetroxide. Iodide carrier was added and oxidized with bismuthate to
prevent distillation of iodine. Molybdenum was added and complexed
with phosphate to prevent distillation of molybdenum. Ruthenium tetroxide
was then distilled from fuming perchloric acid in an air stream. To the
distillate, trapped in 6 $\underline{N}$ sodium hydroxide, ethanol was added and the
solution boiled to coagulate ruthenium oxide. This may be reoxidized
to the higher state and distilled from perchloric acid again for addi-
tional purification. The oxide recovered from the alcoholic sodium
hydroxide was dissolved in hot 6 $\underline{N}$ hydrochloric acid. The solution was
diluted and the ruthenium reduced to the metallic state with magnesium.
The ruthenium was washed, dried at $110^{\circ}C$ and weighed as the metal.

Palladium.-- The target solution containing palladium carrier was
made about 0.4 $\underline{N}$ in nitric or hydrochloric acid. Inactive nickel was
added as a "holdback" agent. The palladium was precipitated with
dimethylglyoxime and the precipitate washed with dilute acid (0.2 $\underline{N}$).
The palladium dimethylglyoxime was dissolved in concentrated nitric
acid (destroying the dimethylglyoxime), the solution diluted, and ferric
hydroxide precipitated twice with ammonia. Silver iodide was precipitated

twice (using excess iodide) and the solution reacidified to 0.4 $\underline{N}$ in

hydrochloric acid. After centrifuging out any silver chloride which

might have been present, the palladium was precipitated with dimethyl-

glyoxime. The procedure could be repeated for additional purification.

After washing, the final palladium dimethylglyoxime precipitate was

dried at 110°C and weighed as Pd-dmg.

Silver.-- The target was dissolved in nitric acid containing silver

carrier (to avoid adsorption of trace amounts of silver by the glass).

The solution was diluted to approximately 4 $\underline{N}$ in nitric acid and silver

chloride precipitated from a boiling solution with stirring. Coagulation

is rapid and complete under these conditions. The silver chloride was

dissolved in aqueous ammonia and ferric hydroxide was precipitated to

remove ammonium hydroxide group contaminants. The silver was recovered

by precipitation as silver sulfide. The sulfide was dissolved in con-

centrated nitric acid which was boiled, then diluted to approximately

4 $\underline{N}$ for precipitation of silver chloride. The cycle was repeated for

additional purification. After dissolving the final precipitate of

silver sulfide, the solution was diluted and made basic with ammonia.

Ferric hydroxide was precipitated to remove any sulfur which might have

been present. The supernatant was made approximately 4 $\underline{N}$ in nitric acid

and silver chloride precipitated. After washing and drying at 110°C,

silver was determined by weighing as silver chloride. All silver

chloride precipitations except the final one should be made in the

presence of palladium and cadmium "holdback" agents to minimize occusion

of radioactive contaminants on the surface of the silver chloride.

Cadmium.-- The target solution containing cadmium carrier and copper
was made basic with ammonia. Cadmium and copper were leached out of the
massive precipitate by hot aqueous ammonia. The completeness of the
extraction was indicated by the blue color of the cupri-ammonium complex.
Iron, lanthanum, and indium were precipitated from the ammoniacal solu-
tion. Cadmium and copper sulfides, precipitated from the supernatant,
were dissolved in concentrated nitric acid. The solution was diluted
to approximately 4 $\underline{N}$ and silver chloride precipitated. The supernatant
was neutralized with ammonia; enough solid potassium cyanide added to
complex copper; and cadmium sulfide precipitated with hydrogen sulfide.
The cadmium sulfide was dissolved in 6 $\underline{N}$ hydrochloric acid, the solution
diluted to 2 $\underline{N}$, and palladium sulfide precipitated from the hot solution.
Antimony sulfide was precipitated from the supernatant. After centri-
fugation and decantation, the supernatant was made basic with ammonia
and cadmium sulfide precipitated. The precipitate was washed, dried
at $110^{\circ}C$, and cadmium determined by weighing as CdS.

Tin.-- The target was dissolved in hydrochloric acid containing tin
carrier. A minimum amount of nitric acid was used to complete the solu-
tion of the uranium. A portion of the target solution was taken and
diluted to approximately 1 $\underline{N}$ in acid. Stannic sulfide was precipitated
from the hot solution. After washing, the sulfide was dissolved in
concentrated hydrochloric acid and the hydrogen sulfide boiled out. The
solution was diluted to 2-2.5 $\underline{N}$ and antimony sulfide precipitated from
the hot solution. The acidity of the supernatant was reduced by the
addition of ammonium hydroxide, and stannic sulfide again precipitated.
After dissolving the sulfide in hydrochloric acid and boiling out hydrogen

sulfide, the solution was diluted and ruthenium, zirconium, niobium,

cadmium and iron carriers added. The solution was made basic with excess

6 $\underline{N}$ sodium hydroxide. Iron was added to the supernatant and the hydroxide

again precipitated. The supernatant was acidified and tin precipitated

with hydrogen sulfide. The sulfide was dissolved in hydrochloric acid

and antimony sulfide reprecipitated. After the tin sulfide was recovered

from the supernatant, it was dissolved in nitric acid and metastannic

acid precipitated by the addition of solid ammonium nitrate. The pre-

cipitate was filtered, ignited, and weighed as SnO_2.

