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*Radiation
Laboratory*

FISSION OF GOLD WITH
112-Mev C^{12} IONS:
A YIELD-MASS AND
CHARGE-DISTRIBUTION STUDY

BERKELEY, CALIFORNIA

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H. Marshall Blann
(Thesis)

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ABSTRACT

Fission-product cross sections have been measured radiochemically and mass-spectrometrically for gold bombarded with 112-Mev C^{12} ions. Cross sections for 43 nuclides have been measured for elements from nickel to barium. Thirty-six yields are either primary fission product yields (independent yields) or have been corrected (with less than 25% correction) so as to represent independent yields. The independent yields have been empirically systematized and a yield-mass curve has been constructed. The yield-mass curve is compared with the yield-mass curves obtained from the fission of bismuth with 22-Mev and 190-Mev deuterons. The yield systematics indicate that the sum of the mass numbers of complementary fission products is 13 ± 1 amu less than that of the compound nucleus, and the sum of the charges of complementary fission products is two units less than that of the compound nucleus. It is postulated that 9 ± 1 neutrons and an alpha particle must have been emitted. Evidence is presented that at least three and possibly more of the neutrons are emitted prior to fission.

The most probable charge of the fission products as a function of mass number has been determined empirically. It is shown that from mass number 80 to mass number 111 the Equal Charge Displacement (ECD) and Constant Charge Ratio (CCR) rules predict the empirical values equally well. For masses less than 80 and greater than 120 the ECD and CCR rules are shown to be equally poor, as the empirical distribution is

between them. The Minimum Potential Energy theory of charge distribution proposed by Present and modified and interpreted by Swiatecki is shown to predict the empirical points over the entire mass region studied ($A = 66$ to $A=135$) within experimental error (± 0.2 charge unit).

I. INTRODUCTION

Since its discovery by Hahn and Strassman in 1939, the fission process has been ambitiously investigated.¹ The general properties most often studied have been the manner in which the mass of the fissioning nucleus is divided between the fragments (mass distribution), the way in which the nuclear charge is distributed at the instant of fission (charge distribution), and the distribution of kinetic energy between the fragments. Various review articles summarize these results.²⁻⁴

Initial work was of necessity confined to fission of uranium with neutrons. But as accelerators became more common the nuclei of U^{233} , U^{235} , U^{238} , Th^{232} , Np^{237} , and Pu^{239} were subjected to merciless bombardment with protons, deuterons, and helium ions. Bohr's concept of the "compound nucleus" contributed significantly toward understanding the results of these surveys.⁵ The compound-nucleus theory predicts fusion of the incident particle with the bombarded nucleus; the incident particle will lose its identity as its energy is evenly distributed between all nucleons of the new compound system, and most important, the compound system will de-excite independently of how it was formed, but dependent only upon its mass, charge, and excitation energy.

Uranium fission produced by thermal and low-energy neutrons is decidedly asymmetric; that is, the yield-mass curve shows two peaks. As the energy of the bombarding particle is increased the fission mass yield curve becomes increasingly symmetric, until at sufficiently high energies there exists a single very broad peak. Newton (1949) was the first to observe the competition between the symmetric and asymmetric modes in his fission-excitation studies of thorium with up to 39-Mev helium ions.⁶ Since then fission excitations have been performed on many heavy-element nuclides with neutrons, protons, deuterons, and alpha particles.

Far fewer investigations have been performed on targets of atomic number less than that of thorium (90). Fairhall and Jensen have bombarded radium with 11-Mev protons and observed three distinct peaks in the mass yield curve corresponding to the asymmetric and symmetric modes.⁷ Fairhall also studied the system Bi^{209} plus 15-Mev and 22-Mev deuterons and observed a single narrow symmetric peak.⁸

Fairhall, Jensen, and Neuzil found a single narrow symmetric peak for the fission of lead isotopes with 25- to 42-Mev helium ions.⁹ Griffioen and Cobble found a symmetric peak for the fission of tantalum and rhenium with helium ions of more than 40 Mev, with asymmetric fission becoming relatively more important below 40 Mev.¹⁰ Much less work has been done on the fission of elements lighter than thorium at high excitation energy (greater than 50 Mev). Probably the best-known work in this region is the investigation by Goeckermann and Perlman of the fission of bismuth with 190-Mev deuterons.¹¹ Unfortunately, when the energy of the bombarding particle exceeds about 50 Mev per nucleon the compound-nucleus model becomes invalid, as direct interactions take place; nucleon-nucleon collisions cause cascade particles to be knocked out of the nucleus, resulting in a large number of excited nuclear species having a broad spectrum of excitation energy.¹²

The Berkeley heavy-ion linear accelerator (Hilac) provides a means of attaining high excitation energy in a single compound nucleus.* Projectiles ranging from He^3 to A^{40} are accelerated to an energy of 10 Mev per nucleon, far too low for the nucleon-nucleon cascade process. Gold was chosen as a target because it is monoisotopic and high-purity foils are available. Bombardments have been performed with C^{12} ions of up to 112 Mev. The predicted compound nucleus is At^{209} , with about 95 Mev of excitation. Such systems formed through heavy-ion bombardment have very high angular momenta; the average predicted for the above case is 40 units.¹³ With a known compound system, the mass distribution is more meaningful, and in particular the charge distribution has far more significance.

* Here we are considering only compound-nucleus formation; stripping reactions are ignored, since they should not contribute significantly to the fission cross section of gold.

Many workers have investigated the charge distribution in fission. The rules of the game have usually been to measure independent* and cumulative fission yields and then by trial and error determine which of several empirical rules gives the best correlation of the results. It is understood that there is no reason why any simple rule must explain the experimental results in the complex process of nuclear fission. Nonetheless, by learning more about the primary distribution of charge in fission, we can gain additional insight into the nature of the fission processes and the properties of nuclear forces.

*"Independent Yield" as used in this work refers to the yield of an isotope from the fission process, and does not include contributions to that yield from β^+ , β^- , or EC decay of other isotopes in the same mass chain.

II. EXPERIMENTAL PROCEDURE

A. The Target and Irradiations

1. The Target

For radiochemical studies the targets consisted of sheets of 0.1-mil gold. The actual arrangement in the target holder was a stack consisting of two 0.7-mil aluminum foils followed by the 0.1-mil gold foil, which was in turn followed by another 0.7-mil aluminum foil. The first aluminum foil served as a vacuum seal for the target holder; the other two aluminum foils sandwiched the gold target foil and served as catcher foils for the recoiling fission products. Photographs of the three types of target holders (Types I, II, and III) used in this work are shown in Appendix C (Figs. 22 and 23). In all runs in which absolute cross sections were to be determined target holder types I and II were used and an additional 0.25-mil aluminum foil ("crud foil") was placed well in front of the target. The purpose of this foil was to fully strip the carbon ions of electrons. It was important to know the charge state of the carbon ions, since they were all stopped by the target holders (I and II), which served as their own Faraday cups. In several bombardments of 3 to 5 minutes duration short-lived bromine and yttrium activities were sought. In these irradiations the interceptor target (III) was used. The interceptor target was used without a Faraday cup and only relative cross sections were determined.

A mass spectrometer was used to measure the relative yields of fission-produced isotopes of cesium and strontium. The target for the bombardment that yielded results consisted of a 1-mil aluminum beam degrader foil followed by a 0.5-mil gold foil. When thinner gold targets were used the number of cesium and strontium atoms formed was insufficient for mass spectrometry.

Spectrographic analysis (optical) of the 0.1-mil and 0.5-mil gold foils set an upper limit of 0.1% for heavy elements. The aluminum foils were 99.5% pure.

2. Irradiations

Irradiations were performed on the Berkeley heavy-ion linear accelerator (Hilac). The irradiations for radiochemical investigations varied from 3.00 minutes to 4 hours. For mass-spectrometric studies the irradiations were from 4 to 12 hours long.

B. Chemistry

The chemistry used in this work consisted mainly of standard radiochemical procedures. For such procedures references are given to the standard compilation with no further discussion. The procedures used to separate various carriers are discussed below, as well as the treatment for the mass-spectrometric target.

1. Chemistry for the Mass Spectrometer

The gold target foil was cut into many small squares (approx 3mmX3mm) and each square was monitored for net activity on a Health Chemistry "Juno" meter.* The two pieces having greatest activity were placed in a quartz cone; two drops of HCl and one drop of HNO₃ were added to dissolve the gold foil. The solution was run through a Dowex A-1 anion-exchange column which had about 5 mm of resin in a 3-mm-diameter quartz column, the bottom of which was stoppered with quartz wool. The column adsorbed the gold and allowed the fission products of interest to pass through into a quartz cone. The fission product solution was evaporated to dryness and was ready to be transferred to the sample filament of the mass spectrometer.

Care had to be taken to keep natural contamination to a minimum lest it completely obscure the fission-produced isotopes. The HCl used was made from conductivity water which had been redistilled through a quartz-condenser still and stored in polyethylene bottles. Hydrogen

*The "Juno" meter is a device for detecting alpha, beta, and (or) gamma radioactivity. It uses an air-filled ionization chamber with an aluminum rod as a collecting electrode.

chloride gas was bubbled through several traps and eventually into the conductivity water to form the HCl used. The HNO_3 used was obtained by boiling reagent grade nitric acid and condensing the nitric acid vapors on a large quartz test tube over the solution. The condensed nitric acid was collected in a quartz cone for use with the target. The column itself was washed alternately with conductivity water and HCl for several weeks before it was used.

