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THE REACTIONS OF TANTALUM WITH 5.7-Bev PROTONS

James Robb Grover
(Thesis)

September 1957

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THE REACTIONS OF TANTALUM WITH 5.7-Bev PROTONS

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September 1957

ABSTRACT

Tantalum was bombarded with 5.7-Bev protons in the Bevatron at the University of California. Product cross sections were measured, by radiochemical techniques, for about 80 radioactive isotopes from mass number 7 (beryllium) to mass number 181 (tungsten). The results are very different from those found at 0.34 Bev; the cross sections in the mass region \( A = 7 \) to 150 have increased by several orders of magnitude while those in the mass region \( A = 160 \) to 175 have decreased appreciably. There is no clear fission peak observable. The bulk of the observed products are neutron-deficient isotopes. On the other hand, these results are similar to the observations made by workers at Brookhaven National Laboratory in the bombardment of lead with 3-Bev protons. The experimental data were treated empirically, in order to establish a semi-analytic "systematics" of cumulative cross sections, for the purpose of estimating (1) unmeasured yields and total isobaric cross sections, and (2) the course of the isobaric independent yield maxima with respect to the beta stability line. The observed distribution of products is discussed with respect to the current ideas concerning high-energy nuclear reactions.
THE REACTIONS OF TANTALUM WITH 5.7-Bev PROTONS

I. INTRODUCTION

The investigation of the interactions of nuclei with particles having energies of about 0.1 Bev or more is becoming an increasingly important approach to the problems of nuclear structure. Such studies have indicated that at these energies the nucleon-nucleon cross sections become small enough that the bombarding particle no longer interacts with the nucleus strictly as if it were an integral unit, but rather as if it were a relatively open cluster of individual nucleons. One hopes, on this basis, that as the body of experimental data grows it may become possible to infer a more detailed picture of the distribution of nucleons within the nucleus than is now possible, or, as the case may be, to test the very idea of nucleon "location." For example, the recent measurements of (p,pn) cross sections at Bev energies by Markowitz and Benioff appear to require some such factors in their interpretation.

Early work in this field (prior to 1947) was confined to the study of stars appearing in nuclear emulsions and cloud chambers exposed to cosmic rays. Despite the facts that the bombarding energies were not clearly defined, the target was necessarily almost restricted to nuclear emulsions, and the role of neutrons in these reactions remained a mystery, many valuable conclusions of a qualitative nature were drawn. The idea of collisions between the bombarding nucleons and individual nucleons within the nucleus, leading to a group of emergent knock-on particles, was proposed to account for some of the observed features of nuclear stars. The early work with cosmic rays is reviewed in an article by Rochester and Rosser.

When machines capable of producing intense beams of particles of well-defined energy in this energy range came into being (about 1946), radiochemical identification and measurement of the reaction products became possible, along with other experiments allowing more quantitative measurement of the phenomena associated with such reactions. In particular, the measurement of the spectrum and angular distribution of emitted
neutrons, protons, alpha particles, etc., became possible for a wide variety of targets and bombarding energies.\textsuperscript{6-9}

One of the important discoveries of the first radiochemical workers was that the reaction products were spread over a wide range of mass and atomic numbers below the target. This was in disagreement with the predictions of the compound-nucleus theory,\textsuperscript{10} but consistent with the wide distribution of star sizes previously observed in nuclear emulsions. At the same time transmission experiments were found to give total inelastic cross sections decidedly smaller than the expected geometric cross sections,\textsuperscript{11} for various nuclei, indicating that nuclei are partially transparent to bombarding nucleons of these energies. Such observations led Serber\textsuperscript{12} to propose the view, already mentioned in the preceding paragraphs, that the bombarding nucleon interacts with individual nucleons within the nucleus, having a mean free path in nuclear matter comparable with the nuclear radius, and that the consequent wide statistical variation in the resulting knock-on cascades and deposition energies is reflected in the wide distribution of products.

More quantitative theoretical work soon followed; Goldberger\textsuperscript{13} approximated the cascade event by a series of Monte Carlo calculations, and Le Couteur\textsuperscript{14} and Yamaguchi\textsuperscript{15} treated the evaporation phase analytically in terms of an excited Fermi gas, with results in fair agreement with observations in nuclear emulsions. An account of the progress up to about 1952, with special attention to radiochemical work, is given in a review by Templeton.\textsuperscript{16}

Radiochemical studies in this energy range have been appearing with increasing frequency, so that the papers now available include a wide range of target nuclei and bombarding energies. A good list of the published results for target nuclei of $A = 75$ and below is given by Rudstam,\textsuperscript{17} and another list, for target nuclei in which fission has been studied at these energies, is given by Shudde.\textsuperscript{18} The following list includes studies for which the target $A$ varies from 89 to 209, and for which some work in the spallation (cascade-evaporation) region has been done.
<table>
<thead>
<tr>
<th>Bomb. Energy (Mev)</th>
<th>Particle</th>
<th>Target Element</th>
<th>Worker</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 to 50</td>
<td>negative muons</td>
<td>iodine</td>
<td>Winsberg(^{19})</td>
</tr>
<tr>
<td></td>
<td>negative pions</td>
<td></td>
<td></td>
</tr>
<tr>
<td>240</td>
<td>protons</td>
<td>yttrium</td>
<td>Caretto and Wiig(^{20})</td>
</tr>
<tr>
<td>240</td>
<td>protons</td>
<td>iodine</td>
<td>Dropsky(^{21})</td>
</tr>
<tr>
<td>60 to 240</td>
<td>protons</td>
<td>cesium</td>
<td>Fink and Wiig(^{22,23})</td>
</tr>
<tr>
<td>340</td>
<td>protons</td>
<td>silver</td>
<td>Kofstad(^{24})</td>
</tr>
<tr>
<td>340</td>
<td>protons</td>
<td>tantalum</td>
<td>Nervik and Seaborg(^{25})</td>
</tr>
<tr>
<td>50 to 190</td>
<td>deuterons</td>
<td>antimony</td>
<td>Lindner and Perlman(^{26})</td>
</tr>
<tr>
<td>380</td>
<td>helium ions</td>
<td></td>
<td></td>
</tr>
<tr>
<td>375 and 450</td>
<td>protons</td>
<td>bismuth</td>
<td>Bennett(^{27})</td>
</tr>
<tr>
<td>450</td>
<td>protons</td>
<td>bismuth</td>
<td>Sugarman et al.(^{28})</td>
</tr>
<tr>
<td>480</td>
<td>protons</td>
<td>lanthanum</td>
<td>Murin et al.(^{29})</td>
</tr>
<tr>
<td>480</td>
<td>protons</td>
<td>bismuth</td>
<td>Vinogradov et al.(^{30})</td>
</tr>
<tr>
<td>280</td>
<td>deuterons</td>
<td>silver</td>
<td>Kurchatov et al.(^{31})</td>
</tr>
<tr>
<td></td>
<td>protons</td>
<td></td>
<td></td>
</tr>
<tr>
<td>550</td>
<td>helium ions</td>
<td></td>
<td></td>
</tr>
<tr>
<td>480 and 660</td>
<td>protons</td>
<td>bismuth</td>
<td>Sugarman et al.(^{29})</td>
</tr>
<tr>
<td>2200</td>
<td>protons</td>
<td>bismuth</td>
<td>Sugarman et al.(^{32})</td>
</tr>
<tr>
<td>600 to 3000</td>
<td>protons</td>
<td>lead</td>
<td>Wolfgang et al.(^{33})</td>
</tr>
<tr>
<td>5700</td>
<td>protons</td>
<td>tantalum</td>
<td>This work</td>
</tr>
</tbody>
</table>

Several workers have also systematically studied the formation of particular products of interest, e.g., tritium\(^{34}\), beryllium-7,\(^{35,36}\) lithium-8,\(^{35,37}\) fluorine-18,\(^{38,39}\) sodium-24,\(^{39}\) terbium-149,\(^{40,41}\) etc., and others have similarly studied particular processes, such as the (p,pxn) reactions.\(^{1,2,42,43}\) More refined theoretical calculations have been and are being done.\(^{17,44,45,46}\) The agreement with experiment for target nuclei of A = 50 to A = 75 (and for nuclear emulsions), for bombarding energies of 0.1 to 0.5 Bev, is quite good. Attempts at comparison with radiochemical work for A ~ 200 has been attempted,\(^{47}\) but the experimental
results in this region are not yet good enough to permit more than a qualitative check. Above 0.5 Bev, coincident with the onset of extensive meson production, the theoretical predictions are no longer so good. Apparently new modes of product formation have appeared to complicate the picture.\(^{33}\)

Recently, reliable proton beams of energies up to 6.2 Bev have become available, with the completion of the Cosmotron and Bevatron. The radiochemical work which has been done at the Cosmotron at energies up to 3 Bev displays striking differences with the work done below 0.5 Bev.\(^ {33,48}\) The distribution of products has become so wide that all elements below the target are produced with cross sections of at least a few millibarns. The cross sections for the formation of products that have \(A < 20\) increase by many orders of magnitude. The pronounced fission peak observed in bismuth at the lower energies has apparently disappeared. A new mechanism, "fragmentation" (discussed in Section V), has been postulated\(^ {33}\) to account for most of the new effects, one of the interesting features being that even in the cascade event the nucleus is no longer regarded as a cluster of noninteracting particles, but that groups of nucleons can behave more or less as units.

The present work was initiated as part of a program at Berkeley to extend the radiochemical observations up to about 6 Bev. Tantalum was chosen as the target material because it falls between copper and uranium, on which work was already in progress.\(^ {18,49}\) Also, the results of Nervik and Seaborg\(^ {25}\) in the bombardment of tantalum with 0.34-Bev protons was available for comparison, and it was of some interest, for example, to see how the small fission peak that they observed would change. Tantalum has the advantage of being monoisotopic, and of being readily available in pure foils of varying thicknesses.

It is hoped that such general, exploratory studies as the one described in this report will act as guides to the design of more definitive experiments, with respect to the determination of nuclear structure and properties.
II. EXPERIMENTAL PROCEDURES

A. The Target

The target always consisted of a stack of aluminum and tantalum foils, the exact number, thickness, and arrangement of which depended upon the requirements of the particular experiment that was being performed. In all target stacks, the first three foils on the "upstream" side (the side of the target stack that was first struck by the impinging proton beam) were a 0.001-inch aluminum jacket foil, a 0.003-inch aluminum monitor foil, and a 0.001-inch aluminum guard foil, respectively. The last foil on the "downstream" side of the stack was usually another 0.001-inch jacket foil. The purpose of these aluminum foils will be explained in a subsequent section.