A modification to this procedure has been suggested by McDonell.[32]

The antimony was removed by reduction to metallic antimony with iron.

The solution was filtered through "fine" grade sintered glass funnels.

Tin was recovered by precipitation of the sulfide from the filtrate.

In the sodium hydroxide precipitation of possible contaminants, ammonium

polysulfide was added to remove certain elements, such as lead, which

would otherwise be soluble in the basic solution. The final precipitate

of stannic sulfide was dissolved in ammonium nitrate solution with small

amounts of 6 $\underline{N}$ nitric acid (instead of in concentrated nitric acid)

to minimize the amount of sulfur formed.

Tellurium.-- The target was dissolved in hydrochloric acid contain-

ing tellurium carrier with the minimum amount of nitric acid necessary

to complete solution. A portion of the target solution, to which sele-

nium carrier has been added, was boiled to near dryness with concentrated

hydrobromic acid two or three times. The residue was taken up in con-

centrated hydrochloric acid, selenium carrier added, and the red allo-

tropic modification of selenium precipitated by the addition of sulfur

dioxide to the solution at $5^{\circ}C$. The supernatant was diluted to approxi-
mately 3 $\underline{N}$ in hydrochloric acid and tellurium was precipitated by the
addition of sulfur dioxide. The tellurium was dissolved in a few drops
of concentrated nitric acid, the excess nitric acid evaporated, and
the solution diluted. 6 $\underline{N}$ sodium hydroxide was added dropwise until
the precipitate of tellurous acid which formed was thoroughly dissolved.
Ferric hydroxide was precipitated from this solution. Tellurium was
recovered by precipitation with sulfur dioxide from the acidified super-
natant. The procedure was repeated for additional purification. The
final tellurium precipitate was washed, dried at $110^{\circ}C$, and weighed as Te.

Cesium.-- (See rubidium procedure, page 46.) The cesium and rubi-
dium perchlorates were taken up in 6 $\underline{N}$ hydrochloric acid and cesium
precipitated with silicowolframic acid. After digestion at $95^{\circ}C$, the
solution was chilled and centrifuged. The precipitate was washed with
6 $\underline{N}$ hydrochloric acid and dissolved in dilute sodium hydroxide. Upon
reacidification, wolframic oxide and silica precipitated. Inert rubidium
was added as a "holdback" agent, and the cesium reprecipitated with
silicowolframic acid. The precipitate was dissolved and cesium repre-
cipitated as before. The precipitate was washed with 6 $\underline{N}$ hydrochloric
acid, dried at $110^{\circ}C$ and the cesium determined by weighing as $Cs_8SiW_{12}O_{42}$.

Barium.-- (See strontium procedure, page 47.) The barium chromate
separated from the strontium was washed with water at $95^{\circ}C$. The pre-
cipitate was dissolved in a minimum amount of hot 6 $\underline{N}$ hydrochloric
acid. The solution was taken up in ether-concentrated hydrochloric
acid, 1:5 by volume, the "ether-HCl reagent." Upon chilling at ice
temperature, the barium chloride precipitated. The chloride was dissolved

in a minimum amount of distilled water and reprecipitated twice. The

final precipitate was washed with absolute alcohol and with ether,

and dried in a vacuum desiccator. The barium was determined by weigh-

ing as $BaCl_2 \cdot 2H_2O$.

Hafnium.-- Hafnium and zirconium were separated together from

other fission products by the above described procedure for zirconium.

Hafnium was separated from zirconium by elution from cation exchange

resin with constant boiling hydrochloric acid.[33] Since the hafnium

is eluted first, tailing from the higher activity zirconium peak does

not interfere.

Osmium.-- After carefully dissolving the uranium target in nitric

acid under reflux, carrier osmium having previously been added to the

nitric acid, iodine and bromine "holdback" agents were added, oxidized

with sodium bismuthate, and osmium tetroxide distilled into 6 N sodium

hydroxide. Hydrogen sulfide gas was passed in to precipitate osmium

sulfide. After washing, the sulfide was dissolved in concentrated

nitric acid, and the distillation-precipitation cycle repeated twice.

The sulfide precipitate was then dissolved in perchloric acid. Molyb-

denum "holdback" agent was added and complexed with phosphate. Osmium

tetroxide was distilled from this solution. After precipitation of

the sulfide, the perchloric acid distillation was repeated. The

solution of osmium in 6 N sodium hydroxide was carefully acidified

with hydrochloric acid and the osmium(VIII) reduced with magnesium

to metallic osmium.