2. Radiochemistry

Carriers were always added before the target was dissolved, and in every case the aluminum catcher foils were dissolved along with the gold target. The headings of the following paragraphs indicate which elements were studied in the indicated bombardment.

Iodine, Bromine. The target was dissolved in aqua regia in a standard taper flask having a condenser leading to a few ml of 0.5 N NaOH. After the target was dissolved the condenser was rinsed with the 0.5N NaOH followed by distilled water; the rinsings were added to the target solution. The solution was diluted and a few drops of 1M NaHSO_3 were added to precipitate the gold. The metallic gold was centrifuged and discarded. A few drops of 1M KNO_2 were added to oxidize I^- to I_2 , which was extracted into carbon tetrachloride.¹⁴ The final precipitate was silver iodide. A few drops of 0.1M KMnO_4 were then added and bromine was extracted into bromoform.¹⁵ The final sample was mounted as silver bromide. Bromoform was used rather than the carbon tetrachloride suggested in the standard procedure because of its much higher extraction coefficient for bromine. In acidic solution there should be no exchange problem.

Niobium, Zirconium. The target was dissolved in a lusteroid cone containing 2 ml of niobium carrier (24 mg), 1 ml of zirconium carrier (18 mg), 12 drops concentrated HF, and 2 ml 9 N aqua regia. The solution was added to a separatory cone with 15 ml conc. HCl; 15 ml of di-isopropyl ketone (DIPK) was added, and the mixture was stirred for 2 minutes. The niobium was extracted into the DIPK; the aqueous phase was saved for the zirconium procedure.¹⁶ The DIPK was washed with two 10-ml aliquots of conc. HCl, after which the niobium was extracted from the DIPK into

4 M HCl.¹⁷ The niobium-hydrochloric acid solution was made basic with ammonium hydroxide precipitating niobium oxide. The niobium oxide was dissolved in oxalic acid and reprecipitated from approx. 8 M HNO₃ by boiling with addition of potassium chlorate. The niobium was mounted as the oxide; the zirconium was mounted as the phenylarsenate.

Barium, Cesium, Niobium, Molybdenum. The target was dissolved in a solution containing the carriers (Ba^{++} , Cs^+ , Nb^{+5} , Mo^{+6}) and a minimum amount of hydrochloric and nitric acid. Hydrogen chloride gas was bubbled through the solution, which was in an ice bath; barium chloride precipitated.¹⁸ A DIPK solvent extraction was performed to remove niobium.¹⁷ The supernatant solution was made 8 M in HCl and put through a Dowex A-1 anion-exchange column, which adsorbed molybdenum.¹⁹ A few mg each of Ce^{+3} , Y^{+3} , La^{+3} , Zr^{+2} , Ba^{+2} , and Sr^{+2} carriers were added to the solution and a standard cesium separation was performed.²⁰ The final precipitates were niobium pentoxide, barium chloride, lead molybdate, and cesium chloroplatinate.

Silver, Molybdenum. The silver and molybdenum carriers were mixed together, precipitating some silver chloride. Hydrogen chloride gas was bubbled into the solution until the silver chloride redissolved as AgCl_2^- . The target was added to this solution plus a minimum amount of nitric acid. The solution was diluted with distilled water to precipitate silver chloride.²¹ The molybdenum was precipitated with α -benzoinoxime and further purified.²² Silver was mounted as silver chloride; molybdenum was mounted as lead molybdate.

Nickel, Molybdenum, Strontium. The target was dissolved in the solution of carriers with 2 ml of 9 N aqua regia. After dissolution the solution was diluted to 20 ml and a few drops of 1 M NaHSO₃ were added to precipitate the gold; the gold was centrifuged and removed. The solution was made basic with ammonia, and nickel was precipitated with dimethylglyoxime.²³ Sodium carbonate was added to the supernate to precipitate strontium carbonate.²⁴ The α -benzoinoxime precipitation was then used to separate molybdenum.²² Nickel was mounted as nickel dimethylglyoxime, strontium as the oxalate, and molybdenum as lead molybdate.

Bromine (Relative Cross Section). The target was dissolved in a centrifuge cone having 20 mg of bromide carrier and 3 ml 9 N aqua regia. Upon dissolution of the target the solution was diluted to 30 ml and a few drops of 1 M NaHSO_3 were added to precipitate the gold. The solution was centrifuged; several drops of 0.1 M KMnO_4 were added to the supernatant solution. The bromine (and some iodine) was extracted into 20 ml of bromoform. The bromoform was washed with 20 ml of water containing a drop of nitric acid. The bromine was back-extracted from the bromoform into 10 ml of water containing a drop of 1 M NaHSO_3 . A few mg of iodide carrier was added to the aqueous phase, two drops of 1 M KNO_2 were added, and the iodine was extracted into two aliquots of carbon tetrachloride. Excess silver ion was added to the aqueous and the silver bromide was filtered, washed with ether, dried, and mounted.

Yttrium (Relative Cross Section). The target was dissolved in the carrier solution with 2 ml of 9 N aqua regia. Hydrofluoric acid was added to the solution and the yttrium fluoride precipitate was centrifuged. The precipitate was washed in an aqueous hydrofluoric acid solution and again centrifuged. The yttrium fluoride was dissolved in a minimum of warm saturated boric acid. Twenty ml of 15 M HNO_3 was added and the yttrium was extracted into an equal volume of tributyl phosphate (TBP). The TBP was then washed with 10 ml of nitric acid, after which yttrium was extracted from the TBP into water.²⁵ The yttrium was precipitated as the oxalate from a slightly acidic solution (pH 3 to 5), filtered, dried under an infrared lamp, and mounted.

Nickel, Arsenic, Strontium, Molybdenum. The target-carrier solution was brought to 4 M in HCl and passed through a Dowex A-1 column which adsorbed the molybdenum.¹⁹ The arsenic was eluted with 9 ml of 3 M HCl . Twenty ml of 12 M HCl and 20 mg of potassium iodide were added to the arsenic solution, and hydrogen sulfide was bubbled in. The arsenious sulfide precipitate was filtered and purified further as described elsewhere.²⁶ The supernatant solution was boiled to remove hydrogen sulfide and evaporated to near dryness. The residue was dissolved in water and ammonium hydroxide was added to make the solution basic.

Nickel was then precipitated with dimethylglyoxime²³ and strontium was precipitated with sodium carbonate.²⁴ Arsenic was mounted as the penta sulfide, nickel as nickel dimethylglyoxime, molybdenum as lead molybdate, and strontium as strontium oxalate.

Yttrium, Niobium. Twenty ml of 15 M HNO_3 was added to the target-carrier solution; the acidic solution was heated nearly to boiling and potassium chlorate was added in small portions, destroying the niobium oxalate complex and precipitating niobium pentoxide. The niobium pentoxide was dissolved in oxalic acid and the solution was made 10 M in HCl ; a DIPK extraction was made for the niobium as previously described.¹⁷ The yttrium was extracted from the original nitric acid solution with TBP.²⁵ The TBP was washed with 10 ml of concentrated nitric acid and the yttrium was extracted from the TBP into 20 ml of water. An yttrium fluoride precipitate was formed by adding hydrofluoric acid to the 20 ml of water containing the yttrium nitrate. The yttrium fluoride was redissolved in warm boric acid and added to 10 ml of concentrated HNO_3 , and yttrium was re-extracted into TBP. Another nitric acid wash and extraction into an aqueous phase were performed, followed by the precipitation of yttrium oxalate. Niobium was mounted as niobium pentoxide.

C. Mounting and Counting of Samples

1. Sample Mounting

A set of glass filtering chimneys (Fig. 24 Appendix C) was machined by the Lawrence Radiation Laboratory glass shop to 1.90 cm inside diameter. Brass collars were made to fit the chimneys. The collars were squeezed together by means of springs or rubber bands, giving a tight seal between the chimney, the filter paper, and the sintered glass disc as shown in Fig. 24 (Appendix C). All samples were filtered with this apparatus, giving a precipitate uniformly distributed over an area of 2.84 cm^2 .

Aluminum plates, 50 mils thick and 2.5 by 3.5 inches, were used as supports for all samples. One such plate, with a 1-inch-diameter hole in its center, was used as a template to help in centering samples. The

template was placed over the sample plate and a 1-inch circle was drawn on the sample plate. Double-sided Scotch tape was placed over this circle, as shown in Fig. 25 (Appendix C). The dried sample on 1-inch filter paper was centered on the circle drawn on the plate and placed on the Scotch tape. The edges of the filter paper were pressed with tweezers to get firm contact all around the circumference. A piece of 0.1-mil rubber hydrochloride (0.5 mg/cm^2) was stretched over the sample and attached to the double-sided tape not covered by the filter paper. This thin film kept the sample from being lost, and also kept the counters from being contaminated.

2. Yields

When absolute cross sections were to be determined, filter papers were pretreated with the same reagents and dried in the same manner as they were to be during the actual "run." The filter papers were then preweighed. The filter papers with samples were dried and weighed, allowing a stoichiometric yield determination. After the samples had undergone sufficient radioactive decay to render further counting impractical, they were sent to the analytical laboratory for yield analysis. In general the yields obtained stoichiometrically and analytically agreed to within 5%.

3. Beta Counting

Beta particles were counted by means of an end-window proportional counter. Since the author had noted a lack of reproducibility in the end-window counters in the community counting room,* a special counter was installed in his laboratory where it could be given careful attention. This counter differed from the community counters in that it had a regulated high-voltage power supply and a new type of scaling unit. The detector unit was the standard Los Alamos design using a 1-inch-diameter window covered with 0.25-mil aluminized Mylar.²⁷ The electrode used was of the hypodermic needle type with a loop, approximately 6 mm in diameter, of 1-mil tungsten wire forming the collecting electrode.