When the nuclides for which cross sections were being measured had atomic numbers between 15 and 71, the total target stack consisted of only the above-mentioned aluminum foils and a single 0.010-inch tantalum foil sandwiched between the aluminum guard foil and the downstream jacket foil.

When nuclides for which cross sections were being measured had atomic numbers less than 15, there was danger that these nuclides, which are also formed in the aluminum foils, would recoil into the tantalum foil and cause high results. In this case a 0.010-inch tantalum foil was used as in the above-described arrangement, except that a 0.001-inch tantalum guard foil was placed on each side of the 0.010-inch foil.

When cross sections were being measured for the production of nuclides with atomic numbers greater than 71, where the results could be seriously affected by the production of secondary particles, a much thinner target was necessary. In this case, both upstream and downstream jacket foils were omitted, and a single 0.001-inch tantalum foil was placed downstream from the aluminum guard foil.

Spectrographic analysis of the tantalum used in these bombardments showed no detectable impurities, except for the presence of a maximum of 0.1% of zirconium in the 0.001-inch tantalum foil. Assuming
reasonable cross sections (based on the work of Markowitz)\textsuperscript{1} for the production of zirconium isotopes from zirconium, it is estimated that the error in the measured cross sections for the production of zirconium isotopes from tantalum will be less than 3\%. Typical detection limits for other elements were: < 0.1\% for tungsten, hafnium; < 0.05\% for niobium, calcium; < 0.01\% for zirconium, molybdenum, iron.

All foils were carefully cleaned of grease and other foreign matter before bombardment by washing with chloroform and ethanol. In addition, the tantalum foils that were used to measure the production of nuclides with atomic numbers less than 15 were soaked overnight in aqua regia and rinsed in distilled water before bombardment, in order to remove any possible surface deposits of iron which might have come from processing the foil.

Since it is critical that all the foils in a target stack be of identical size, all the foils that were to be used as targets throughout this study were clamped together in one stack and machined to 0.75 by 2 inches.

To prepare a target for bombardment, the aluminum monitor and tantalum target foils were carefully weighed with an analytical balance. Then the stack of foils that was to be used was carefully aligned and held together with scotch tape, including a piece of scotch tape on the leading edge. Alignment was never quite perfect; inspection with a low-power binocular microscope revealed the misalignment between the leading edges of the tantalum target foil and the aluminum monitor foil to be always less than about 0.006 inch, where the aluminum would be slightly ahead on one side of the leading edge, and the tantalum would be slightly ahead on the other side. Subsequent activity distribution studies indicate that the error introduced into the results by this misalignment is of the order of 5\% or less. The target stack thus prepared was then mounted on a lucite target holder, which in turn was mounted on a removable aluminum probe head. The probe head was designed to fit the air-driven target ram of the Bevatron.

In the above-described arrangement, the target stack protruded 1-1/4 inch beyond the end of the lucite. In the "thick target" cases,
the entire 0.010-inch tantalum foil was taken for subsequent radiochemical analysis. However, in the "thin target" case, where secondary particles could be troublesome, only the leading 1 inch of the stack was taken, and was cut off with scissors immediately following the bombardment.

The way in which the Bev protons are made to strike the target is as follows: about 0.01 second before the radio-frequency accelerating potential is turned off, the target is plunged by means of an air-driven ram into its bombardment location, which is at a slightly smaller radial distance than the radius of the orbiting protons. The magnetic field is allowed to increase after the proton acceleration has ceased, which forces the orbiting protons to spiral inward until they strike the target. Since they first encounter the "leading edge" of the target stack, one might expect to find that the leading edge has relatively more activity per unit volume of material than the rest of the target. Such is indeed the case, as shown in Figure 1, which was obtained by cutting the aluminum monitor (for a thick-target bombardment) into strips parallel to the leading edge, and measuring the production of Na$^{24}$ in each strip. Fully one-half of the total activity appears in the first 8 or 9 mm of the target, and 20% of the activity appears in the first 1.5 mm.

B. The Beam Monitor

In order to measure absolute cross sections, it was necessary to know how many protons had passed through the target stack. For this purpose, the reaction Al$^{27}$ (p,3pn) Na$^{24}$ was used. After the bombardment, the 0.003-inch aluminum monitor foil was removed from the target stack, cut into pieces, the dimensions of which corresponded approximately to the dimensions of the radioactive chemical samples (to be described later), and mounted on cards in the same way as the chemical samples. The amount of Na$^{24}$ that was present was then measured by placing these cards in a calibrated proportional counter and following
Fig. 1. Integrated fraction of total Na$^{24}$ activity found in monitor foil, that occurs beyond D, the distance from the leading edge.
the decay of the 15-hour half-life component, which was the only activity detectable from 24 hours after bombardment onward (except for a very small amount of Na$^{22}$ and Be$^7$).

Unfortunately the cross section for this reaction at 5.7 Bev is only approximately known at present. Pending the announcement of a better value, 10.5 mb has been adopted. This number was obtained by using the cross section for the reaction C$^{12}$ (p,pn) C$^{11}$, which has been measured up to 4.1 Bev$^{50,51}$ and the ratio of cross sections for the reactions C$^{12}$ (p,pn) C$^{11}$ and Al$^{27}$ (p,3pn) Na$^{24}$, which has been measured up to 3 Bev$^{52,53}$ to calculate an excitation function for the Al$^{27}$ (p,3pn) Na$^{24}$ reaction. This excitation function was then extrapolated to 5.7 Bev. The error in the value thus obtained is thought to be less than ±20%.

Since large numbers of secondary particles are formed in the interaction of Bev protons with nuclei of large A, it is possible that these secondary particles could cause some interference with the monitor reaction, especially due to the reaction Al$^{27}$ (n,α) Na$^{24}$. Such an interference has been reported by Wolfgang et al.$^{33}$ in 1- to 3-Bev bombardments of lead. Based on their results, a correction of 10 to 15% has been applied in this work wherever a "thick" target was used.

C. Sample Preparation

After each bombardment, the tantalum target foil was worked up to obtain radiochemically pure samples of several of the elements that are formed. These samples were to be subjected to measurements of their radioactivity, sometimes over a period of months, and it was necessary to contain the chemical sample in a reasonably sturdy mount. The various procedures involved in accomplishing this aim are described in the following paragraphs.

After the bombardment the tantalum target foil was removed from the target stack and dropped into a 125-ml platinum evaporating dish containing 25 ml of hot, concentrated HF, amounts of suitable carriers
varying from 1.5 mg for rare earths to 30 mg for tungsten, and maintained at nearly 100°C on a steam bath. In those "thick target" cases where the tantalum target foil was flanked on either side by only the 0.001-inch aluminum guard and jacket foils, these two foils were also added to the HF. In all other cases, only the tantalum foil was added. The aluminum foils, when present, dissolved quickly. The tantalum was then dissolved by cautiously adding concentrated HNO₃ dropwise, at a rate sufficient to cause vigorous, but not violent, foaming. Complete dissolution of even the thick tantalum foils generally took only ten minutes or so. As soon as the tantalum was completely dissolved, concentrated formic acid was added, 0.5 ml at a time, until no more reaction was observable, in order to destroy the excess nitric acid present. The target solution was then poured into a lusteroid cone kept in a cold water bath. From this point on, the treatment of the target solution varied, depending on the particular elements being isolated. It may be of some value to mention briefly how different groups of elements were brought out of this target solution, which usually contained gram amounts of tantalum, and concentrated HF.

A combined precipitate, which may be formed by having carriers of both barium and some rare earth present, adding a drop or two of concentrated H₂SO₄, and allowing to stand for a minute or so, was found to contain all the rare earths, magnesium, calcium, strontium, and barium.

Several elements may be precipitated directly by passing H₂S through the target solution; they are copper, arsenic, selenium, molybdenum (saturate with H₂S for 20 minutes), cadmium (copper must be present also), and tellurium.

If the pH of the target solution is raised to the point where tantalum just begins to precipitate (pH ~ 4.5), by cautiously adding concentrated ammonium hydroxide dropwise, accompanied by vigorous stirring, to the target solution which is kept in a cold water bath, the resulting solution will be found to precipitate palladium, and antimony when saturated with H₂S.
Zirconium and hafnium are precipitated upon the dropwise addition of saturated barium nitrate solution. Tantalum, when present in moderate amounts, is precipitated when saturated potassium fluoride solution is added; tungsten may be recovered from the remaining solution by evaporating the HF cautiously on the steam bath.

When 2 ml of concentrated sulfuric acid is added to the cooled target solution, followed by 2 ml of \( \frac{1}{8} M \) silicotungstic acid, the cesium is precipitated, together with some of the rubidium, upon short standing.\(^{54}\)

Silver is best precipitated by adding a few drops of HCl. Many of the elements for which no precipitant can be found may be recovered by first extracting most of the tantalum into di-isopropyl ketone.\(^{25}\) The target solution is made \( 18 \) N in \( H_2SO_4 \), and the tantalum is extracted by shaking with three successive 20-ml portions of di-isopropyl ketone. It is best to use a polyethylene bottle to do the shaking and to use polyethylene pipettes (which are easily fabricated from polyethylene tubing) to separate the layers, when an impurity-sensitive element such as sodium is being separated. The remaining aqueous layer is poured into a 600-ml beaker (or a large platinum dish if sodium is being sought), and boiled gently (there is much foaming) until fumes of \( SO_3 \) appear. Concentrated nitric acid is then added cautiously until the black color disappears, after which the solution is fumed nearly to dryness, cooled, slurried with water, transferred to a 40-ml cone, and centrifuged. The resulting supernatant solution has been found to contain beryllium, sodium, phosphorus, manganese, cobalt, nickel, and zinc.

After removal from the target solution the various elements were subjected to standard radiochemical purification procedures,\(^{25,55,56}\) which were usually used with little or no modification. A description of the purification procedures followed for each element studied will be found in the appendix.

The radiochemically purified elements were precipitated in a suitable form for filtration, and filtered onto a dried, tared disk of No. 42 Whatman filter paper, 7/8 inch in diameter, using the filter-
chimney arrangement described by Friedlander and Kennedy. The sample thus prepared was usually quite uniform in thickness, was circular, and had an area of 2.4 cm$^2$.

The sample on its filter paper was dried at 110$^\circ$ for 15 minutes or more, weighed quickly to provide an approximate assay of the chemical yield, and mounted for counting.