Gold.-- Gold was separated as the metal by a series of precipita-

tions from chloride-free nitric acid solution with sulfur dioxide in

the presence of mercury, tellurium, and platinum "holdback" agents.

The gold was then dissolved in a minimum of aqua regia and taken up

in 2 $\underline{N}$ hydrochloric acid from which it was extracted by ethyl acetate.

Gold was recovered by evaporating the organic layer over dilute nitric

acid and precipitating metallic gold with sulfur dioxide.

Radium.-- (See barium procedure, page 54.) The final precipitate

of barium chloride was washed with alcohol to remove any excess hydro-

chloric acid. The precipitate was then dissolved in water and equili-

brated with Dowex-50 cation exchange resin. By elution through a

column packed with resin, using pH 8 ammonium citrate solution, the

barium and radium fractions were separated.[34] Samples for alpha count-

ing were mounted on platinum disks and flamed to destroy and remove

citrate, leaving essentially weightless samples.

General Procedure for Isolating and Separating Rare Earths[*]

General.-- One milligram of each rare earth to be isolated (except

promethium, for which tracer was added) was added as carrier, the

total amount being of the order of twelve milligrams. The group was

separated by precipitation of the fluorides from 2 $\underline{N}$ nitric acid

solution by addition of hydrofluoric acid. (Some uranyl fluoride may

be carried if the concentration of uranium is high enough. For one

of the bombardments which used an eight gram uranium target, the

uranium was extracted with ether from the nitric acid solution before

the rare earth fluorides were precipitated.) The fluorides were dis-

solved in 8 $\underline{N}$ nitric acid saturated with boric acid, and the rare earths

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[*]Modification of a procedure used by Wilkinson and Hicks.[25]

were recovered as hydroxides by precipitation with ammonia gas. The
hydroxides were washed to remove excess ammonium salts, and dissolved
in dilute hydrochloric acid. Selected sulfides (copper, arsenic,
antimony, tin, silver, palladium, bismuth) were precipitated from this
solution by addition of hydrogen sulfide gas to remove contaminants
belonging to the acid sulfide group. From a hydrochloric acid con-
centration of the order 0.3 N, the rare earths were then precipitated
as oxalates. After washing, the oxalates were dissolved in concentrated
nitric acid containing potassium bromate. The solution was diluted
and the rare earth hydroxides reprecipitated. The hydroxides were
dissolved in 0.2 N hydrochloric acid which was then diluted to between
pH 1 and pH 2. The solution was equilibrated with Dowex-50 ion ex-
change resin for five minutes at $95^{\circ}C$.

Cerium.-- In addition to separation by the general procedure,
cerium has also been separated from the other rare earths by oxidation
to cerium(IV) and precipitation of ceric iodate. Ceric phosphate
precipitations have also been used to separate cerium from the other
rare earths.

Europium.-- Europium was separated from the other rare earths by
reduction to europium(II) with zinc amalgam in a nitrogen atmosphere.
The other rare earths were precipitated as hydroxides from a carbonate
free solution leaving the europium(II) in solution. The europium
was recovered by ozone oxidation to europium(III) and hydroxide or
oxalate precipitation. The cycle may be repeated for additional
purification.

Column Separations

The resin, with adsorbed rare earths, was placed in a band at the top of a glass column 4 mm in diameter and 10 cm high, previously packed with Dowex-50 resin, and eluted with constant boiling (~6 $\underline{N}$) hydrochloric acid at a temperature of $87^{\circ}C$, using a flow rate of approximately 0.1 ml per minute. After about two hours, the rare earths from lutetium through neodymium had been eluted, leaving behind about 30 percent of the praseodymium, 50 percent of the cerium, 60 percent of the lanthanum and all of the actinium with its thorium daughter activities. The rare earths were recovered from the 6 $\underline{N}$ hydrochloric acid solution by precipitation of the hydroxides with ammonia gas. The hydroxides were washed, dissolved, and equilibrated with resin as before. The resin was then placed at the top of a glass column 7 mm in diameter and 20 cm high, previously packed with resin, and eluted with 0.25 $\underline{M}$ ammonium citrate-citric acid solutions at $87^{\circ}C$. Lutetium and ytterbium were separated by elution with pH 3.19 citrate at a flow rate of 0.14 ml per minute. Ytterbium, thulium, erbium, and holmium were separated by elution with pH 3.23 citrate at a flow rate of 0.21 ml per minute. For the lighter rare earths, pH 3.30 and pH 3.5 citrate solutions were used successively. Dysprosium-yttrium and europium-gadolinium were not separated under these conditions. The dysprosium-yttrium separation was made by elution with pH 3.05 citrate on another resin-packed column at room temperature.