*The proportional counters in the community counting room suffered from a nonregulated high-voltage supply, faulty high-voltage neon tube dividers, need of replacement of collecting electrode, and contamination of detector units.

When "house gas" was used as a source of flowing methane, it was possible to get plateaus 400 v wide having a slope of approx 0.3% per 100 v when the electrode was new. After approximately a month of use, the electrode characteristics changed until the plateau was only 1% per 100 v over a 400-v width. The β radiations from a uranium standard were counted each day for 5×10^5 to 1×10^6 counts. In general the count rate remained at $13,200 \pm 30$ cpm on Shelf 3 over a period of several months. When this count rate varied by 1% or more the electrode was changed, restoring the original characteristics.

Decay curves of one or two components were resolved graphically for activity at the end of bombardment. All decay curves of more than two components were resolved by using an IBM 704 least-squares program. This program (named "Frenic") is described in Appendix A.

When the decay curves had been analyzed there remained the problem of converting count rate to disintegration rate. In the past a series of corrections (from various sources) for such factors as geometry, backscattering, self-scattering, self-absorption, air and window absorption, etc. had been used. The need for these separate corrections was eliminated by conducting an empirical counter calibration by the method suggested by Bayhurst and Prestwood.²⁸ The empirical calibration curves obtained are of two types. The first is a family of curves representing the efficiency* on a given shelf as a function of sample thickness for β^+ and β^- emitters of various energies. The second is a plot of the efficiency versus average β energy for a given shelf, taken at some arbitrary sample thickness; such curves are shown in Appendix B. Shelf positions 1, 3, 5, and 8 were calibrated in this fashion in this work. The two families of efficiency curves made it possible to estimate an efficiency for any nuclide having a fairly well-known decay scheme and a β end-point energy of 400 kev or greater.

* "Efficiency" when applied to β counting in this work means the count rate of the sample as recorded by the end-window proportional counter scaler divided by the rate of emission of charged particles by the sample.

They also show how dangerous it would be to estimate efficiencies for betas of less than 400 kev. I believe the efficiencies estimated by this method in this work have less than 10% error. A fuller discussion of all phases of the counter calibration, including tracer preparation, 4 π counting, pipette calibration, and sample preparation, may be found in Appendix B of this work. The standards used were Pm¹⁴⁷ (230-kev β^-), Na²² (540-kev β^+), Cs¹³⁷ (510 kev β^-), Na²⁴ (1.39-Mev β^-), P³² (1.71-Mev β^-), Y⁹⁰ (2.24-Mev β^-), and K⁴² (3.55-Mev β^-).²⁹

4. Gamma Counting.

A commercially available 100-channel pulse-height analyzer (PENCO) was used with two different Tl-activated NaI crystals; a 1-1/2 $\times$ 1-inch crystal having a 4-mil Be window, and a 3 $\times$ 3-inch crystal having a 25-mil aluminum window. Because much work has been published on NaI crystal efficiencies for gamma counting, it was necessary only to determine the geometrical factors of the various shelf positions used.

The geometrical factors of the shelf positions used were calculated by use of an Am²⁴¹ standard made to the author's specifications by Mr. Albert E. Salo of the Health Chemistry Division. The Am²⁴¹ was fairly evenly spread over a 1.9-cm-diameter circle on a 10-mil-thick aluminum plate. The rate of alpha-particle emission from the Am²⁴¹ was then measured in an alpha chamber of a "FAB" counter to 5×10^5 counts; the alpha-particle emission rate of a Bureau of Standards Am²⁴¹ sample was measured on the same FAB counter under the same geometry conditions immediately after the Health Chemistry sample. Since the disintegration rates of both samples were within 20% of each other, dead-time differences were negligible. Geometrical factors were equal, and the disintegration rate of the Am²⁴¹ sample prepared by Mr. Salo was found to be 187,000 alpha particles per minute or 67,200 60-kev gamma rays per minute.*

*Using the value 0.36 gamma rays per alpha emission from Reference 29.

For the 3X3-inch crystal an empirically determined correction of 3% was applied to the 60-kev photopeak for the aluminum-window absorption of 60-kev gamma radiation. Typical geometrical factors found after correction for the x-ray escape peak are 0.345 and 0.132 for shelf positions 1 and 2 of the 1-1/2X1-inch crystal, and 0.284 for shelf 1 of the 3X3-inch crystal. Geometrical factors were rechecked each time the pulse analyzer was used.

A set of gamma standards was prepared consisting of U^{235} , Bi^{207} , Na^{22} , Cs^{137} , and Co^{60} . When high-energy photopeaks, (>250 kev) were being measured, resolution was performed graphically by starting with the highest-energy photopeaks and subtracting off photopeaks and their associated Compton-electron and backscattered gamma contributions in order of decreasing energy, as described in Reference 30. The above-mentioned standards were used to obtain standard peak shapes for gamma rays of various energies. High-energy spectra were all measured on the 3X3-inch crystal. Efficiencies for the 3X3-inch crystal were taken from Reference 30.

Low-energy spectra (<250 kev) were measured on the 1-1/2X1-inch crystal. Background was subtracted from the photopeak on a linear basis; that is, a straight line was drawn through the non-Gaussian base of the photopeak and everything below this line was subtracted. When there was an overlapping of peaks, as in I^{121} - I^{123} (210 kev to 160 kev), both peaks were integrated together and the sums were plotted versus time as a decay curve. The counting rates of the two nuclides were then obtained by decay-curve analysis. All photopeaks measured were measured at various times to be sure that they had the expected half life. The values in the report by Hollander and Kalkstein were used for estimating photopeak efficiencies of the 1-1/2X1-inch crystal.³¹

III. EXPERIMENTAL RESULTS

A. Nuclides Observed

Nickel

The decay curves obtained by proportional counting always showed two components which were easily resolved graphically. The components represented half lives of 2.56 hours, corresponding to Ni^{65} , and 54 hours, corresponding to Ni^{66} in equilibrium with its 5.1-minute Cu^{66} daughter.

Arsenic

End-window proportional counting gave a decay curve that was resolved, through the use of the IBM 704 Frenic program (described in Appendix A), into four components having half lives of 17.5 days (As^{74}), 39 hours (As^{77}), 26.7 hours (As^{76}), and 90 minutes (As^{78}). Because the 26.7-hour component was less abundant than the 39-hour group, and because the half lives were not well separated, resolution of the 26.7-hour component had a large standard deviation associated with it. Therefore the 26.7-hour As^{76} gamma radiation was also observed on the 3x3-inch NaI crystal on the 100-channel pulse-height analyzer.

Bromine

In the long bombardments (1 to 2 hours) three components were observed in the beta decay curves, corresponding to the isotopes with half lives of 2.3 hours (Br^{83}), 4.4 hours ($\text{Br}^{80\text{m}}$), and 35.9 hours (Br^{82}). In a 3.00-minute bombardment components of 6.00 minutes ($\text{Br}^{84\text{m}}$), 32 minutes (Br^{84}), 2.3 hours (Br^{83}), 4.4 hours ($\text{Br}^{80\text{m}}$), and 35.9 hours (Br^{82}) were observed. All bromine decay curves were analyzed through the use of the IBM least-squares program. Although the $\text{Br}^{80}\beta^-$ activity of 18-minute half life was put into the guessed decay curve for the IBM least-squares program, the least-squares program would not converge promptly; rather, it showed the 18-minute activity at the end of the bombardment as zero counts per minute.

Strontium

The ratio of the yield of Sr^{89} to that of Sr^{90} was determined by use of the mass spectrometer. Strontium-89 and Sr^{91} yields were determined radiochemically by using the proportional counter. Two-component decay curves resulted which were easily resolved graphically. Twenty hours elapsed after the end of the irradiation before strontium oxalate was precipitated. Several yttrium hydroxide precipitations were performed just prior to the precipitation of the strontium to remove Y^{91} and Y^{92} . The 20-hour delay permitted 80% of the Sr^{91} to decay to Y^{91} , as well as removing the Sr^{92} - Y^{92} pair. Yttrium-91 has a 58-day half life and would appear as Sr^{89} (50.5-day half life) in the decay-curve analysis. A small correction was applied to the Sr^{89} disintegration rate for the Y^{91} that was present. This method was quite accurate; the correction was very small.

Yttrium

The proportional counter was used to count β^- particles from the decay of yttrium isotopes from four different bombardments of from 5.00 minutes to 2 hours. The yttrium fraction from the 5.00-minute bombardment showed β -decay components corresponding to half lives of about 16 minutes, 50 minutes (Y^{91m}), 3.6 hours (Y^{92}), 10.5 hours (Y^{93}), 64.2 hours (Y^{90}), and 57.5 days (Y^{91}). The 16-minute component was further resolvable into components of 10-minute (Y^{95}) and 19.5-minute (Y^{94}) half life. The decay curve from the 5.00-minute bombardment was analyzed through the use of the IBM 704 least-squares program, as were all the yttrium decay curves. The decay curves from the longer bombardments (20 minutes to 2 hours) yielded β -decay components with half lives of 19.5 minutes (Y^{94}), 50 minutes (Y^{91m}), 3.6 hours (Y^{92}), 10.5 hours (Y^{93}), 64.2 hours (Y^{90}), and 57.5 days (Y^{91}).