In the earlier part of the work, the sample-mounting arrangement was almost identical to that described by Shudde; the bottom of the filter-paper disk was stuck to the sticky side of a piece of inch-wide "Texcel" brand cellophane tape, and a piece of 0.00025-inch DuPont Mylar (about 1 mg/cm$^2$) was placed on top. The sample was then centered under a one-inch hole in a 2.5-by-3.5-inch cardboard "counting card," another such counting card (also with a one-inch hole) 2.5 by 3.0 inches was placed below for rigidity, and the entire assembly was firmly secured with Scotch tape (Fig. 2).

In the later part of the work, the bottom of the filter-paper disk was stuck to a 1-inch-by-1-inch piece of two-sided Scotch tape that had been previously stuck to the center of a standard 2.5-by-3.5-inch aluminum "counting card" (350 mg/cm$^2$), and a piece of rubber hydrochloride (0.7 mg/cm$^2$) was placed on top of the sample (Fig. 3).

The intensities of the radiations from the samples mounted thus were measured in appropriate detection equipment and their decay followed. When the radiation intensities had become so feeble as to be undetectable, the samples were accurately analyzed to determine the fraction of the original carrier that was recovered. These analyses were performed by members of the Analytical Chemistry Group.

**D. Radiation Detection and Measurement**

Two different radiation-detection instruments were used to measure the radiation intensity of the samples. When the radiations of interest consisted of beta particles, positrons, or conversion electrons, an end-window gas-flow proportional counter was used. When
Fig. 2. Mounting of sample for radiation measurements in the earlier portion of the work.
Fig. 3. Mounting of sample for radiation measurements in the later portion of the work.
the radiations consisted of photons, i.e., gamma rays or x-rays, a sodium iodide crystal scintillation counter was used. Pertinent features of these instruments will be described below.

1. Proportional Counter

The counting chamber was cylindrical (aluminum wall) with a window 1 inch in diameter. In the first part of the study the window consisted of 1.0 mg/cm² aluminized Mylar, and in the second part of the study, it was 0.9 mg/cm² silvered nylon. The counting gas was commercially available pure methane, and was kept flowing constantly through the chamber.

Nine shelves spaced at intervals from 0.130 inch to 2.630 inch from the window provided a selection of geometries that could be used, according to the counting rate of the sample. Usually the counting rates were low, and the first or second shelf was used.

The entire assembly was contained in a lead housing with walls about two inches thick, the inside of which was lined with aluminum about 1/4 inch thick to minimize scattering.

The effective geometry of the various shelves was determined by means of a Na²⁴ source kindly donated for the purpose by Dr. Donald Barr. This source consisted of a piece of 0.003-inch aluminum foil, of nearly the same size and mounted in exactly the same way as the aluminum monitor foils in the second part of this study. The absolute disintegration rate of the Na²⁴ was measured by means of the coincidence counting technique. A factor, F, was obtained which related the observed counting rate on some particular shelf to the absolute disintegration rate:

\[ F = \frac{\text{observed counting rate}}{\text{disintegration rate}} \]

The value of F for Shelf One was 0.459, and for Shelf Two was 0.356. This corresponds to "pure" geometries of 0.323 and 0.251 respectively, when appropriate corrections for self-scattering, self-absorption, backscattering, gamma-ray counting efficiency, and air-window absorption are applied.
This calibration was also done using a RaDEF source calibrated by the National Bureau of Standards. The pure geometry for the second shelf obtained in this way was 0.258.

2. Sodium Iodide Scintillation Counter

The radiation-sensitive element of the scintillation counter was a thallium-activated sodium iodide crystal in the form of a cylinder 1 inch high by 1-1/2 inches in diameter. The window consisted of a thin sheet of beryllium, and a thin layer of magnesium oxide which formed part of the crystal mounting. The effective window thickness was found to be almost negligible with respect to americium L x-radiation (about 19 kev).

The output of the scintillation counter was fed into a 50-channel pulse analyzer which displayed the photon spectrum of the radioactive samples.

A number of shelves were spaced at intervals below the window, to permit the use of different geometries. The crystal mounting and shelf arrangement were contained in a large lead box, with walls two inches thick.

The geometry factors of the various shelves were measured by the 59.7-kev gamma ray emitted by an Am$^{241}$ source. The absolute disintegration rate was measured by alpha-counting the source in a chamber having 52% geometry. The branching ratio between the 59.7-kev gamma ray and the alpha group was taken to be 0.37. Typical geometries determined in this way, corrected for finite source width to correspond to actual samples, were 0.327 for Shelf One and 0.113 for Shelf Two.

The geometry factors were also determined by means of the 1.37-Mev gamma ray emitted by the same Na$^{24}$ source as was used to calibrate the proportional counter. The geometry factors thus obtained, using the photopeak counting efficiencies of Kalkstein and Hollander, agreed with the above values to within 4%.
III. TREATMENT OF DATA

A. Decay-Curve Analysis

In general, each element which was separated contained more than one radioactive isotope, with the result that the decay curves which were obtained had more than one component. In order to obtain the formation cross sections for the isotopes which were detected, it was necessary to resolve the decay curves into their individual components. The methods by which this was done will be described in the following paragraphs.

Radioactive decay is governed by a first-order rate law, so that a plot of the logarithm of the "activity" of a single decaying species versus time will be a straight line. The slope is characteristic of the species being studied and is conveniently expressed by specifying a "half life," which is defined as that time required for the number of active nuclei present to be diminished by one-half.

When two or more active species are present, a similar plot yields a curved line which represents the sum of the activities of each of the individual species. Such curves will "tail" into a straight line with a slope corresponding to the species present which has the longest half life. If this straight line is extrapolated back to the time the measurements were begun, the contribution to the total decay curve of this particular species can be subtracted. The results of the subtraction may then be plotted, and the whole process repeated until the decay curve has been completely "resolved."

There are two complications that may arise to render inadequate the procedure just described. One possibility is that two of the active species present may have half lives so similar that the decay curve never quite goes completely to a definitely linear "tail" before the activity of the sample becomes too feeble to measure. The other complication, which was often encountered in this work, was the situation in which one or more of the decaying species had daughter activities that were also radioactive, with half lives that were roughly the same order of magnitude as the parent half lives.
These cases were usually resolved by the use of a least-squares analysis of the decay data. A theoretical decay law was written in which the half lives of the various components were assumed to be the same as the best values available in the literature, and the most appropriate modification of the least-squares procedures described by Shudde¹ was used.

In certain fortunate circumstances, one of the decaying species would be emitting easily observed gamma rays of known energy and abundance, and in these cases the decay rate of the species could be obtained without resorting to a decay-curve resolution. These cases, and other cases where special techniques were used in the resolution of decay curves, will be mentioned in a subsequent section, where the experimental observations are described for each element studied.

B. Calculation of Disintegration Rates

After a decay curve has been successfully resolved into its components, it is necessary to obtain for each decaying isotope some quantity which will express quantitatively the number of its atoms that were present at the end of the bombardment. It is this quantity which must be used later on in the calculation of the absolute formation cross section for the isotope.

A convenient quantity to use for this purpose is the "disintegration rate," which is defined as the number of atoms of a particular isotope that undergo decay in unit time. Its connection with the number of atoms of the isotope that are present at any given time \( t \) is expressed by

\[
D(t) = -\frac{dN(t)}{dt} = \lambda N(t), \text{ where } \lambda = \ln 2 \frac{T_{1/2}}{T_{1/2}}
\]

where \( D(t) \) is the disintegration rate, \( T_{1/2} \) is the half life, and \( N(t) \) is the number of atoms.

Each component of a successfully resolved decay curve may be extrapolated back to the time the bombardment ended, yielding a set of
radiation intensities whose relation to the corresponding set of disintegration rates of the species involved depends partly on the instruments that were used to establish the decay curve, and partly on certain other factors. It is the purpose of the following paragraphs to describe, for each instrument, how the radiation intensities were converted into disintegration rates.

1. **Proportional Counter**

   The measured radiation intensity (counting rate) must be corrected for geometry, counting efficiency, back-scattering, air and window absorption, self-scattering, and self-absorption, and radiation abundance. The coincidence correction was negligible for all counting rates encountered in this work.

   (a) **Geometry**

   The geometry correction is that fraction of the total radiation which would enter the sensitive volume of the detector in the absence of interfering effects such as scattering, absorption, etc., and depends on the effective solid angle subtended by the counter window as seen from the sample position. Ordinarily the aluminum monitor and the sample were counted on the same shelf so that the geometry factor cancelled. However, in those many cases in which the monitor was counted with the proportional counter and the sample was counted with the scintillation counter, the geometry factors of both counters were very important. The calibration of the proportional counter with respect to geometry has already been described in Section II.

   (b) **Counting efficiency**

   The counting efficiency is that fraction of the radiation entering the detector's sensitive volume which is counted. It was assumed that beta particles, positrons, and conversion electrons counted at 100% efficiency, and that all photons counted with negligible efficiency. Actually, typical counting efficiencies for photons were \( \sim 0.2\% \) for 25-kev photons and \( \sim 1\% \) for 1-Mev photons.
(c) Backscattering

In the first part of the work, the samples were backed by cellophane tape and filter paper having a total thickness of 19 mg/cm². Since this was seldom a "saturation backscattering" situation, the fractional backscattering curve for polystyrene established by Zumwalt was used, together with the curve of saturation backscattering for cardboard as a function of maximum beta energy that was measured by Burtt and the range-energy relation of beta-particles in aluminum established by Glendenin. Using these curves, it was possible to construct a backscattering curve approximately valid for 19 mg/cm² of cellulose backing as a function of the maximum beta-particle energy. Typical values of the backscattering correction factor thus computed were: 1.08 at 0.25 Mev, 1.13 at 0.45 Mev, 1.14 at 0.8 Mev, 1.10 at 1.5 Mev, 1.06 at 2.5 Mev.

In the second part of the work, where 22 mg/cm² of cellulose backing was backed in turn by 350 mg/cm² of aluminum, the value for saturation backscattering in aluminum taken from the curves by Burtt was used. This assumption was checked by Dr. Barr and found to hold approximately true, possibly because the two-sided Scotch tape used in this phase of the work contained some zinc.

Since the backscattering correction for positrons is less than that for beta particles of the same energy, the data of Seliger were used to correct the beta-particle backscattering correction. The specific correction that was adopted was

\[ f_{bs+} = 1 + 0.7 (f_{bs-} - 1) , \]

where \( f_{bs+} \) is the backscattering correction for positrons at a given maximum energy and \( f_{bs-} \) is the corresponding correction for beta negative particles at the same energy.

(d) Air and window absorption

The correction for absorption of beta particles and positrons in the air, sample cover, and counter window, which usually totaled 2 to 3 mg/cm² in total thickness, was based on the procedure devised by Gleason et al., assuming the correction factors to be the same as for an equal
thickness of aluminum, and the correction to be the same for positrons and beta particles.