Recovery and Chemical Yield

By means of a turntable actuated by a microflex timer, samples
of the elutriant were collected at regular intervals. Small aliquots
of these samples were mounted on counting plates for radioassay. From
the plot of relative activity versus sample number it was possible to
identify the various peaks representing the separated rare earths.
Identification of the peaks was made more definite by spectrographic
analysis.

From the radioassay plot it was possible to select the samples
comprising a given peak which were free from any contamination by the
material comprising neighboring peaks. These samples were combined
in a small erlenmeyer flask and fumed with sulfuric acid to which a
few drops of selenium(IV) solution were added as a catalyst. The
destruction of the citrate was completed by adding perchloric acid
and heating. The rare earth was recovered by precipitation as the
hydroxide. After washing out the ammonium perchlorate, the hydroxide
was dissolved and the oxalate precipitated. The oxalate was dried
in a vacuum desiccator at room temperature, and weighed to determine
the percentage recovery of the rare earth. (The oxalate was selected
for the gravimetric analysis because of the high molecular weight of
its stable hydrate. In this way small amounts of a rare earth could
be determined with greater accuracy since there are two to three
grams of the compound per gram of rare earth.)

APPENDIX II

Let us consider data which are of the following form: (1) the
total yield of isobars of charge Z-1 or less; (2) the independent yield
of the isobar of charge Z; and (3) the total yield of isobars of charge
Z + n + 1 or greater, where n is the number of isobars not available
for chemical separation, i.e., stable or short lived isobars. Let
the area under the histogram be equal to unity. Now let us define
a function E(X) such that:

$$E(X) = \int_{X}^{+\infty} f(Z)dZ \qquad\qquad (AII-1)$$

X being the abscissa with the center of the curve taken as the origin,
X being expressed in units of the standard deviation of the curve.
Then if we define y as shown in Figure 6 (taking y as a positive number
when in the direction shown), and the width of one block of the histo-
gram (in X units) is a, the experimental data may be described as
proportional to $E(y)$, $E(y-a) - E(\bar{y})$, and $E(b)$.

Figure 7 gives $E(y)/E(y-a)$ as a function of y for various values
of a. From the experimental value for $E(\bar{y})/E(y-a)$ one obtains from
this chart a table giving y as a function of a.
Now

$$\left| b \right| = (n + 1)a - \left| \bar{y} \right| , \qquad\qquad (AII-2)$$

so we have

$$y = f_1(a) \qquad\qquad (AII-3)$$

and

$$b = f_2(a) \qquad\qquad (AII-4)$$

therefore

$$E(y) = f_3(a) \qquad\qquad (AII-5)$$

$$E(b) = f_4(a) \qquad\qquad (AII-6)$$

therefore

$$E(b)/E(y) = f_5(a) \qquad\qquad (AII-7)$$

where $E(b)/E(y)$ can be obtained from the experimental data. Equation (AII-7), of the form $f_5(a) = $ constant, can then be solved by graphical means, giving a value for a, hence for y, and hence locating Z_0 the center of the curve as

$$Z_0 = Z - 1/2 + y/a . \qquad\qquad (AII-8)$$

Since a histogram block has a width of 1 charge unit and a standard-deviation units, the standard deviation of the curve is $1/a$.

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LIST OF ILLUSTRATIONS

Figure

1. The distribution of total fission product yields as a function of mass number.

2. Decay schemes for isobaric chains of mass 140 and of mass 166.

3. Standard curve of error histogram for mass 140 isobaric chain.

4. Standard curve of error histogram for mass 166 isobaric chain.

5. Hypothetical contour representation of bombardment product fission probabilities and contour representation of fission product yields.