Zirconium

The β -decay curve resulting from decay of zirconium isotopes was resolved graphically into components corresponding to half lives of 17 hours (Zr^{97}) and 65 days (Zr^{95}). The 17-hour component was in equilibrium with the 72-minute Nb^{97} . The cross section for the production of Zr^{97} was also measured by using the 3X3-inch NaI crystal and pulse analyzer to observe the gamma radiation from decay of Zr^{97} .

Niobium

Least-squares analysis of the niobium β -decay curves yielded components corresponding to half lives of 52 minutes (Nb^{98}), 72 minutes (Nb^{97}), 23 hours (Nb^{96}), 84 hours ($\text{Nb}^{95\text{m}}$), and 35 days (Nb^{95}). The Nb^{95} decay was observed additionally on the 3X3-inch crystal by measuring the 760-kev gamma radiation.

Molybdenum

The β -decay rate of molybdenum was always measured on shelf 3 of the end-window proportional counter. The β decay always yielded a single component of 66- to 67-hour half life (Mo^{99}).

Silver

β -proportional counting was used to observe the decay of silver isotopes. IBM least-squares analysis yielded decay components having half lives of 21 minutes (Ag^{115}), 3.2 hours (Ag^{112}), 5.3 hours (Ag^{113}), and 7.5 days (Ag^{111}).

Iodine

Yields of iodine isotopes were determined by observing gamma radiation on the 3X3-inch and 1-1/2X1-inch NaI crystals, which were connected to the 100-channel pulse-height analyzer. Peaks were resolved from the total gamma spectra by using standards as explained in the section on gamma counting, and the integrated resolved photopeaks were plotted versus time to give decay curves. Resolution of these decay curves allowed quite certain identification of iodine isotopes (121, 123, 124, 125, 126, 128, 130) as well as allowing determination of their yields.

Cesium

The 3X3-inch NaI crystal and pulse analyzer were used to measure the 670-kev gamma radiation of Cs^{132} as well as the 380-kev gamma radiation of Cs^{129} .

Barium

The 1-1/2X1-inch NaI crystal with beryllium window and pulse-height analyzer were used to measure the cesium K x-rays from Ba^{131} as well as the Xenon K x-rays from decay of the Cs^{131} daughter.

B. Cross Sections and Results

The cross sections for all nuclides measured are listed in column 2 of the Table of Results. If the cross sections have been corrected so as to represent independent yields, these are listed in column 3. The other column headings are self-explanatory. The limits of errors quoted are generous, allowing usually 20% for the absolute errors between nuclides of different elements. The relative errors for isotopes of a fixed Z are much smaller. Where difficulty was encountered in resolving decay components, as in the gamma spectra resolution resulting from decay of iodine isotopes, or where the half life was open to some uncertainty, as with Y^{94} , a larger absolute error has been indicated. The results of the lower-energy bombardments are listed at the end of the table. The niobium isotopes from the target bombarded with 76-Mev C^{12} ions have no uncertainty for their yields listed because an unidentified contamination made decay-curve resolution very uncertain. The yttrium cross sections from the lower-energy bombardments have less than 20% errors indicated. The error is lower because these yttrium cross sections are being compared only with the yttrium cross sections from the 112-Mev bombardments and not with cross sections of other nuclides.

Equations for calculating cross sections are available in many references. All equations used in this work may be found in the discussion by Pappas.³²

Table of Results

Nuclide	Cross Section (mb)		Radiation detected	Assumed radiation abundance (particles per disintegration)	Number of determinations	Half life	Reference if other than 29.
	Measured	Corrected to independent yield					
With 112-Mev C^{12} ions							
Ni ⁶⁵	1.36±.14		β^-		5	2.56 hr	
Ni ⁶⁶	1.25±.13	1.0±.2	β^-	2	5	54.6 hr	
As ⁷⁴	.47±.10	.47±.10	β^+, β^-	0.62	1	17.5 d	
As ⁷⁶	2.8±1.4		β^-		1	26.4 hr	
	3.7±.8	3.7±.8	560-kev γ	0.36 γ /D		26.4 hr	
As ⁷⁷	10.0± 2		β^-	1	1	38.7 hr	
As ⁷⁸	5.6± 1		β^-	1	1	90 min	
Br ^{80m}	5.4±1.1	5.4±1.1	Br ⁸⁰ β^-, β^+	.95	3	4.4 hr	
Br ⁸²	12±2.4	12±2.4	β^-	1	3	35.9 hr	
Br ⁸³	17.5±3.5		β^-	1	3	2.30 hr	
Br ^{84m}	5.5±1.1	7±1.5	β^-	1	1	6.0 min	
Br ⁸⁴	2±.4		β^-	1	1	32 min	
Sr ⁸⁹	38.8±8.8		β^-	1	2	50.5d	
Sr ⁹⁰	33±5		(Mass spec ratio Sr^{89}/Sr^{90})		2		
Sr ⁹¹	20.4±3	18±3	β^-	1	2	9.67 hr	
Y ⁹⁰	16.3±3	16.3±3	β^-	1	4	64.2 hr	
Y ⁹¹	24±5	24±5	β^-	1	4	57.5 d	
Y ⁹²	25.7±5	25.7±5	β^-	1	4	3.60 hr	
Y ⁹³	24.5±5	20.5±5	β^-	1	4	10.4 hr	

Table of Results (continued)

Y ⁹⁴	12.8±3.6	11.0±3.6	β ⁻	1	4	16.5m, 20m	33
Y ⁹⁵	4.4±2.2	4.4±2.2	β ⁻	1	1	10.5m	
Zr ⁹⁵	34±7	30±7	β ⁻	1	1	65 d	
Zr ⁹⁷	11.6±2	11.6±2	β ⁻	2	1	17 hr	
	11.5±2		747+665-keV γ's	2	1		
Nb ⁹⁵	17±3	17±3	765-keV γ	1	2	35 d	
Nb ⁹⁶	29±6	29±6	β ⁻	1	2	23.3 hr	
Nb ⁹⁷	38±8	37±8	β ⁻	1	2	72.1 min	
Nb ⁹⁸	24±6	20±6	β ⁻	1	2	51.5 min	34
Mo ⁹⁹	45±10		β ⁻	1	6	66 hr	
Ag ¹¹¹	17±3	16.4±3	β ⁻	1	1	7.6 d	
Ag ¹¹²	10.8±2	10.5±2	β ⁻	1	1	3.20 hr	
Ag ¹¹³	7.6±1.5	6.9±1.5	β ⁻	1	1	5.3 hr	
Ag ¹¹⁵	.92±.20	.8±.2	β ⁻	1	1	21.1 min	
I ¹²¹	.04±.01	.04±.01	210-keV γ	0.92	2	2.0 hr	
I ¹²³	.44±.12	.42±.12	160-keV γ	0.83	2	13.0 hr	
I ¹²⁴	1.20±.30	1.20±.30	600-keV γ	0.64	2	4.2 d	35
I ¹²⁵	3.2±.6	3.2±.6	Te-K x-rays	1.39	1	60 d	
I ¹²⁶	3.1±.6	3.1±.6	380-keV γ	0.33	2	13.3 d	
I ¹²⁶	3.1		480-keV γ	0.05			
I ¹²⁶	3.2		650-keV γ	0.33			
I ¹²⁸	1.0±.3	1.0±.3	460-keV γ	0.17	2	25.0 min	
I ¹³⁰	≥.077±.02	≥.088±.02	410-keV γ	≤0.30	1	12.6 hr	
I ¹³⁰	≥.087		530-keV γ	≤1.00			

Table of Results (continued)

I ¹³⁰	≥.100		660-keV γ	≤0.90			
Cs ¹²⁷	.079±.02	.075±.02	mass spec, γ		1	6.3 hr	
Cs ¹²⁹	.6±.12	.54±.12	380-keV γ	≤1.00	1	30.7 hr	
			mass spec				
Cs ¹³¹	1.1±.22	1.1±.22	mass spec		1	9.6 d	
Cs ¹³²	.9±.18	.9±.18	670-keV γ	0.98	1	6.2 d	
			mass spec				
Cs ¹³⁵	.090±.018	.086±.018	mass spec		1	3×10 ⁶ Yr	
Ba ¹³¹	.16±.04	.15±.04	Cs&Xe K	0.94	1	11.5 d	35
			x-rays	0.73			
With 95-MeV C ¹² ions							
Y ⁹⁰	8.9±.9	8.9±.9	β ⁻	1	1	64.2 hr	
Y ⁹¹	17.5±2	17.5±2	β ⁻	1	1	57.5 d	
Y ⁹²	23±2	22±2	β ⁻	1	1	3.6 hr	
Y ⁹³	27±3		β ⁻	1	1	10.5 hr	
Y ⁹⁴	21±4		β ⁻	1	1	19.5 m	33
Nb ⁹⁵	9.4±2	9.4±2	765-keV γ	1	1	35 d	
Nb ⁹⁶	16.0±3	16±3	β ⁻	1	1	23 hr	
Nb ⁹⁷	32±6	32±6	β ⁻	1	1	72.5 m	
Nb ⁹⁸	15±3		β ⁻	1	1	51.5 m	34

Table of Results (continued)

With 76-Mev C^{12} ions

Y^{90}	1.05 ± 1	1.05 ± 1	β^-	1	1	64.2 hr
Y^{91}	3.00 ± 3	3.00 ± 3	β^-	1	1	57.5 d
Y^{92}	5.30 ± 5	4.9 ± 7	β^-	1	1	3.6 hr
Y^{93}	8.7 ± 9		β^-	1	1	10.5 hr
Y^{94}	8.4 ± 1.6		β^-	1	1	19.5 m
Nb^{95}	≤ 1.3		765-kev γ	1	1	35 d
Nb^{96}	9.7		β^-	1	1	23 hr
Nb^{97+98}	18		β^-	1	1	≈ 70 m

DISCUSSION

A. Correlation of Results

In most fission product studies the fission fragments are very neutron-rich. Thus it is simple to measure many chain yields but very difficult to measure a large number of independent yields. In this work the neutron-deficient properties of the probable compound nucleus (At^{209}) and the high excitation energy (95 Mev) made the reverse true. That is, the fission products varied from slightly neutron-rich to neutron-deficient, and it was possible to measure many independent yields. Because the products were close to stability it was very difficult to measure chain yields.* Both the yield-mass and charge-distribution curves depend upon a knowledge of chain yields. Therefore a method of converting independent yields to chain yields was needed.