(e) Self-scattering and self-absorption

Since radiations are absorbed and scattered by samples of finite mass, it is necessary to apply a corresponding self-absorption and self-scattering correction. Several studies of this extremely troublesome correction exist,\textsuperscript{1,67,68,69} but they usually apply accurately only to special sets of well-defined conditions, and are far from comprehensive in the number of maximum beta energies examined in each precipitate. Under the circumstances it was thought wise to use only the widely available study by Nervik and Stevenson.\textsuperscript{67} When the precipitate contained no element heavier than strontium, their curves for NaCl were used; in all other cases the curves for \textit{Pb(NO$_3$)$_2$} were used. It is thought that the error in using this convention seldom exceeded 5%.

(f) Radiation abundance

The radiation-abundance correction depended on the exact decay scheme of each isotope studied. These were taken from the compilation by K. Way et al.,\textsuperscript{70} and from the original literature, when known. When they were not known, assumptions were made, which are noted in the section in which the elements studied are discussed individually. In particular, when positron branching ratios were unknown while positron energies had been measured, the branching ratios were estimated from the calculations of Feenberg and Trigg\textsuperscript{71} and Brysk and Rose,\textsuperscript{72} assuming allowed transitions.

When conversion electrons were being counted, the corrections were applied as if the conversion electrons formed a beta-particle group with a maximum energy that was twice the energy of the conversion electrons.
2. Scintillation Counter

The counting rates of K x-rays and gamma rays were obtained by integrating the appropriate photopeaks in the scintillation spectra of the samples. Figures 4 and 5 illustrate the manner in which the integration was performed in an "easy case" and in a "difficult case" respectively. When the situation was such that the photopeak of interest was distorted by other peaks near by and (or) had an underlying hash composed of Compton "smear" from higher-energy gamma rays, backscattered "peaks" from the floor of the lead housing, etc., it is evident that the convention which has been adopted (i.e., that of estimating a smooth "bottom" for the photon peak) yields only roughly approximate results at best. When such complications were encountered, they will be noted when the decay of the particular isotope involved is discussed in a subsequent section.

The raw photon-counting rates that were obtained by the above techniques are subject to several corrections in order that they may be converted to disintegration rates. These corrections include factors for counter geometry, crystal counting efficiency, escape peak, absorber, dead time, "stack-up" loss, and radiation abundances.

(a) Counter geometry

The geometry correction is the same type of factor as was discussed in the proportional-counter section above. The geometry calibration of the scintillation counter has already been discussed in Section II.

(b) Crystal counting efficiency

The crystal counting efficiency is that fraction of photons of a particular energy entering the crystal which count in the corresponding photopeak observed in the resulting spectrum. For this correction, the curves determined by Kalkstein and Hollander were used.
Fig. 4. K x-ray photopeak appearing in the electron capture decay of a sample of lutetium. This represents a case where the photopeak integration was easily performed.
Co\textsuperscript{55} annihilation radiation
$E = 511$ keV

Co\textsuperscript{58} photopeak
$E = 805$ keV

assumed "hash" background

(time = 485.0 minutes)
(c) Escape peak

The escape-peak correction is necessary because the photopeak may be reduced in size when K x-radiation from the iodine in the sodium iodide escapes from the crystal without further interaction with the crystal material. In favorable cases the result of this process may be observed as a small satellite "escape peak" located at a spectral position corresponding to the photon energy minus 29 kev. Curves expressing the size of this escape peak relative to the main photopeak have been published by P. Axel,73 and were used throughout this work.

(d) Absorption

In many cases, the photons were counted through several hundred mg/cm$^2$ of beryllium in order to screen out energetic beta particles or positrons. The absorption coefficients by Davisson and Evans74 may be used to estimate this correction. The window thickness was assumed to be negligible for all the photons that were observed.

(e) Register dead time

The dead-time correction arises because the 50-channel pulse analyzer was equipped with mechanical registers for which the dead-time loss was measured to be 7% per 100 counts/min. The total correction, which was serious only for the highest counting rates observed in this study, seldom exceeded 10%.

(f) Stack-up loss

When an isotope has coincident photons in its decay, a photopeak may appear at a position corresponding in energy to the sum of the energies of the photons. The photopeak integrations must be corrected for this effect by adding the integrated intensity of the stack-up peak to each real photopeak involved. This correction was usually avoided by counting at a lower geometry, when practical. In the cases where high-geometry counting was employed, no serious stack-up peak was ever observed.
(g) Radiation abundances

When decay schemes were sufficiently well known that absolute abundances of either specific gamma rays or K x-rays (depending on the particular case) could be inferred, the compilation by K. Way et al., 70 or the best data available in the literature was used. When the same sources reported decay schemes which were partially but still insufficiently known to permit absolute abundances to be inferred, the K x-ray abundance was usually estimated. The various factors necessary for this estimate were obtained by using the beta-decay energy systematics of Way and Wood, 75 the electron amplitudes in the nuclear volume computed by Brysk and Rose, 72 the K-shell fluorescence yields compiled by Gray, 76 and the K- and L-shell binding energies of Hill, 77 and assuming the decay to be such that \( |\Delta I| = 0.1 \). Where necessary the computed conversion coefficients of Rose, 78 and of Sliv 79 were used to estimate the contribution to the K x-ray abundance from gamma conversion. Where the decay schemes were totally unknown, suitable guesses were made. All the K x-ray abundances that were used, together with references to the pertinent decay-scheme work, are listed in Table II, in Section IV B.

On some occasions, the scintillation counter was used to count positrons, taking advantage of the characteristic 511-kev annihilation radiation. For this purpose, the sample was sandwiched between sufficient thicknesses of beryllium (or stainless steel) absorber to insure that most of the positron annihilation events occurred near the sample at a reasonably well-defined geometry.

An indication of the agreement that was attained between the cross sections obtained by counting photons and those obtained by counting positrons is given in Table I.
Table I

Comparison of Photon Counting With Particle Counting

<table>
<thead>
<tr>
<th>Bombardment Number</th>
<th>Nuclide</th>
<th>Cross section obtained by photon counting</th>
<th>Cross section obtained by particle counting</th>
</tr>
</thead>
<tbody>
<tr>
<td>17</td>
<td>Ba\textsuperscript{128}</td>
<td>0.92</td>
<td></td>
</tr>
<tr>
<td>21</td>
<td>Ba\textsuperscript{128}</td>
<td>0.92</td>
<td></td>
</tr>
<tr>
<td>22</td>
<td>Ba\textsuperscript{128}</td>
<td>0.87</td>
<td></td>
</tr>
<tr>
<td>25</td>
<td>Zr\textsuperscript{89}</td>
<td>1.19</td>
<td></td>
</tr>
<tr>
<td>26</td>
<td>Zr\textsuperscript{89}</td>
<td>1.36</td>
<td></td>
</tr>
</tbody>
</table>

C. Calculation of Cross Sections

Once the disintegration rate of a given radioactive product in a sample at the end of bombardment has been determined, it remains necessary only to correct for the percent recovery of the original carrier in order to find the total disintegration rate of the product in the target foil. That disintegration rate can then be used, together with the disintegration rate of the Na\textsuperscript{24} in the monitor foil, to calculate the formation cross section of the product according to the formula

\[
\sigma_p = \frac{D_p^0 W_m \lambda_T (1-e^{-\lambda m \cdot t})}{D_m^0 W_T \lambda_m (1-e^{-\lambda_p \cdot t})} \sigma_m',
\]

where \(D_p^0\) and \(D_m^0\) are the disintegration rates of the product, \(p\), and of the beam monitor, Na\textsuperscript{24}, at the end of the bombardment; \(W_T\) and \(W_m\) are the weights of the tantalum foil and the monitor foil; \(M_T\) and \(M_m\) are the atomic weights of tantalum and aluminum; \(\lambda_p\) and \(\lambda_m\) are the decay constants of the product, \(p\), and of Na\textsuperscript{24}; \(t\) is the length of the bombardment; and \(\sigma_m\) is the cross section for the reaction Al\textsuperscript{27} (p,3pn) Na\textsuperscript{24}, which was taken to be 10.5 mb for this work (see Section II, B).
IV. OBSERVATIONS AND RESULTS

A. Nuclides Observed

Beryllium

A gamma ray (which decayed with a half life of 53 days) was observed at ~ 480 kev with the scintillation counter. No beta or positron emission could be observed with the proportional counter, thus the possibility that the observed gamma ray was due to positron annihilation was eliminated. Thus, the activity was assumed to be due to Be⁷, and the cross section was calculated assuming the 477-kev gamma ray to be present in 12% of the disintegrations.

Sodium

The activity observed with the proportional counter decayed with a 15.1-hr half life, assumed to be due to Na²⁴, for more than six half lives, until it finally turned into a very long-lived tail with a counting rate of about one count per min. The counting rate of the long-lived activity was too low to permit a half-life determination to be made, therefore, it was used to calculate an upper limit for the cross section for the production of 2.6-yr Na²².

Magnesium

Only one component, having a half life of 22 hr, was observed in the decay of the activity of the magnesium fraction, using the proportional counter. It was assumed to be Mg²⁸ in equilibrium with its 6.6-min Al²⁸ daughter.

Phosphorus

The activity was observed with a proportional counter, and displayed a half life of ~ 18 days, with a small but definite curvature perceptible in the decay data when plotted on semilog paper. This activity was assumed to be a mixture of 14.3-day P³² and 24.9-day P³³.
Resolution of the two activities was accomplished by performing a least-squares calculation on the data. In two separate determinations, the ratios of the cross section for the production of $^{32}\text{P}$ to that for the production of $^{33}\text{P}$ were 1.4 and 1.1.

**Calcium**

The decay of the activity in the calcium fraction could be resolved into a long-lived tail with a half-life of ~200 days, assumed to be due to 164-day Ca$^{45}$, and a shorter-lived component of complex shape which could be satisfactorily fitted by a least-squares calculation assuming it was due to 4.7-day Ca$^{47}$ with its 3.42-day Sc$^{47}$ daughter.

**Manganese**

The decay curve obtained with the proportional counter was easily resolved into two components having half lives of 2.6 hr and 5.8 days, due to 2.58-hr Mn$^{56}$ and the 5.66-day isomer of Mn$^{52}$, respectively. In addition, a gamma ray of ~850-900 kev was observed with the scintillation counter and found to have a half life of ~300 days. This activity was assumed to be 291-day Mn$^{54}$, which decays by electron capture entirely to an excited level in Cr$^{54}$ which decays in turn with the emission of an 840-kev gamma ray.