6. Illustration for the fitting of a normal curve of error to isobaric yield data.

7. $E(y)/E(y-a)$ as a function of y for various a.
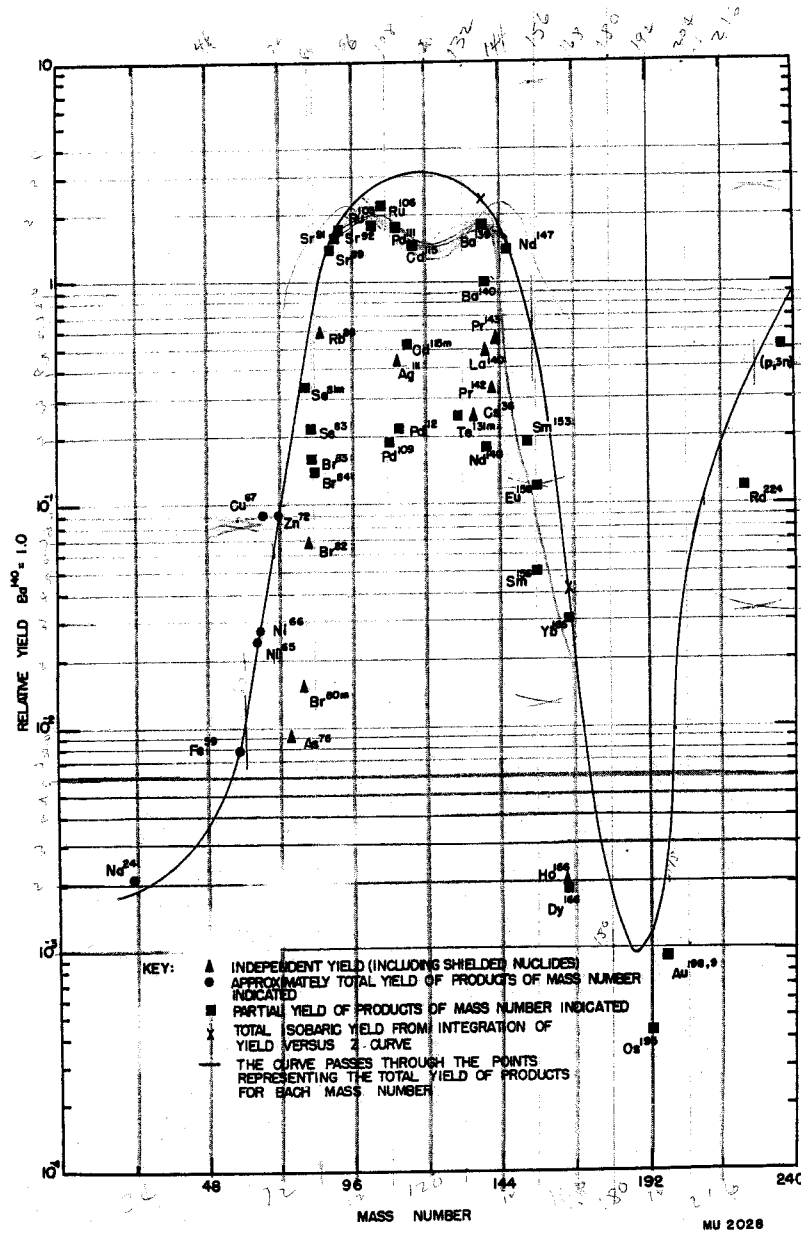