It was assumed that the distribution of independent yields for any element fitted a fixed curve; that is, for any Z a graph of cross section for the production of the nuclide with mass number A versus A could be normalized to the same curve as for any other Z used. This assumption was tested by plotting relative cross sections for the production of cesium and yttrium isotopes versus mass number, as may be seen in Fig. 1. The yields of cesium isotopes were measured very accurately on the mass spectrometer; the yields of yttrium isotopes were carefully measured radiochemically. The yields of Y^{93} and Y^{94} are partial chain yields and should not (and do not) fall on the independent-yield curve. The points representing yields of yttrium isotopes were normalized so that the Y^{90} and Y^{91} yield points were on the smooth curve drawn through the cesium yield points. The consistency of the yttrium yield points with the curve drawn through the cesium yield points confirms that the shape of the curves for cesium and yttrium yields is the same within experimental error. It is unlikely that there is any great difference in this shape for other fission product yields.

*The term "chain yield" is used in a rather loose sense in this work; it is used to mean the cumulative isobaric yield, even though the "chain" may contain β^- , β^+ , EC, and stable isotopes.

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Fig. 1. Relative cross sections for the production of Y and Cs isotopes versus mass number.

For each of the elements arsenic, bromine, niobium, zirconium, silver, cesium, and iodine, two or more independent yields were measured; the curve determined for the yields of cesium and yttrium isotopes was normalized to these points (as may be seen by the solid curves of Fig. 2). By interpolating curves (Fig. 2) where experimental data were lacking, it was possible to estimate the independent yield of any nuclide. Thus it was also possible to estimate any chain yield in the mass region studied. In order to construct interpolated curves on Fig. 2 it was necessary to know only the mass number at which the maximum yield for a Z occurred ($A_{\max}$) and the cross section of the maximum yield ($\sigma_{\max}$). These numbers were both obtained from an interpolation of the solid curves (Fig. 2). A graph of the mass number at which the maximum yield occurred for each Z , plotted versus Z , allowed interpolation of the mass number for the maximum yield for unmeasured Z (Fig. 3), and a second graph of $\sigma_{\max}$ versus Z was used to interpolate the cross section at the maximum for any Z (Fig. 4).

Actually there are some finer points to justify. First, the cesium isotopes come from the fission of 0.5-mil gold foil; all other points are from the fission of 0.1-mil gold foil. The thicker foil degrades the beam more (by 11 Mev). Does it form a broader fission product distribution? Second, the measured yields of Cs^{127} , Cs^{129} , and Cs^{135} are not, strictly speaking, independent yields. How can these be corrected so as to represent independent yields?

The second question may be answered more easily. By studying the interpolated distribution for barium and xenon, and by using the formulas of radioactive growth and decay as discussed in Reference 32, one finds that the yield of Cs^{127} represents an independent yield and the yields of Cs^{129} and Cs^{135} may be corrected so as to represent independent yields with a correction of less than 10%. In order to answer the first question it was necessary to perform an experiment; three 0.1-mil gold foils, each sandwiched between 0.7-mil aluminum catcher foils, were bombarded with full-energy carbon ions. The average energy of the beam through the first and second gold foils was the same as that in the 0.5-mil gold foil; the total energy difference of the beam between the first and second foils was nearly the same as in the 0.5-mil gold foil.

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Fig. 2. The "standard shape" of Fig. 1 normalized to measured independent yields (solid lines). Several interpolated curves are shown as broken lines. The atomic number corresponding to each curve is written directly above the curve.

Fig. 3. The graph used to interpolate the mass number at which a Z should have its maximum yield.

Fig. 4. Curve used to determine the average number of charged particles emitted per fission event.

Yttrium and niobium fractions were extracted from each of the three foils by the previously mentioned radiochemical procedures. The yttrium isotopes from the first two foils were chosen for study because resolution of the yttrium decay curves gives more reliable data than the niobium decay curves. The yttrium cross sections are plotted versus mass number in Fig. 5. The broken curve is the curve resulting from summing the curves representing the yttrium yields from the first and second foils. It shows only a very slight broadening due to superposition of several excitation states. Corrections for both broadening and independent yields were applied to the cesium isotopes, and the whole process of constructing Fig. 2 was repeated. The cesium-yttrium yield curves (Fig. 5) and the family of curves shown in Fig. 2 actually are the corrected curves. Very little change resulted from the corrections.

The yield-mass curve was constructed by summing independent yields from Fig. 2 to get chain-yield values. Wherever possible mass numbers were chosen for which most of the chain was experimentally measured. The resulting yield-mass curve is shown in Fig. 6, where measured values are shown as triangles, and circles represent the total estimated chain yield.

B. Yield-Mass Curve

The yield-mass curve (Fig. 6) is symmetric about mass number 98 and is 27 mass numbers wide at half maximum. The yield-mass curves measured for the fission of bismuth with 22-Mev deuterons and bismuth with 190-Mev deuterons are shown in Fig. 7 as broken lines.^{8, 11} Fairhall's work (bismuth + 22-Mev d) shows a symmetric peak centered about mass number 103.5 and having a 17-unit width at half maximum. The work of Goeckermann and Perlman ($\text{Bi}^{209} + 190\text{-Mev d}$) shows a symmetric distribution centered about mass number 99. The comparison shows a narrower peak in the low-energy bombardment than in the higher-energy bombardments, which could be attributable to lower excitation in the fissioning nucleus, less variety of fissioning nuclei, or both. Comparison of the two high-energy peaks shows that the system bismuth + 190-Mev deuterons gives a fissioning nucleus with approximately the same average excitation energy as the system $\text{Au}^{197} + 112\text{-Mev C}^{12}$.

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Fig. 5. Yttrium excitation curves. For the production of yttrium isotopes from gold with carbon ions of 112 Mev, 95 Mev, and 76 Mev.

Fig. 6. Yield-mass curve for fission of Au^{197} with 112-Mev C^{12} ions.

MU - 20488

Fig. 7. Yield-mass curves for $\text{Bi}^{209} + 22\text{-Mev d}$ ---
 $\text{Bi}^{209} + 190\text{-Mev d}$ - · - · -
 $\text{Au}^{197} + 112\text{-Mev C}^{12}$ ———

This follows from the observation that both peaks are in nearly the same position as a function of mass number. The greater width of the peak resulting from the fission of bismuth with 190-Mev deuterons is most likely attributable to a greater spread of excitation energies about the average than for the $\text{Au}^{197} + \text{C}^{12}$ system, as well as to a greater variety of fissioning nuclei due to nucleon-nucleon cascade reactions. It is difficult at present to make any assessments, solely from the experimental data, of the effect of high angular momentum on the fission process.

The cross section for fission found in this work was 0.9 ± 0.3 barn, which may be compared to a predicted compound-nucleus cross section of 1.9 barns.¹³ Goeckermann and Perlman found a 0.2-barn cross section for fission of bismuth with 190-Mev deuterons, and Fairhall found a fission cross section of about 10^{-5} barn for bismuth + 22-Mev deuterons.^{11, 8}

C. Neutrons and Alpha Emission

How many nucleons were emitted in the fission act? An estimate of the total nucleon emission may be obtained by "folding" the yield-mass curve. Such folding is accomplished by summing mass numbers having the same yield, as read from Fig. 6. These mass numbers may be said to be yield-complementary. Masses of complementary fragments so defined summed to 196 amu, thirteen less than the 209 of the assumed compound nucleus. The fragments are truly complementary only if the number of postfission neutrons is the same for each fragment. This assumption should surely be good to within one nucleon. By folding the yield-charge curve (Fig. 4) in a similar manner, one finds the peak to occur at $Z=41.5$. Thus the sum of the charges of complementary fragments is 83, two less than the 85 of the compound nucleus, and two of the nucleons were emitted as charged particles. Thus it is found that in the average fission process there were 13 ± 1 nucleons emitted, two of them being charged particles, i. e. 11 ± 1 neutrons plus two protons, or 9 ± 1 neutrons plus an alpha particle.

Were the charged particles emitted as protons or as an alpha particle? An answer to this question may be obtained from energy-balance considerations. Consider Table II.