**Cobalt**

The activity observed with the proportional counter could be resolved into components having half lives of 1.6 hr, 16 hr, and $\gg$ 15 days. The first two components were assumed to be 1.65-hr Co$^{61}$ and 17.9-hr Co$^{55}$, respectively. No attempt was made to resolve the long-lived tail directly, because of its low counting rate, although it was undoubtedly due to a mixture of Co$^{58}$, Co$^{57}$, and Co$^{56}$. However, a weak gamma ray of ~830 kev was observed with the scintillation counter, and this was assumed to be principally the 805-kev gamma ray prominent in the decay of 72-day Co$^{58}$. A similar gamma ray is to be found among the radiations of 70-day Co$^{56}$, but the cross section for the production of Co$^{56}$ is expected to be much lower than that for Co$^{58}$. 
Nickel

The decay data recorded using the proportional counter were easily resolved into components having half lives of 2.6 hr and 56 hr. In addition, the scintillation counter revealed a weak gamma ray at 525 kev, after the 2.6-hr component had died out, but before the 56-hr component had decayed appreciably. The 2.6-hr and 56-hr components were assumed to be due to 2.56-hr Ni$^{65}$, and 56-hr Ni$^{66}$ in equilibrium with its 5.1-min Cu$^{66}$ daughter, respectively. The gamma ray was used to estimate an upper limit for the cross section for the 56-hr positron-emitting Ni$^{57}$, assuming that it was the 511-kev positron annihilation radiation, since the half life of this gamma ray could not be definitely established.

Copper

Three components were resolved from the copper decay curve, obtained with the proportional counter, having half lives of 62 hr, 12.5 hr, and 3.1 hr. These components were assumed to be due to 58.6-hr Cu$^{67}$, 12.82-hr Cu$^{64}$, and 3.32-hr Cu$^{61}$. The radiation abundances that were used were 100%, 57%, and 66% respectively.

Zinc

The proportional-counter decay curve obtained from the zinc sample contained a prominent component of about 10.5 hr and a very small amount of a long-lived tail. The 10.5-hr component was subjected to a least-squares analysis, assuming that it consisted of 9.33-hr Zn$^{62}$ in equilibrium with its 9.73-min Cu$^{62}$ daughter, and 13.8-hr Zn$^{69m}$. The assumed counting efficiencies for Zn$^{69m}$ + Zn$^{69}$ and Zn$^{62}$ + Cu$^{62}$ were 1.13 and 1.10 respectively.

Selenium

Proportional counting of the selenium sample yielded a three-component decay curve. One component was a good 7.1-hr line, and was assumed to be due to Se$^{73}$ (ground state). The next component was of complex shape, appearing to be due to a 8.9-day parent with a ~25-hr
daughter growing in, and was assumed to be due to 9.7-day Se$^{72}$ with its 26-hr As$^{72}$ daughter. The last component was a very small amount of a long-lived tail, which was probably due to the 127-day Se$^{75}$. The radiation abundances that were assumed for Se$^{73}$ were 0.72 for positrons and 0.21 for the conversion electrons from the 66-kev transition. The radiation abundances assumed for Se$^{72}$ and As$^{72}$ were 0.00 and 0.30, respectively.

**Strontium**

The decay curve that was obtained by proportional-counting the strontium was rather complicated. The components into which the curve was resolved were ~98 min, ~31 hr, ~81 hr, 27.5 days, and a small amount of long-lived tail. For calculations, the 27-day component was assumed to be 25.5-day Sr$^{82}$ in equilibrium with its 1.25-min Rb$^{82}$ daughter, and the radiation abundance for this system was assumed to be 0.96. The 98-min activity was probably a mixture of 70-min Sr$^{85m}$ and 2.8-hr Sr$^{87m}$. The 31-hr activity could have been the 38-hr Sr$^{83}$, and the long-lived tail could have been due principally to Sr$^{85}$ and Sr$^{89}$. The 81-hr activity, which was present in small amount, is difficult to explain.

Two of the most prominent photopeaks observed with the scintillation counter appeared at 230 kev and 390 kev. The 230-kev peak decayed with a 73-min half life and was assumed to be the 225-kev gamma ray associated with the internal transition decay of 70-min Sr$^{85m}$. In calculating the cross section for this product a gamma-ray abundance of 0.84 was used. The 390-kev photopeak yielded a two-component decay curve; the components were 2.6 hr and 35.5 hr. The 2.6-hr component was assumed to be the 388.2-kev gamma ray associated with the internal transition decay of 2.83-hr Sr$^{87m}$, and a gamma-ray abundance of 0.73 was used in calculating the cross section. The 35.5-hr component of the 390-kev gamma ray is probably associated with the decay of 38-hr Sr$^{83}$, and is quite prominent.
Zirconium

The decay curve obtained with the proportional counter was resolved into 78-hr and ~100-day components. The shorter-lived component was assumed to be 79.3-hr Zr$^{89}$, and a cross section was calculated taking the positron abundance to be 0.25 and the total contribution from the 913-kev gamma ray to the effective radiation abundance to be 0.02. The long-lived tail was probably due to 85-day Zr$^{88}$ with its 105-day Y$^{88}$ daughter. A prominent photopeak at ~910 kev which decayed with a half life of about 79 hr was observed with the scintillation counter. Another cross-section determination of Zr$^{89}$ was based on this observation, assuming the radiation abundance of the 913-kev gamma ray to be 0.99.

A comparison of the two different determinations of the formation cross section of Zr$^{89}$ will be found in Table I. A prominent photopeak at ~390 kev decaying with a half life of 86 days was assumed to be the 395-kev gamma ray associated with the decay of 85-day Zr$^{88}$, and a cross section was calculated assuming the gamma-ray abundance to be 0.97.

Molybdenum

The decay curve taken with the proportional counter appeared to have at least three components, ~11 hr, 68 hr, and a very small amount of long-lived tail. The 11-hr component was actually rather gently curved, varying from an 8.0-hr "half life" near the beginning of the count to one of about 14 hr before the data became unreliable. The interpretation of this observation is that the decaying species responsible for the short-lived component must consist of a mixture of 5.7-hr Mo$^{90}$ with its 14.60-hr Nb$^{90}$ daughter and 6.9-hr Mo$^{93m}$. A least-squares analysis of the data was made in which the above-quoted half lives were assumed, together with the following radiation abundances: Mo$^{90}$, 0.33 for positrons, 0.16 for the 240-kev conversion electrons, and 0.39 for the 100-kev conversion electrons; Nb$^{90}$, 0.33 for positrons and 0.18 for the 125-kev conversion electrons; and Mo$^{93m}$, 0.7 for the ~250-kev conversion electrons. The 68-hr component was assumed to be due to the presence of Mo$^{99}$, for which a radiation abundance of 1.00
for beta particles and 0.09 for ~130-kev conversion electrons was assumed. The long-lived tail could have been due to ~2-yr Mo\textsuperscript{93}.

Palladium

The decay of the palladium fraction was followed by counting the K x-rays of rhodium and ruthenium with the scintillation counter. The three components that were resolved were: a 9.0-hr half life, assigned to 8.5-hr Pd\textsuperscript{101}; a 4.1-day half life with evidence for a daughter activity of 18.5-hr half life, which was assumed to be 4.1-day Pd\textsuperscript{100} with its 20.8-hr Rh\textsuperscript{100} daughter; and a 17-day half life, which was assigned to 17.0-day Pd\textsuperscript{103}. A least-squares analysis was employed in order to ascertain the contribution from the Pd\textsuperscript{100} + Rh\textsuperscript{100} component. The estimated K x-ray abundances that were used in the cross section calculations are as follows: for Pd\textsuperscript{100}, 0.65; for Pd\textsuperscript{101}, 0.66; for Pd\textsuperscript{103}, 0.78.

Silver

The decay curve obtained using the proportional counter was resolved into components having half lives of 65 minutes, ~4 hr, and a small amount of a ~10-day tail. The 65-min activity was assumed to be entirely due to the 65-min Ag\textsuperscript{103}, and a calculated positron abundance of 0.31 was used in computing the cross section. The 4-hr component was present in very small amount, and an accurate half life could not be obtained. It was thought to be due to the presence of a mixture of 3.12-hr Ag\textsuperscript{112} and 5.3-hr Ag\textsuperscript{113}, and a combined cross section was estimated using the measured "half life" of 3.5 hr. The 10-day tail could have been due to a mixture of 40-day Ag\textsuperscript{105}, 8.3-day Ag\textsuperscript{106}, 270-day Ag\textsuperscript{110}, 7.5-day Ag\textsuperscript{111}, and 17-day Pd\textsuperscript{103}.

Scintillation counting of the electron-capture K x-rays in a sample containing no Pd\textsuperscript{103} permitted the resolution of two more components, 40-day Ag\textsuperscript{105} and 8.3-day Ag\textsuperscript{106}. In obtaining cross sections, the K x-ray abundances that were assumed were 0.80 for Ag\textsuperscript{105} and ~0.7 for Ag\textsuperscript{106}. 

-36-
Cadmium

The K x-ray photopeak decay curve was dominated by a 6.9-hr component which ultimately turned into a ~500-day tail. There was also some evidence for a ~1-hr component. These components correspond to 6.74-hr Cd$^{107}$, 470-day Cd$^{109}$, and a mixture of 59-min Cd$^{104}$ and 54.7-min Cd$^{105}$ respectively. The K x-ray abundances for Cd$^{107}$ and Cd$^{109}$ were estimated to be 1.05 and 0.93 respectively in calculating cross sections.

Scintillation counting of the positron annihilation photopeak yielded a decay curve dominated by a complex component which appeared to include a 1-hr parent and a 1-hr daughter growing in. This component should be due to a mixture of 59-min Cd$^{104}$ with its 27-min Ag$^{104}$ daughter and 54.7-min Cd$^{105}$. A small contribution from a longer-lived component was also present. No attempt was made to interpret these data further, however, owing to a lack of reported information concerning positron abundances.

Several attempts were made to observe 54-hr Cd$^{115}$, without success; therefore an upper limit for the formation cross section of Cd$^{115}$ (ground state) was determined.