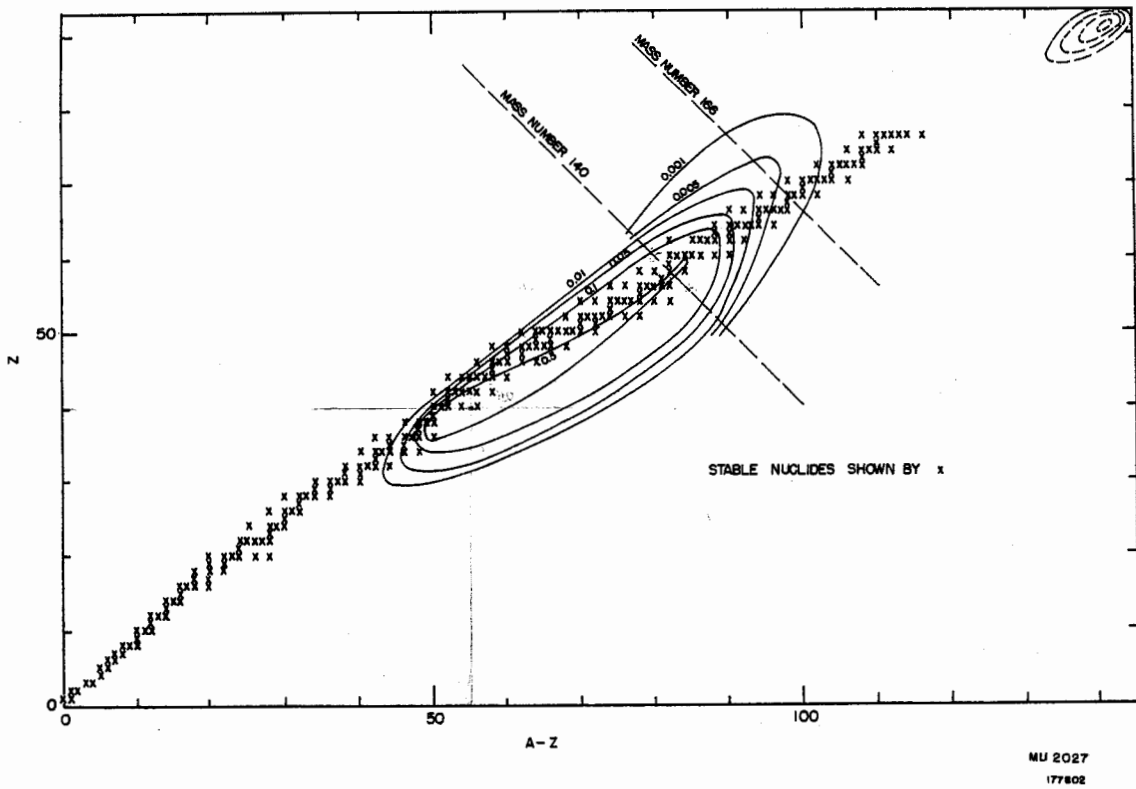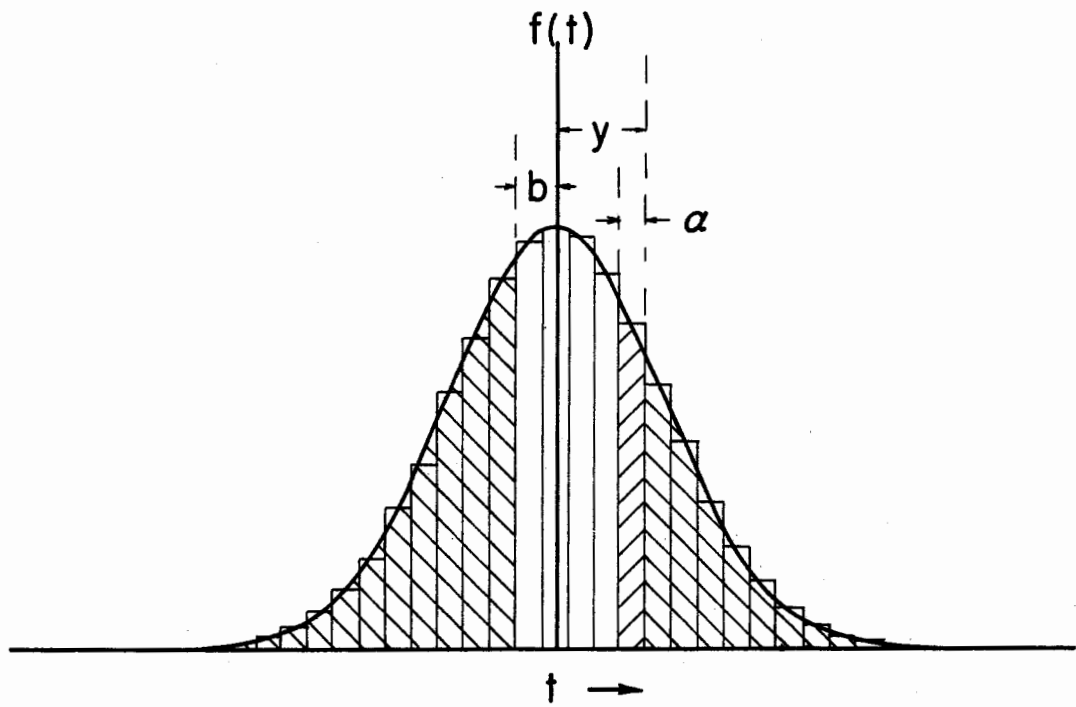
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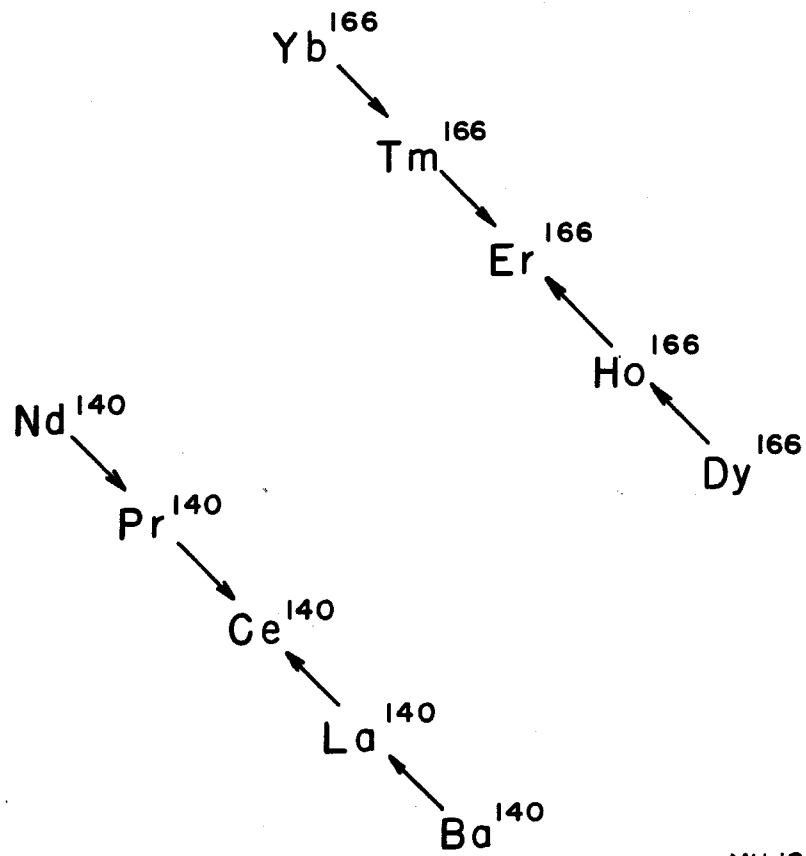
MU 1288