Table II

Comparison of two possible modes of nucleon evaporation for ${}_{85}\text{At}^{209}$ Excited to 89 Mev (c. m.)		
	Assumed final products (most probable)	
	${}_{41}\text{Nb}^{98} + {}_{42}\text{Mo}^{98} + 11\text{n} + 2\text{p}$	${}_{41}\text{Nb}^{98} + {}_{42}\text{Mo}^{98} + 9\text{n} + \alpha$
Observed K. E. of fission fragments (c. m.) ^a (Mev)	144	144
Energy available for n, p, or α K. E. or gamma emission (Mev)	8	36
Expected K. E. of neutrons (Mev)	22	18
Expected total K. E. of charged particles (Mev)	18	18
Total expected K. E. of neutrons plus charged particles (Mev)	40	36
Comparison with available energy	Inconsistent	Consistent
^a From Gordon. ³⁷		

Mass values for Table II were taken from the mass tabulation by Cameron, with the exception of mass-excess values for ${}^6\text{C}^{12}$, ${}^4\text{He}$, ${}^1_0\text{n}$, and ${}^1_1\text{p}$, which were taken from the Chart of the Nuclides.^{38,239} As may be seen from the tabulation, the 28-Mev binding energy of the alpha particle is necessary to give a balance between the energy available for nucleon kinetic energy and the energy required. One would expect 6 to 8 Mev of energy to be lost due to gamma emission. That this energy is apparently not available may be attributed to the one-neutron uncertainty (one less neutron lost would make an additional 8 Mev available) or to experimental error in the 144-Mev value used for kinetic energy of the fission fragments. Actually alpha-particle emission from the system gold + 160-Mev ${}^{16}_0\text{n}$ ions has been seen by Anderson, Quinton, and Knox.⁴⁰ Preliminary results indicate that the average KE of the emitted alpha particles is 18 Mev in the center-of-mass system.⁴⁰

Are the neutrons emitted before or after fission? Can any estimate be made from radiochemical results? A study of Fig. 8 indicates that it can. Two modes of de-excitation are shown in Fig. 8: evaporation of six neutrons and an alpha particle followed by fission (1); and emission of an alpha particle followed by fission, with all neutrons emitted after fission (2). Both modes give the same final result within the consistency of the mass tables. The important difference is that the average neutron binding energy is greater than 8.5 Mev per neutron for prefission neutrons (1), and is less than 6.3 Mev for postfission neutron evaporation (2). It is likely that in either case the neutrons have about 2 Mev of kinetic energy; thus as seen in Table III (column 3) it should require approx 10.5 Mev to remove a neutron before fission, but only approx 8.3 Mev to remove a neutron from a fission fragment. Thus it is necessary to experimentally determine the energy required to remove a neutron from the system $\text{Au}^{197} + \text{C}^{12}$. The yttrium yield excitation function shown in Fig. 5 represents such a determination. When the energy of the incident C^{12} ion is increased from 76 Mev to 112 Mev, the most probable neutron number in the yttrium fraction is decreased by $1.3 \pm .2$ neutrons. In addition, Gordon's results imply that the total fragment KE was increased by 3 Mev as the bombarding energy was increased from 76 Mev to 112 Mev.³⁷

Fig. 8. Comparison of prefission and postfission neutron evaporation thermodynamics.

Table III

Comparison of neutron-evaporation systematics

	<u>Prefission</u>	<u>Postfission</u>
1. Average neutron binding energy (Mev)	8.5	6.3
2. Expected average KE per neutron (Mev)	2	2
3. Total energy required to remove a neutron (Mev)	10.5	8.3
4. Experimentally found energy required to remove a neutron (Mev)	11.9±1.6	
5. Consistency with experimental value	Consistent	Inconsistent

Assuming the same neutron change in the complementary fragment (ruthenium), it is clear that 31 Mev (c.m.) additional excitation energy caused $2.6 \pm .4$ additional neutrons to be emitted. As may be seen in Table III (row 4), this implies an average of 11.9 ± 1.6 Mev required to remove each of the last three neutrons from the system $\text{Au}^{197} + 112\text{-Mev } \text{C}^{12}$ ions. This value is consistent with prefission neutron evaporation.

The assumptions being made in the above argument are

(a) gamma-emission competition remains constant as a function of bombarding energy from 76 Mev to 112 Mev, (b) nucleon emission is considered to take place before beta decay, (c) the ruthenium fragment is assumed to show the same change in neutron number versus energy as the yttrium fragment, (d) the yttrium fraction is assumed representative of the most probable fission products, (e) a compound nucleus is formed for nuclei which eventually fission, and (f) Cameron's masses are assumed to give neutron binding energies accurate to ± 0.5 Mev in the regions used. The foregoing argument serves more to show how radiochemistry coupled with thermodynamic data may be used to gain an insight into neutron emission kinetics than as an "airtight" argument for prefission neutron evaporation. One should have data on the entire yield-mass curve (and therefore ν) in order to make such a calculation.

D. Charge Distribution

The two most useful empirical rules for correlating independent yields are the Equal Charge Displacement (ECD) and Constant Charge Ratio (CCR) rules. The ECD rule as originally suggested by Glendenin, Coryell, and Edwards predicts that the most probable chain length for a light fission fragment is equal to that for its complementary heavy fragment; that is $(Z_A - Z_p)_{\text{light}} = (Z_A - Z_p)_{\text{heavy}}$, where Z_A is the most stable charge for the mass number "A" and Z_p is the most probable charge for the same mass A.⁴¹ Neither Z_A nor Z_p is required to be integral. Glendenin et al. used an empirically determined continuous Z_A function and considered the fission fragments after neutron evaporation.

Several modifications of ECD exist; Pappas considered the fission fragments before neutron emission and used a Z_A function which was discontinuous at shell edges.⁴² Grummitt and Milton used a Z_A function which showed no

sharp discontinuities at shell edges.⁴³

The ECD rule has had its greatest success in the correlation of thermal-neutron and low-energy fission data. Examples are found in the fission of U^{235} , U^{238} , and Pu^{239} with thermal neutrons, Th^{232} with pile neutrons, Cm^{242} spontaneous fission, and U^{235} with 14-Mev neutrons.^{43, 44, 45} The Grummitt and Milton and Glendenin Z_A functions were used in the majority of the above-mentioned works.

Goeckermann and Perlman's fission study of bismuth with 190-Mev deuterons fits the CCR rule better than the ECD rule. The CCR rule predicts the same neutron-to-proton ratio in the fission products as in the fissioning nucleus. The CCR rule seems to be superior to the ECD rule in high-energy fission. In his very careful mass-spectrometric study of fission yields from U^{235} and U^{238} plus 45.7-Mev and 24-Mev helium ions, Chu concluded that perhaps something intermediate between a non-shell-affected ECD and a CCR rule might best explain the experimental results.⁴⁶ Del Marmol studied fission of U^{235} with 13.6-Mev deuterons and 23.5-Mev helium ions, and concluded that the ECD rule gave a "satisfactory" interpretation of the fractional chain yields for the excitation studied.⁴⁷ Alexander and Coryell investigated fission of U^{238} and Th^{232} with 13.6-Mev deuterons, and concluded that the ECD rule was valid.⁴⁸ In a very careful radiochemical study of fission of uranium isotopes with 25- to 40-Mev helium ions, Colby and Cobble have found that the CCR rule gives excellent correlation for nuclides away from the 82-neutron shell.⁴⁹

A third approach to the charge-distribution problem, an approach having a theoretical basis, is that by Present.⁵⁰ Present's theory is based on classical electrostatics; for example, when a spherical charged conductor is distorted, there results a redistribution of charge at the surface to effect the minimum potential energy for the charge on the body. It is this tendency toward redistribution of charge with change of shape that is the basis of Present's theory.

As the excited nucleus goes through its various modes of distortion, it is possible for the charge distribution to be constantly changing also, showing a tendency toward minimizing electrostatic potential.

True equilibrium in the sense of classical electrostatics is never attained, owing to the competition of nuclear forces which tend to keep a uniform proton distribution within the nucleus; nevertheless the competition between the electrostatic force and the nuclear restoring force would be expected to attain a position of minimum potential energy, Such a tendency would give a light fission fragment more than its share of charge, a heavy fragment less.

Making the simplifying assumption that the shape of the deformed nucleus at the time of scission may be approximated by two tangent spheres (as was done by Present), Swiatecki has worked out the equation for estimating the distribution of charge in fission:⁵¹

$$\frac{\frac{Z_1}{A_1}}{\frac{Z}{A}} = 1 + \frac{A^{2/3}}{8 \text{ COEFF}} \frac{e^2}{r_0} F(\lambda), \quad (1)$$

where Z_1/A_1 is the charge-to-mass ratio of a fission fragment, Z/A is the charge-to-mass ratio of the fissioning nucleus, A , e , and r_0 have the customary definitions,² COEFF refers to the coefficient of the $\frac{(N-Z)^2}{A}$ term (binding energy) in the nuclear mass equation (25.4 mmu) $\lambda = R_1/R_2$, or the ratio of the radii of the two "spheres" or fission fragments.

$$\text{and } F(\lambda) = \frac{(1-\lambda) [6/5(1+\lambda)^3 - 1 - \lambda - \lambda^2]}{(1+\lambda)(1+\lambda)^{5/3}}.$$

After numerical substitution, the charge-distribution equation simplifies to

$$\frac{\frac{Z_1}{A_1}}{\frac{Z}{A}} = 1 + .00625 A^{2/3} F(\lambda). \quad (2)$$

Swiatecki has also formulated the problem in more general terms. He has expressed the potential energy as a function of the displacement of the most probable charge from the most stable charge for an effective fragment separation D as

$$E = \frac{1}{2} B_{A_H} (Z_{P_H} - Z_{A_H})^2 + \frac{1}{2} B_{A_L} (Z_{P_L} - Z_{A_L})^2 + \frac{e^2}{D} Z_{P_H} Z_{P_L} + \text{constants}. \quad (3)$$