Antimony

The K x-ray photopeak decay curve was rather difficult to interpret unambiguously, because it had a large number of components and most of the counting rates were low owing to small formation cross sections. The longer-lived section of the curve displayed a "half life" of 48 hr just before the radiation became immeasurably feeble. Since the expected half lives in this region would be due to 38.0-hr Sb$^{119}$ and 5.8-day Sb$^{120}$, this portion of the curve was "resolved" analytically assuming these half lives. The resulting cross section for the formation of Sb$^{120}$ is, of course, somewhat shaky, although the formation cross section for Sb$^{119}$ shouldn't be too bad. The earlier portion of the decay curve, which was obtained by subtracting the extrapolated section discussed above, had an initial effective half life of ~3 hr, and displayed some curvature toward longer half lives. This section was assumed to be a mixture of 2.8-hr Sb$^{117}$...
and 5.1-hr $^{118}$Sb, and was resolved analytically. The contribution of the 5.1-hr activity was small, and therefore somewhat uncertain. In calculating cross sections, the following K x-ray abundances were adopted: for $^{120}$Sb, 1.0; for $^{119}$Sb, 0.86; $^{118}$Sb, 0.76; $^{117}$Sb, 0.80.

Proportional counting yielded a decay curve which was resolved into 3.0-hr, ≈4-day, and ≈1-yr components. The activity level of the 4-day component was quite low (from 2 cpm to 9 cpm), although the data appear to form a fairly good curve. Both 2.8-day $^{122}$Sb and 5.8-day $^{120}$Sb might contribute to a 4-day resultant effective "half life", hence an analytical resolution was based on this assumption. The resulting cross section for the formation of $^{122}$Sb is thus probably an upper limit. The reason for the 1-yr tail is unknown; perhaps it is due to 250-day $^{119}$Sn.

Tellurium

The decay curve obtained using the proportional counter resolved into 2.4-hr, ≈16-hr, and 6.0-day components, indicating the probable presence in the sample of 2- to 4-hr $^{116}$Te with its 10- to 20-min $^{116}$Sb daughter, 2.5-hr $^{117}$Te with its 2.8-hr $^{117}$Sb daughter, 16-hr $^{119}$Te, and 6.0-day $^{118}$Te with its 3.5-min $^{118}$Sb daughter. In calculating cross sections, it was assumed that the 2.5-hr component was due entirely to $^{117}$Te, and that the positron abundance in its decay was 0.69. The positron abundance associated with the decay of $^{118}$Te and its $^{118}$Sb daughter was calculated to be 0.83.

Scintillation counting of the K x-ray photopeak yielded a decay curve from which components of 17 days, 5 days, 38 hr, and ≈2.8 hr were resolved. The 17-day component was evidently $^{121}$Te, and a K x-ray abundance of 0.73 was used to obtain the formation cross section. The 5-day component could have been due to a mixture of 6.0-day $^{118}$Te and 4.5-day $^{119}$Te, but the data were too uncertain to permit a really trustworthy analytical resolution. The 38-hr component presented something of a puzzle. The earlier portion of this component gave the appearance of tending to a somewhat shorter half life than 38 hr, but the overwhelming 2.8-hr component largely obscured this feature. The tentative
explanation for this and other data to be described shortly is that
4.5-day Te$^{119}$ is really a metastable state of Te$^{119}$, and 16-hr Te$^{119}$
is the ground state. The nonobservation of a strong 16-hr component in
the K x-ray decay is seen to be reasonable if one makes a synthetic plot
of a case where a 16-hr parent grows into a 38-hr daughter; only a very
gentle curvature appears in the early part of the curve.

A very prominent photopeak appeared in the scintillation spectra
at 640 kev, and was found to decay with a half life of 16.0 hr. This
gamma ray was assumed to be associated with the decay of Te$^{119}$, and
might be identified with the 0.56-Mev gamma ray reported by Dropesky.$^{21}$

The disintegration rate of 16-hr Te$^{119}$ at the end of bombard-
ment was calculated from the 38-hr Sb$^{119}$ component assuming the K x-ray
abundance to be 0.86, and also from the 640-kev gamma ray assuming its
abundance to be 1.0, with the following result:

$$\frac{\text{disintegration rate based on 640-kev gamma ray}}{\text{disintegration rate based on Sb}\text{\textsuperscript{119} daughter}} = 0.89.$$ 

The reported cross section for 16-hr Te$^{119}$ is based on the 38-hr Sb$^{119}$
component.

If it be assumed that 4.5-day Te$^{119}$ is the upper state, and must
decay by internal transition to 16-hr Te$^{119}$ and thence through the 640-
kev gamma ray, a 4.5-day component in the decay of that gamma ray should
have been seen. Since it was not, an upper limit for the production of
4.5-day Te$^{119}$ was calculated. A partial check of this assumption was
made by assuming that the 5-day component in the K x-ray decay was due
entirely to 6-day Te$^{118}$, and computing a cross section for the production
of Te$^{118}$ on that basis. The cross section so obtained agreed within
experimental error with the cross section for Te$^{118}$ obtained from pro-
portional-counting data. Thus it seems probable that the cross section
for the formation of 4.5-day Te$^{119}$ is indeed low.

After nearly all of the radiations had died away, the scintil-
lation counter revealed a weak but definite photopeak at 212 kev, which
had a long half life (> 70 days). This radiation was assumed to be the
213-kev gamma ray associated with the decay of 150-day Te$^{121m}$, and a
cross section was calculated assuming the gamma ray abundance to be 0.91.
Cesium

The K x-ray photopeak decay curve could be resolved into components having half lives of 7 hr, ~33 hr, and 20 days. The corresponding activities were assumed to be 6.3-hr Cs$^{127}$, 32.8-hr Cs$^{129}$, and a mixture of 9.7-day Cs$^{131}$ and 36.4-day Xe$^{127}$, respectively. An analytical "resolution" of the long-lived tail (for which counting rates were very low) was used to estimate an upper limit for the cross section of Cs$^{131}$. K x-ray abundances assumed in calculating cross sections were: for Cs$^{127}$, 1.0; for Cs$^{129}$, 1.26; and for Cs$^{131}$, 0.73.

Barium

The decay curve obtained using the proportional counter was resolved into 1.8-hr, 2.4-day, and 11.5-day components, with a very small amount of long-lived tail. Scintillation counting of the annihilation radiation yielded a decay curve resolvable into 1.9-hr and 2.4-day components. Scintillation counting of the K x-ray photopeak produced a decay curve which could be resolved into components having half lives of ~2 hr, ~30 hr, 2.4 days, and a complex long-lived portion, which appeared to be a ~12-day activity having a radioactive daughter of nearly the same half life.

The ~2-hr activity was assumed to be a mixture of 1.6-hr Ba$^{126}$ with its 1.6-min Cs$^{126}$ daughter, and 1.9-hr Ba$^{129}$. The cross section for the production of Ba$^{129}$ alone was determined by an experiment in which its 32.8-hr Cs$^{129}$ daughter was isolated and counted separately. The cross section for the production of Ba$^{126}$ alone was then calculated from the average "total effective counting rate" of the mixture, in which both K x-ray and positron counting rates were included, and from which the calculated total contribution of Ba$^{129}$ was subtracted. A positron-counting efficiency of 0.82 was assumed for Ba$^{126} +$ Cs$^{126}$, together with a K x-ray counting efficiency of 0.89. The corresponding quantities in Ba$^{129}$ could not be accurately determined; however, the experimental data indicate that the positron-counting efficiency is small, roughly 0.1.
The ~30-hr component in the K x-rays probably arises from a mixture of 39-hr Ba$^{133m}$, 29-hr Ba$^{135m}$, and Cs$^{129}$. Attempts to resolve this mixture were unsuccessful, although they indicated that the formation cross sections for Ba$^{133m}$ and Ba$^{135m}$ were probably small.

The 2.4-day activity was presumably due to Ba$^{128}$ with its 3.8-min Cs$^{128}$ daughter. Counting efficiencies of 0.76 for positrons, 0.10 for conversion electrons (~250 kev), and 1.14 for K x-rays were adopted for the Ba$^{128}$ + Cs$^{128}$ mixture.

The complex ~12-day component was assumed to be due to 11.5-day Ba$^{131}$ with its 9.6-day Cs$^{131}$ daughter, and its relative contribution was established by least-squares analysis of the K x-ray data. K x-ray abundances of 0.94 and 0.73 were used for Ba$^{131}$ and Cs$^{131}$ respectively. 12.8-day Ba$^{140}$ could interfere with this analysis, if present, but the proportional counting data indicated that its formation cross section was negligible, < 0.5 mb.

The ratio
\[
\frac{\text{photon abundance of 375-kev + 420-kev gamma rays}}{\text{photon abundance of K x-rays}} = 0.66
\]
was measured for the nearly clean Cs$^{129}$ sample obtained in the course of this work, and was combined with the data of several other workers to estimate a K x-ray abundance of 1.26.

**Cerium**

Four components were resolved from the decay curve obtained by proportional counting of the sample. The first component was definitely curved, and appeared to be a case in which a 6- or 7-hr activity was the parent of a 4- or 5-hr activity. The other components had half lives of ~24 hr, 72 hr, and ~30 days. The first component was probably 6.3-hr Ce$^{133}$ with its 4-hr La$^{133}$ daughter. Complications involved in estimating the positron abundances involved in this system prevented the calculation of an absolute cross section for the formation of Ce$^{133}$; the reported cross section is based on an assumed 100% positron abundance for Ce$^{133}$. The 24-hr component was probably a mixture of 39-hr Ba$^{133}$, 22-hr Ce$^{135}$, 19-hr La$^{135}$, and 35-hr Ce$^{137}$. No attempt was made to
resolve this mixture. The 72-hr component, which was very prominent, was undoubtedly due to 72.0-hr Ce\(^{134}\) with its 6.5-min La\(^{134}\) daughter. A positron abundance for the decay of La\(^{134}\) of 0.44 was used in obtaining the formation cross section for Ce\(^{134}\). The ~30-day component was very feeble, and probably represented a mixture of 140-day Ce\(^{139}\) and 32-day Ce\(^{141}\).

Attempts were made to follow the positron annihilation and K X-ray photopeaks, using the scintillation counter, in order to help resolve the complicated initial portion of the decay curve, but the results were inconclusive.

**Neodymium**

Proportional counting yielded a decay curve from which components corresponding to half lives of 5.5 hr and 3.3 days were resolved. The first component was probably due to 5.5-hr Nd\(^{139}\) with its 4.5-hr Pr\(^{139}\) daughter. No cross section is reported for the formation of Nd\(^{139}\), because in such systems, where the daughter half life is comparable to the parent half life, the calculated disintegration rates are extremely sensitive to the assumed positron abundances. The 3.3-day activity was assumed to be Nd\(^{140}\) with its 3.4-min Pr\(^{140}\) daughter, and a positron abundance for Pr\(^{140}\) of 0.58 was assumed.