Fig. 2
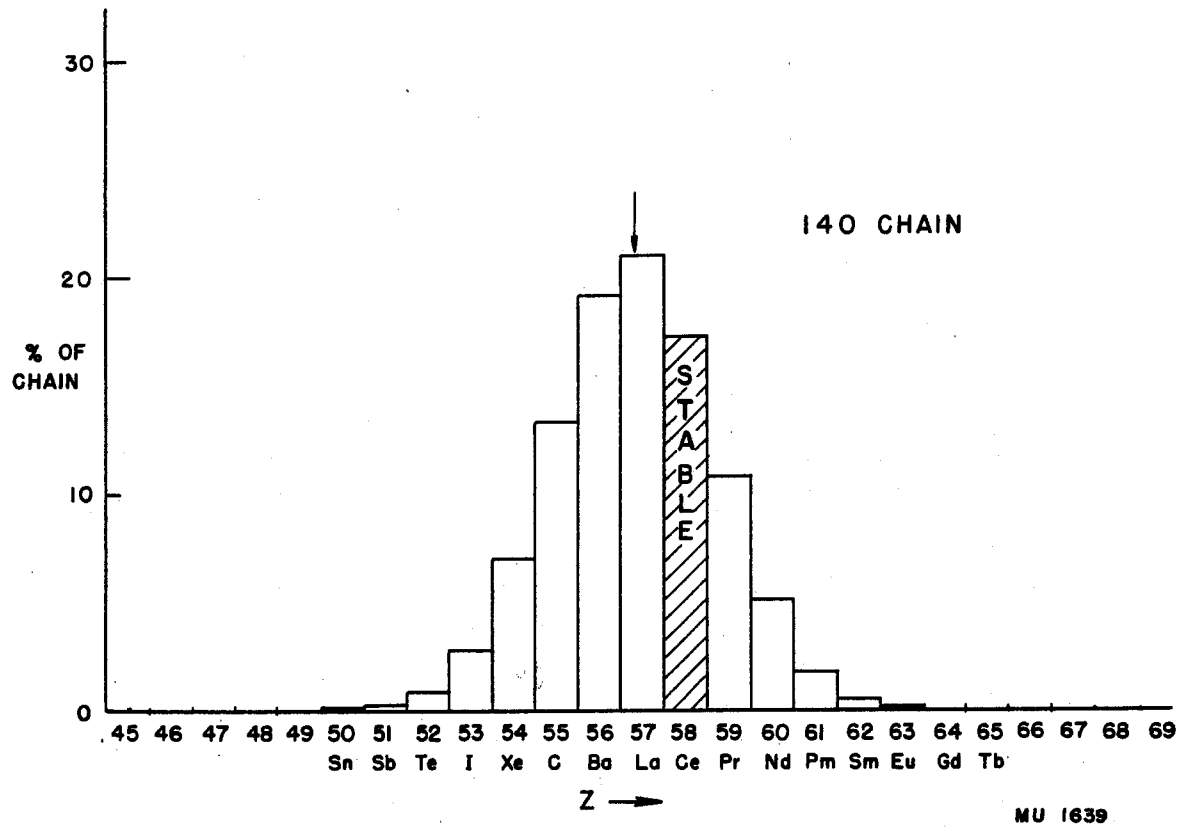
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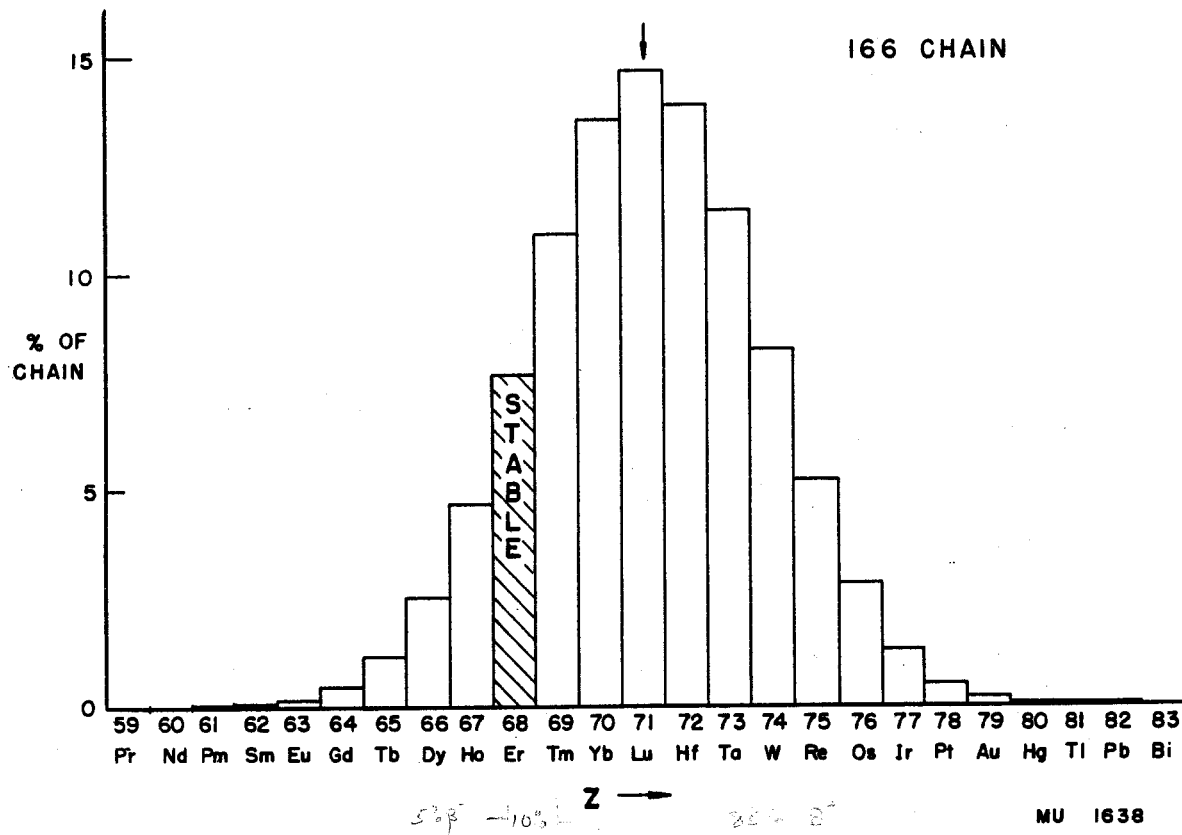
Fig. 4