The B_A of Eq.(3) are parameters representing the curvature of the mass parabolas. The subscripts L and H refer to the light and heavy fragments respectively. Minimizing the potential energy with respect to Z_{PH} gives

$$\frac{\partial E}{\partial Z_{PH}} = B_{A_H}(Z_{PH} - Z_{A_H}) - B_{A_L}(Z_{PL} - Z_{A_L}) + \frac{e^2}{D}(Z_{PL} - Z_{PH}) = 0. \quad (4)$$

It is of interest to note that this calculation was done by Wigner and Way, using the first two terms on the right-hand side of Eq. (2), and apparently the identical calculation was done by Way.⁵² Making the assumption that $B_A = \frac{8C}{A}$, where C represents the coefficient of the $\frac{(N-Z)^2}{A}$ term of the mass equation (25.4 mmu),⁵³ simplifies Eq. (4) to

$$\frac{203.2}{A_H}(Z_{PH} - Z_{A_H}) - \frac{203.2}{A_L}(Z_{PL} - Z_{A_L}) + \frac{e^2}{D}(Z_{PL} - Z_{PH}) = 0, \quad (5)$$

where D is to be expressed in Fermis.⁵¹ Equation (5) coupled with the relation $Z_{PH} + Z_{PL} = Z_{\text{fissioning nucleus}}$ permits calculation of Z_p 's. If a non-shell-affected Z_A function is used (such as the function obtained by using Green's values), Eq. (5) gives essentially the same results as Eq.(2).⁵² If a shell-affected Z_A value is used in Eq. (5) (such as the Z_A 's reported by Grummitt and Milton), the resulting Z_p function shows shell effects. The Z_p functions calculated from Eq. (5) for several fissioning systems will be seen in Fig. 10, and are discussed on p. 45.

Experimental discrimination between various charge-distribution rules requires empirically determined Z_p 's. However, Z_p 's are difficult to determine empirically, since it is hard to measure a sufficient number of independent yields for a given target-projectile system. As previously noted, the system gold plus 112-Mev carbon ions gives a highly excited compound nucleus (At^{209}), which is neutron-deficient and gives many fission products near the beta stability line. The yields measured were corrected so as to represent independent yields (as previously discussed), and the independent yields were converted to fractional chain yields by dividing them by their total chain yield as read from the smooth yield-mass curve (Fig. 6).

The large number of independent yields obtained allowed determination of Z_p 's empirically. Two methods were used. First, by reading along a fixed mass number in Fig. 2, values of the cross sections of independent yields of a given mass number were read and plotted versus Z ; a smooth curve was drawn through these points, the peak being taken as Z_p . It is believed that the empirical Z_p so obtained is easily accurate to ± 0.2 charge unit. The fractional chain yields were next plotted versus $Z - Z_p$ to see if they would determine a unique charge-distribution curve. That this is true may be seen from Fig. 9, in which a smooth charge-distribution curve has been drawn through the experimental points. The curve obtained integrates (graphically) to 0.92, which may be considered unity within experimental error. The charge-distribution curve obtained allows calculation of a second set of empirical Z_p 's by the method due to Wahl.⁴⁴ It is assumed that all independent yields should fall on the curve; they are indeed forced onto the curve and the $Z - Z_p$ value so determined gives an empirical Z_p .

The Z_p 's determined by both methods described above have been plotted versus mass number (A) in Fig. 10. It was desired to plot the Z_p 's in such a way as to show any fine structure. Since Z varies approximately as $0.4 A$ in this region, the Z_p 's were plotted as $Z_p - 0.4 A$ versus A , which clearly shows any detail in the distribution. Included for comparison in Fig. 10 are the Z_p 's predicted by the ECD rule, the CCR rule, and the Minimum Potential Energy theory (MPE) of Present as modified by Swiatecki. Both shell-affected and non-shell-affected Z_p 's have been calculated by the MPE theory, as may be seen in Fig. 10. In all cases a "fissioning nucleus" of Bi^{196} was assumed. For comparison the ECD, CCR, MPE, and empirical Z_p 's for U^{235} + thermal neutrons (Wahl, Chu, and Wolfsberg), and U^{235} + 45.7-Mev helium ions (Chu) are shown below the gold-plus-carbon data of Fig. 10.^{44, 46, 54}

The MPE theory gives the best prediction of the empirical Z_p 's for the system gold plus carbon ions, as Fig. 10 clearly shows. It is not possible to decide whether the shell-affected or non-shell-affected MPE prediction gives the empirical Z_p 's better, since there is an uncertainty of ± 0.2 unit associated with each empirical point. For U^{235} + 45.7-Mev helium ions, the non-shell-affected MPE prediction appears to be best. For U^{235} plus thermal neutrons, the non-shell-affected MPE

MU-20522

Fig. 9. Charge-distribution curve for fission of $\text{Au}^{197} + 11.2\text{-MeV C}^{12}$ ions.

MU-20489

Fig. 10. Comparison of empirical Z_p 's and various predicted Z_p 's for the systems of $\text{Au}^{197} + 112\text{-Mev } \text{C}^{12}_p$, $\text{U}^{235} + 45.7\text{-Mev } \text{He}^4$, and $\text{U}^{235} + \text{thermal neutrons}$.

- Empirical points found by the method due to Wahl.
- △ Empirical points from Fig. 2.
- CCR
- ECD
- Liquid-drop (non-shell-affected) MPE.
- o o o and o--o--o MPE using Grummit and Milton Z_A and liquid-drop B_A .

prediction shows the proper trend but fails to reproduce the 82-neutron shell effect. The shell effect is predicted well by the ECD rule, or as may be seen from Fig. 10, by the MPE theory using the same shell-affected Z_A 's as were used in calculating Z_p 's by the ECD rule.

It is of interest that the shell-affected Z_p 's calculated by the MPE theory for thermal neutron fission of uranium agree so well with those calculated by the ECD rule. The ECD rule is thus given a degree of physical significance, as pointed out by Halpern.⁵⁵ It is also interesting that the shell-affected MPE predictions for gold plus carbon ions do not agree with the ECD rule predictions. Because of its physical significance, the MPE theory may eventually allow calculation of such parameters as effective separation of fragments at time of scission or nuclear shape at time of scission. Such calculations would require accurately determined empirical Z_p 's. The MPE theory should not be overlooked when fission data are being correlated.

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APPENDIX A. THE LEAST-SQUARES PROGRAM

The least-squares program used in this work was FRENIC, a Los Alamos Scientific Laboratory program written by R. H. Moore and R. K. Ziegler. FRENIC performs a least-squares analysis for radioactive decay curves of up to 10 components. The function minimized is

$$\sum_{i=1}^n W_i (y_i - \sum_{j=1}^k a_j e^{-\lambda_j t_i})^2, \quad (A1)$$

where W_i is the weighting factor associated with each data point y_i , y_i represents the counting rate at t_i minutes after the end of bombardment, and a_j and λ_j represent the values of the intercept and decay constant of the j th component. It is necessary to put in guesses for the initial activity of each component (a_{j0}) and the decay constants (λ_{j0}), because of the iterative nature of the program. That is, a Taylor expansion is taken about a_{j0} and λ_{j0} and all terms higher than the first-order terms in $(a_{j0} - \Delta a_{j0})$ and $(\lambda_{j0} - \Delta \lambda_{j0})$ are discarded before the normal equations are solved. Thus the program starts with a set of guessed values a_{j0} and λ_{j0} , computes the weighted sum of the squares of the deviations according to Eq. (A1), applies a first-order correction to a_{j0} and λ_{j0} , and repeats the process until convergence is obtained. Convergence has been defined in the FRENIC program as the set of values a_{j0} , for which $\frac{\Delta a_j}{a_j} < 10^{-4}$, and $\frac{\Delta \lambda_j}{\lambda_j} < 10^{-4}$. Any or all of the a_{j0} or λ_{j0} may be held constant during the computation. Any number of data point cards up to $n = 500$ may be used. After convergence, the output consists of a listing of the parameter guesstimates (a_{j0} , λ_{j0}), final least-squares answers, the standard deviation associated with each parameter, a listing of each W_i , y_i , t_i , y_i calc, and Δy_i , the number of iterations performed, and the weighted variance,

$$S^2 = \frac{1}{n-k} \sum_{i=1}^n W_i (y_{\text{obs}} - y_{\text{calc}})^2, \quad (A2)$$

where $y_{\text{calc}} = \sum_{j=1}^k a_j e^{-\lambda_j t_i}$. The weighted variance is useful when

there is some doubt as to the number of components. The number of components for which S^2 is a minimum may be taken as the best number.

The weighting factor W_i originally had three choices available; $W_i = 1$, $W_i = \frac{1}{y_i}$, or W_i was arbitrarily chosen and punched individually on each data point card. I have added the option $W_i = (\frac{1}{y_i})^2$ to the program. This option is used if all counts are taken to nearly equal statistics. In almost all of my work this was the case and the $(\frac{1}{y_i})^2$ weighting factor was used for the least-squares analysis. For a further discussion of this program one should consult Reference 56.

APPENDIX B. PROPORTIONAL-COUNTER CALIBRATION

1. Pipet Calibration

Micropipets of one to 100 microliters were mercury-calibrated. The pipets were filled to the mark with mercury and emptied into a tared weighing bottle. The process was repeated until three consecutive weighings agreed to $\pm 0.3\%$. The 1- λ pipet was only 0.97 λ , whereas all other pipets (4 λ , 10 λ , 40 λ , 100 λ) were within $\pm 0.3\%$ of their rated volumes. The pipets so calibrated are calibrated "to contain."