**Europium**

The decay curve obtained by counting the K X-rays with the scintillation counter was resolved into components having half lives of 5 days, 24 days, and >1 yr. These components were thought to be due to 5-day Eu\(^{145}\), 24-day Eu\(^{147}\), and the 1-yr Sm\(^{145}\) daughter of Eu\(^{145}\) mixed with 120-day Eu\(^{149}\). No definite evidence was seen to indicate the presence of 58-day Eu\(^{148}\), although the formation cross section of this "shielded" isotope is of considerable interest. Also, a 38-hr component, which would correspond to Eu\(^{146}\), was not observed. This result is surprising, since in one case counting was begun less than 14 hr after the end of the bombardment, and one would expect that even
the independent yield of Eu\(^{146}\) would be measurable. Also, the long-lived tail displayed a "half life" which, although not accurately measured, seemed too long to be explained on the basis of known isotopes. In calculating cross sections, K x-ray abundances of 1.0 were assumed for both Eu\(^{145}\) and Eu\(^{147}\).

Gadolinium

The K x-ray photopeak decay curve was resolved into components having half lives of 33 hr, 9 days, 26 days, and \(\sim 1\) yr. The isotopes responsible for the first two components were very likely 29-hr Gd\(^{147}\) and 9-day Gd\(^{149}\). The 1-yr tail never became a straight line so that a definite half life could be obtained. It was probably due to a mixture of 150-day Gd\(^{151}\), 236-day Gd\(^{153}\), and 120-day Eu\(^{149}\). The 26-day component is unexplained. Although 24-day Eu\(^{147}\) is expected to appear as the daughter of Gd\(^{147}\), the 26-day activity was far too prominent to be accounted for in this way, and moreover, was present in the same relative amount in both determinations (one of which was done after the Gd\(^{147}\) had decayed away). One might be led to suspect europium contamination in the gadolinium samples, except that the computed "cross section" of the 26-day component is so high that one is forced to postulate that 100\% of the europium was present in the gadolinium sample each time. This possibility is thought to be very unlikely because previous experience with the ion-exchange separation technique that was used has shown that the europium-gadolinium separation is good; and also, in taking the gadolinium sample from the elution tubes, only the center of the gadolinium peak was taken, while the trailing side of the peak (which would be sensitive to europium contamination) was rejected. It is thought that perhaps the nonobservation of Eu\(^{146}\) could mean that its Gs\(^{146}\) parent is relatively long-lived, which provides weak evidence that the unassigned 26-day component is Gd\(^{146}\). The calculated cross section for the 26-day component is not inconsistent with this possibility. Shirley, \(^{81}\) who has bombarded Sm\(^{147}\) with 46-Mev helium ions, did not observe a 26-day component among the products, which suggests that the 26-day material may be of mass number 146 or less.
After the 26-day component had disappeared, two clearly discernible photopeaks (in addition to the K x-rays) were observed in the scintillation spectra, appearing at 104 kev and 155 kev. The 104-kev photopeak was assumed to be due to the 102-kev gamma ray associated with the decay of 236-day Gd$^{153}$, and a gamma-ray abundance of 0.56 was assumed in calculating the cross section. The 155-kev gamma ray was assumed to be the 154-kev gamma ray reported by V. Shirley to be associated with the decay of 150-day Gd$^{151}$. A gamma-ray abundance of 0.065 was assumed in computing a cross section.

**Erbium**

The K x-ray photopeak decayed with a 29-hr half life straight into background. This activity was assumed to be 29.4-hr Er$^{160}$ in secular equilibrium with its 5.0-hr Ho$^{160}$ daughter, and a K x-ray abundance of 0.82 was assumed for each isotope.

**Thulium**

The decay curve obtained by following the K x-ray photopeak could be resolved into three components. The first component was of a complex shape which was shown by means of a synthetic plot to be what one would expect if a 29-hr parent were decaying to a 10-hr daughter, and therefore this section of the curve was assumed to be due to 29-hr Tm$^{165}$ with its 9.9-hr Er$^{165}$ daughter. A K x-ray abundance of 0.97 was used for Tm$^{165}$ in calculating the cross section. The second component was ~9.5 days, and was assumed to be due to 9.4-day Tm$^{167}$. The K x-ray abundance of Tm$^{167}$ was estimated to be 0.97. The third component, in which the counting rates were rather low, appeared to have a half life of ~80 days. It was assumed to be 87-day Tm$^{168}$, and an approximate cross section was estimated assuming a K x-ray abundance of 0.97.

**Lutetium**

The decay curve obtained by scintillation counting of the K x-ray photopeak was resolved into components having half lives of 1.7 days,
7.7 days, and ~200 days. The first component was assumed to be due to 1.7-day Lu\textsuperscript{170}, and the cross section was calculated assuming a K x-ray abundance of 1.0. The 7.7-day component could have been due to a mixture of 8.5-day Lu\textsuperscript{171m} and 6.7-day Lu\textsuperscript{172}, with perhaps a small contribution from 6.8-day Lu\textsuperscript{177}. A combined "cross section" was calculated for this component, assuming a K x-ray abundance of 1.0. The 200-day component, for which the counting rates were rather low, was probably a mixture of 1.4-yr Lu\textsuperscript{173} and 165-day Lu\textsuperscript{174}. An attempt was made to resolve this section analytically, but the results were inconclusive because contamination by 32-day Yb\textsuperscript{169} is somewhat likely, and although the presence of a small amount of contamination was not definitely proved, neither could the possibility be definitely eliminated.

**Hafnium**

The K x-ray photopeak decay was resolved into components having half lives of 23 hr and 70 days, and a long-lived tail. The 23-hr component could have been due to a mixture of 16-hr Hf\textsuperscript{171} and 23.6-hr Hf\textsuperscript{173}. In one of the determinations, the final radiochemical purification of the hafnium was done several days after the end of the bombardment, so that the observed 24-hr component was relatively rich in Hf\textsuperscript{173} and poor in Hf\textsuperscript{171}. Moreover, in this decay curve, no 8.5-day component, corresponding to the Lu\textsuperscript{171m} daughter of Hf\textsuperscript{171}, was detected, which indicates that Hf\textsuperscript{171} was present in only small amounts if at all. In the other run, in which the final radiochemical purification was done sooner, a ~7-day component was observed, which probably was due to 8.5-day Lu\textsuperscript{171} appearing as the daughter of Hf\textsuperscript{171}. The cross section of Hf\textsuperscript{171} was based on the ~7-day component assuming the K x-ray abundance of Lu\textsuperscript{171} is 1.0. Since the 7-day component was rather weak, the reported cross section for Hf\textsuperscript{171} is only roughly approximate. The Hf\textsuperscript{173} formation cross section was calculated from the 24-hr component, assuming a K x-ray abundance of 0.97. The 70-day component was assumed to be due to 70-day Hf\textsuperscript{175}, and the cross section was based on a K x-ray abundance of 0.97. The long-lived tail could have come from a mixture of 1.4-yr Lu\textsuperscript{173} and 5-yr Hf\textsuperscript{172}. 
Prominent photopeaks were observed in the scintillation spectra at 120 kev, ~300 kev, and ~340 kev. The 120-kev photopeak decayed with a half life of ~28 hr, and was assumed to be the prominent 121-kev gamma ray reported by Wapstra et al. to be associated with the decay of 23.6-hr Hf$^{173}$. A cross section was calculated for the formation of Hf$^{173}$ based on this data, assuming the 121-kev photon abundance to be 1.1 (this was based on Wapstra's reported 121-kev photon intensity: K x-ray intensity). The resulting cross-section value was lower by a factor 1.3 than the cross section determined from K x-ray data. The reported cross section is based on the 121-kev photon data, because of the uncertain contribution of Hf$^{171}$ decay to the K x-ray data.

The 340-kev photopeak decayed with a good 70-day half life, and was assumed to be due to the 343-kev gamma ray reported by Hatch et al. and others to be prominent in the decay of 70-day Hf$^{175}$. A cross section for the formation of Hf$^{175}$ was calculated taking the 343-kev photon abundance to be 0.86, based on the reported photon abundances. The agreement between the cross sections based on K x-rays and those based on the 343-kev photons is rather poor, the K x-ray data yielding results lower by a factor 1.4. The agreement between two different runs with respect to each type of cross section is quite good, however, hence, the difficulty must be in the interpretation of the decay schemes involved. Since the total decay energy is unknown, it was assumed that the K x-ray abundance was in error, and the reported cross section is based on the 343-kev gamma-ray data.

Tantalum

Scintillation counting of the K x-ray photopeak yielded a decay curve which could be resolved into components having half lives of 8.5 hr, 2.3 days, ~21 days (very weak), and a long-lived tail with a half life > 1 yr. The 8.5-hr activity was probably due to a mixture of 8.0-hr Ta$^{176}$ and 8.1-hr Ta$^{180m}$, and a combined cross section was calculated using a K x-ray abundance of 0.79 (this was a weighted average, using 0.94 for Ta$^{176}$ and 0.74 for Ta$^{180m}$). The 2.3-day component was
assumed to result from the presence of 53-hr Ta$^{177}$, and an estimated K x-ray abundance of 0.87 was used in calculating the cross section. The 21-day component was assumed to be due to a small amount of W$^{178}$ contamination in the tantalum sample, since the complete separation of tungsten from tantalum is known to be difficult. The long-lived tail, which was present at an easily measurable counting rate in one sample, was assumed to be due entirely to 600-day Ta$^{179}$. This assumption is thought to be reasonable, barring the existence of other undiscovered long-lived, neutron-deficient tantalum isotopes, because the K x-ray photopake counting is quite restrictive to the region of tantalum, and no such long-lived tail was observed in the tungsten sample. The formation cross section for Ta$^{179}$ was based on the K x-ray abundance of 0.40, which was measured by Bisi et al.