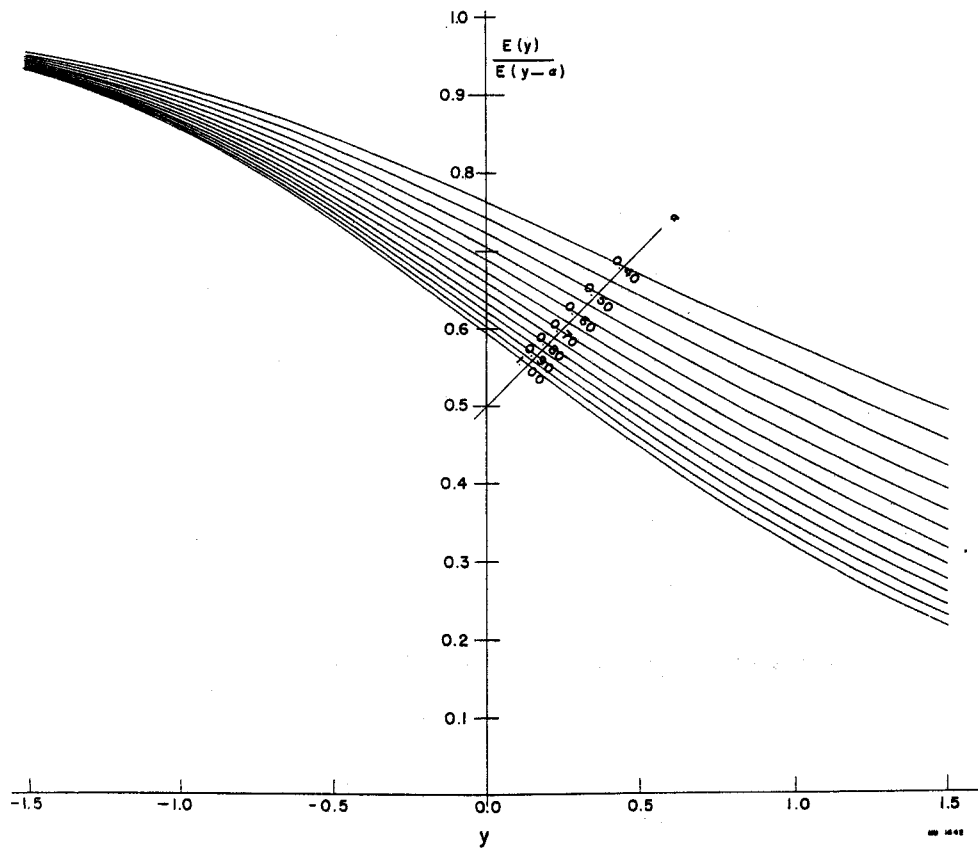
Fig. 5

$$E(x) = \int_x^{\infty} f(t)\, dt$$

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Fig. 6

Fig. 7