2. 4 π Sample Plates and 4 π Counting

Ten-mil-thick aluminum discs, each with a 1/2-in. or 3/4-in. diameter hole in its center, were used for making 4 π sample plates. The actual films used were VYNS* films (a polyvinyl acetate copolymer) made as described by Pate and Jaffe.⁵⁷ After a film was dry a layer of gold was evaporated onto it to give a very slight bluish tint. The gold is necessary to prevent charge build-up on the sample plate. The plates prepared as described above and in the literature were of the order of 10 $\mu\text{g}/\text{cm}^2$ thick.

The 4 π counter used was an Oak Ridge model as modified by Dr. Herman P. Robinson of Lawrence Radiation Laboratory. The plateaus showed no appreciable slope from 3200 v to 3900 v ($< 0.1\%$ per 100 v). A uranium standard always gave the same count rate within statistics ($\pm 0.7\%$). The counter was operated with methane gas as a proportional counter.

3. Tracers

The tracers used in this work were Pm^{147} , Cs^{137} , Na^{22} , Na^{24} , Au^{198} , P^{32} , Y^{90} , and K^{42} . The Na^{22} , Na^{24} , and Au^{198} samples were obtained commercially with their disintegration rates guaranteed to $\pm 3\%$. After correction for the factor-of-10 error made by the issuing

* I am grateful to Bakelite Co. for the VYNS resin furnished free of cost,

company, the Na^{22} 4π counted to within 0.5% of the value stated by the company. The Na^{24} was assumed to be correct; the gold solution could not be checked by 4π counting. This was because there was approximately 10 times as much residue in the solution as stated by the supplier. The Au^{198} results were discarded because of the uncertainties in disintegration rate and exchange problems arising from the cyanide complex form in which the gold "tracer" came. A commercial P^{32} solution was tried. The 4π counting results were in excellent agreement with the manufacturer's stated value--after the manufacturer had sent in a correction sheet on the original value. Actually P^{32} obtained from Donner Laboratory was used in this work. Approximately 0.2 ml of $\text{H}_3\text{P}^{32}\text{O}_4$ was diluted to 25 ml with conductivity water. Three 10- λ aliquots were 4π counted. The disintegration rate was found to be $98,600 \pm 600$ dpm per 10 λ . The Pm^{147} solution was made by diluting 4 λ of Pm^{147} solution from Oak Ridge to 500 ml with conductivity water. The activity found was $60,000 \pm 600$ dpm per 10 λ . Cesium-137 was obtained from Health Chemistry in a pure tracer form; the stock solution was diluted 1:3 with conductivity water, giving a tracer solution of $58,600 \pm 300$ dpm per 10 λ . The Y^{90} and K^{42} tracers were made by irradiations of yttrium oxide (99.9%) and potassium carbonate (ACS specs.) in the Livermore Pool Type Reactor (LPTR). A few mg of each were irradiated for 10 minutes. The irradiated crystals were dissolved in a few drops of "ACS spec." 12 M HCl and diluted to 5 ml in their respective volumetric flasks. Three 4π plates were made of each tracer solution and the decay rate was followed for several half lives on the 4π counter. Both tracers had the proper half life and were assumed to be pure.

4. Standard Samples

a. Promethium-147

To each of six centrifuge cones 10 λ of Pm^{147} tracer was added. Lanthanum carrier was added in varying amounts to each centrifuge cone so that the final precipitates would vary from 5 to 70 mg. Oxalic acid was added to precipitate lanthanum oxalate and coprecipitate promethium oxalate. The precipitates were filtered with the standard chimney previously described, dried, and mounted for radiation counting.

When counting was completed the samples were sent to the analytical laboratory for analysis. The percent yield determined for the lanthanum oxalate was assumed to be the yield of Pm^{147} . Since the lanthanum oxalate yields were nearly 100%, and since the Pm^{147} was used to get an upper limit only, this assumption is certainly safe.

b. Sodium-22

Aliquots of Na^{22} , 10 λ each, were added to each of six centrifuge cones. Varying amounts of warm saturated Na_2CO_3 were added and cooled to precipitate sodium carbonate. The absolute disintegration rates of the samples were determined by comparing the count rate of the samples with those of 4π plates on a "gross gamma" counter. The "gross gamma" counter consisted of a 1-1/2x1-in. NaI crystal and photomultiplier, amplifier, and scaler. The discriminator was set to detect all gamma radiation above 100 kev. The 4π plates of known disintegration rate were counted under the same low-geometry conditions as the samples, with 700 mg/cm² of aluminum absorber. The disintegration rate of the samples was equal to the gamma counting rate of sample $\times$ disintegration rate of 4π plate $\div$ gamma counting rate of 4π plate.

c. Cesium-137

Cesium-137 was precipitated as the chloroplatinate. Yields were determined stoichiometrically and by gross gamma counting against 4π plates. Agreement between the two methods was within 1%.

d. Sodium-24

Forty λ of Na^{24} tracer was added to each of five centrifuge cones. The approximate amount of sodium chloride carrier to give the desired range of sample thicknesses was added to each centrifuge cone. The solutions were evaporated to dryness, slurried in acetone, and filtered. The samples were sent to the analytical laboratory for yield determination.

e. Phosphorus-32

The standard magnesium ammonium phosphate precipitation was performed for P^{32} samples. Forty λ of tracer was used for each sample. The yields were determined by the analytical laboratory by igniting the magnesium ammonium phosphate to the pyrophosphate.

f. Yttrium-90

Forty λ of Y^{90} tracer was added to each of five centrifuge cones. Yttrium carrier in amounts sufficient to give precipitates of 2 to 24 mg/cm^2 was added and yttrium was precipitated as the oxalate. The yields were determined by ignition to the oxide, Y_2O_3 . The analyses were performed by the analytical laboratory.

g. Potassium-42

Forty λ of K^{42} tracer was added to each of six centrifuge cones. Enough carrier was added to each centrifuge cone to give precipitates of 3 to 28 mg/cm^2 . The potassium was precipitated as potassium chloroplatinate and the yield was determined stoichiometrically, analytically, and by gross gamma counting. All three methods agreed to within 1%. Another set of K^{42} samples was mounted as $K_2NaCo(NO_2)_6$. Their disintegration rate was determined by gross gamma counting. Samples of 1 to 37 mg/cm^2 were made as the $K_2NaCo(NO_2)_6$ precipitate.

5. Results and Discussion

The results are shown in the following graphs, Figs. 11-14. The first series are counting-efficiency curves as a function of average β energy for samples 8 mg/cm^2 thick. Curves in the second series represent the efficiency versus sample thickness, Figs. 15-21. The efficiency-versus-energy curves were constructed by taking a "cut" through the efficiency-versus-thickness curves at the arbitrary thickness of 8 mg/cm^2 . The efficiency-energy curves may be constructed for any thickness in the region studied (0 to 25 mg/cm^2). The efficiencies listed for zero thickness correspond to tracer spread over double-sided Scotch tape occupying the position the sample ordinarily was in. Further use of these curves for estimating β -counting efficiencies is discussed in the paper by Bayhurst and Prestwood²⁸ and need not be discussed further here.

It is of interest to note that in the potassium thickness curves the efficiency values versus sample thickness for the cobaltinitrite and chloroplatinate precipitates agree within experimental error; in earlier work done with I^{131} in silver iodide, thallous iodide, and silver chloride it was found that for a given sample thickness there was no more than 5% difference in counting efficiencies.

These results are consistent with those found by Bayhurst and Prestwood for Tl^{204} as thallous chromate, thallous iodide and thallous chloroplatinate. That is, there does not appear to be a dependence upon the average Z of the precipitate. All the curves run in this calibration were normalized to a uranium standard. The normalization was always less than 1%.

MU-20491

Fig. 11. Beta-counting efficiency vs average β energy for Shelf 1 of end-window proportional counter.

MU-20492

Fig. 12. Beta-counting efficiency vs average β energy for Shelf 3 of end-window proportional counter.

MU-20493

Fig. 13. Beta-counting efficiency vs average β energy for Shelf 5 of end-window proportional counter.

MU - 20494

Fig. 14. Beta-counting efficiency vs average β energy for Shelf 8 of end-window proportional counter.

MU-20495

Fig. 15. Beta-counting efficiency vs sample thickness for Pm^{147} .

MU-20496

Fig. 16. Beta-counting efficiency vs sample thickness for 520-keV β of Cs^{137} .

MU-20497

Fig. 17. Beta-counting efficiency vs sample thickness for Na²².

MU-20498

Fig. 18. Beta-counting efficiency vs sample thickness for Na^{24} .

MU-20499

Fig. 19. Beta-counting efficiency vs sample thickness for P^{32} .

MU-20500

Fig. 20. Beta-counting efficiency vs sample thickness for Y⁹⁰.

MU-20501

Fig. 21. Beta-counting efficiency vs sample thickness for K^{42} .

APPENDIX C. APPARATUS

The following pages show the target holders, Figs. 22 and 23; filtering apparatus, Fig. 24; and sample card, Fig. 25.

I Target holder

ZN-2496

Fig. 22. Helium-cooled Hilac target holder.

ZN-2497

Fig. 23. Target holders.

ZN-2498

Fig. 24. Filtering apparatus.

MU-20490

Fig. 25. Sample-mounting card.

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