It is of interest to obtain the cross sections for Ta$^{176}$ and Ta$^{180m}$ separately, because of interest in Ta$^{180m}$ as the (p,pn) product of Ta$^{181}$. Two attempts were made to accomplish this. In the first attempt the samples were counted through 19.4 mg/cm$^2$ of aluminum absorber, to eliminate the contribution from conversion electrons, and the 8-hr component of the resulting decay curve was made the basis of a cross-section calculation for Ta$^{180m}$, assuming a beta-particle abundance of 0.21. The second attempt was to count the 202-kev gamma ray reported by Felber to be present in the decay of Ta$^{176}$. A feeble gamma ray was observed at ~200 kev, and found to decay with a half life of ~10 hr, although there was considerable scatter in the points. A cross section was calculated for Ta$^{176}$ based on a 202-kev photon abundance of 0.063. The sum of these two determinations is only 0.67 of the combined cross section determined using K x-rays, and their relative magnitudes are not what one would expect from other work of this nature in this energy region. These cross sections, although reported, are thought to be untrustworthy.
The decay curve of the K x-ray photopeak was resolved into 2.5-hr, 8-hr, 2.4-day, and 22-day components. The first component could have been due to a mixture of 1.3-hr W$_{176}$ and 2.2-hr W$_{177}$, although the half life seems a little too long. Since counting was not begun until about 8 hr after bombardment, it was thought that most of the 1.3-hr activity had decayed away, and the cross section for W$_{177}$ was therefore estimated from the 2.5-hr component assuming a K x-ray abundance of 1.0. Obviously this cross section represents only an order of magnitude at best. The 8-hr and 2.4-day components would be due to Ta$_{176}$ and Ta$_{177}$ respectively, which would appear as daughter activities to W$_{176}$ and W$_{177}$. In principle it is possible to estimate the cross sections for W$_{176}$ and W$_{177}$ from these components, but the likelihood of tantalum contamination in the tungsten, to which these components would be especially sensitive (if the tungsten sample was 1% by weight of the original tantalum, the expected 8-hr component would have been doubled), rendered such a procedure unwise, although the presence of tantalum contamination was not proved. It is worth recording that if the tungsten samples were clean, the cross sections for the formation of W$_{176}$, W$_{177}$, and W$_{178}$ were all of the same order of magnitude, about 2 or 3 mb. The 22-day component was probably due to 22.0-day W$_{178}$ with its 9.3-min Ta$_{178}$ daughter. The K x-ray abundance of the mixture was calculated from the data of Felber$^{86}$ to be 1.54.

The 22-day tail was followed at length, until the counting rate was so low that further measurements were not feasible, in an attempt to detect the presence of 145-day W$_{181}$, a product of especial interest. No such component was detectable, and an upper limit for the formation cross section of W$_{176}$ was calculated assuming the K x-ray abundance of 0.37 which was measured by Bisi et al.$^{87}$
B. Lists of Photon Abundances

The following two tables list the photon abundances that were used in converting the measured photon-counting rates into disintegration rates.

Table II applies to K x-rays. The first and second columns list the nuclide and its value of K x-ray photons per disintegration, respectively. The third column is a reliability estimate: G means that enough information concerning the decay scheme was available, or could be inferred, that the listed K x-ray abundance is thought to be as reliable as the photo-peak integrations; M means that the listed K x-ray abundance is based on such fragmentary decay scheme data that one or more of the factors in its estimation involves a plausible guess; P means that the listed value is only a guess, based on more reliable figures near by. (See Section III, B for further remarks.)

**Table II. K x-ray abundances**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>K x-rays per disintegration</th>
<th>Estimated reliability</th>
<th>reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rh(^{100})</td>
<td>0.65</td>
<td>G</td>
<td>88</td>
</tr>
<tr>
<td>Pd(^{100})</td>
<td>0.65</td>
<td>P</td>
<td>88</td>
</tr>
<tr>
<td>Pd(^{101})</td>
<td>0.66</td>
<td>G</td>
<td>89</td>
</tr>
<tr>
<td>Pd(^{103})</td>
<td>0.78</td>
<td>M</td>
<td>90, 91</td>
</tr>
<tr>
<td>Ag(^{105})</td>
<td>0.80</td>
<td>G</td>
<td>92, 93</td>
</tr>
<tr>
<td>Ag(^{106})</td>
<td>~0.7</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>Cd(^{107})</td>
<td>1.05</td>
<td>G</td>
<td>94</td>
</tr>
<tr>
<td>Cd(^{109})</td>
<td>0.93</td>
<td>G</td>
<td>95, 96</td>
</tr>
<tr>
<td>Sb(^{117})</td>
<td>0.80</td>
<td>G</td>
<td>97</td>
</tr>
<tr>
<td>Sb(^{118})</td>
<td>0.76</td>
<td>M</td>
<td>98</td>
</tr>
<tr>
<td>Sb(^{119})</td>
<td>0.86</td>
<td>G</td>
<td>99</td>
</tr>
<tr>
<td>Te(^{121})</td>
<td>0.73</td>
<td>M</td>
<td>101, 102</td>
</tr>
<tr>
<td>Cs(^{127})</td>
<td>1.0</td>
<td>P</td>
<td>103</td>
</tr>
<tr>
<td>Cs(^{129})</td>
<td>1.26</td>
<td>M</td>
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<tr>
<td>Cs(^{131})</td>
<td>0.73</td>
<td>G</td>
<td>105</td>
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</tbody>
</table>

(continued)
<table>
<thead>
<tr>
<th>Nuclide</th>
<th>K x-rays per disintegration</th>
<th>Estimated reliability</th>
<th>reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba$^{126}$ + Cs$^{126}$</td>
<td>0.89</td>
<td>G</td>
<td>106</td>
</tr>
<tr>
<td>Ba$^{128}$ + Cs$^{128}$</td>
<td>1.14</td>
<td>G</td>
<td>107, 108</td>
</tr>
<tr>
<td>Ba$^{131}$</td>
<td>0.94</td>
<td>G</td>
<td>109, 110</td>
</tr>
<tr>
<td>Eu$^{145}$</td>
<td>1.0</td>
<td>P</td>
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</tr>
<tr>
<td>Eu$^{147}$</td>
<td>1.0</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>Gd$^{147}$</td>
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<td>P</td>
<td></td>
</tr>
<tr>
<td>Gd$^{149}$</td>
<td>1.0</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>Er$^{160}$</td>
<td>0.82</td>
<td>M</td>
<td>25</td>
</tr>
<tr>
<td>Ho$^{160}$</td>
<td>0.82</td>
<td>P</td>
<td>25</td>
</tr>
<tr>
<td>Tm$^{165}$</td>
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<td>P</td>
<td></td>
</tr>
<tr>
<td>Tm$^{167}$</td>
<td>1.0</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>Tm$^{168}$</td>
<td>0.97</td>
<td>M</td>
<td>111</td>
</tr>
<tr>
<td>Lu$^{170}$</td>
<td>1.0</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>Lu$^{171}$</td>
<td>1.0</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>Hf$^{173}$</td>
<td>0.97</td>
<td>G</td>
<td>82, 112</td>
</tr>
<tr>
<td>Hf$^{175}$</td>
<td>0.97</td>
<td>M</td>
<td>83</td>
</tr>
<tr>
<td>Ta$^{176}$</td>
<td>0.94</td>
<td>G</td>
<td>86</td>
</tr>
<tr>
<td>Ta$^{177}$</td>
<td>0.87</td>
<td>G</td>
<td>86</td>
</tr>
<tr>
<td>Ta$^{179}$</td>
<td>0.40</td>
<td>G</td>
<td>84</td>
</tr>
<tr>
<td>Ta$^{180m}$</td>
<td>0.74</td>
<td>G</td>
<td>85</td>
</tr>
<tr>
<td>W$^{177}$</td>
<td>1.0</td>
<td>P</td>
<td></td>
</tr>
<tr>
<td>W$^{178} +$ Ta$^{178}$</td>
<td>1.54</td>
<td>G</td>
<td>86</td>
</tr>
<tr>
<td>W$^{181}$</td>
<td>0.37</td>
<td>G</td>
<td>87</td>
</tr>
</tbody>
</table>
Table III is a list of the various gamma rays that were used in establishing cross sections. The first, second, and third columns list the nuclide, the energy of the gamma ray which was used, and the abundance of that gamma ray, respectively. The fourth column is a reliability estimate similar in meaning to the corresponding column in the K x-ray table.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Gamma-ray energy (kev)</th>
<th>Gamma-rays per disintegration</th>
<th>Estimated reliability</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be$^7$</td>
<td>477</td>
<td>0.12</td>
<td>G</td>
<td>80</td>
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<tr>
<td>Mn$^{54}$</td>
<td>840</td>
<td>1.00</td>
<td>G</td>
<td>70</td>
</tr>
<tr>
<td>Co$^{58}$</td>
<td>805</td>
<td>1.00</td>
<td>G</td>
<td>70</td>
</tr>
<tr>
<td>Sr$^{85m}$</td>
<td>225</td>
<td>0.84</td>
<td>G</td>
<td>70</td>
</tr>
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<td>Sr$^{87m}$</td>
<td>388</td>
<td>0.78</td>
<td>G</td>
<td>70</td>
</tr>
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<td>Zr$^{88}$</td>
<td>395</td>
<td>0.97</td>
<td>G</td>
<td>70</td>
</tr>
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<td>Zr$^{89}$</td>
<td>913</td>
<td>0.99</td>
<td>G</td>
<td>70</td>
</tr>
<tr>
<td>Te$^{119}$</td>
<td>~640</td>
<td>~0.9</td>
<td>G</td>
<td>This work</td>
</tr>
<tr>
<td>Te$^{121m}$</td>
<td>213</td>
<td>0.91</td>
<td>G</td>
<td>94</td>
</tr>
<tr>
<td>Cs$^{129}$</td>
<td>375 + 420</td>
<td>0.52</td>
<td>G</td>
<td>This work</td>
</tr>
<tr>
<td>Gd$^{151}$</td>
<td>154</td>
<td>0.065</td>
<td>G?</td>
<td>81</td>
</tr>
<tr>
<td>Gd$^{153}$</td>
<td>~102</td>
<td>0.56</td>
<td>G</td>
<td>113, 114</td>
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<tr>
<td>Hf$^{173}$</td>
<td>121</td>
<td>1.1</td>
<td>G</td>
<td>82</td>
</tr>
<tr>
<td>Hf$^{175}$</td>
<td>343</td>
<td>0.86</td>
<td>G</td>
<td>83</td>
</tr>
<tr>
<td>Ta$^{176}$</td>
<td>202</td>
<td>0.063</td>
<td>G</td>
<td>86</td>
</tr>
</tbody>
</table>

C. Experimental Cross Sections

The following table lists the experimentally measured cross sections. The first column designates the nuclide for which the formation cross section was measured, and the second column lists the observed cross section. The third column designates whether the listed cross section corresponds to a
cumulative chain yield, designated by C, or an independent cross section, designated by I. In several cases, where the measured cross section lay between cumulative and independent, the result was corrected to completely cumulative or completely independent, either by indirectly measuring the parent cross section by using early and late separation times in successive bombardments, or by estimating the cross section for the parent by means of the semi-analytic "yield surface" to be described shortly. These cases are designated in the third column by placing the C or I in parentheses.

The fourth column is an estimate of the reliability of the observed cross section (exclusive of the error in the monitor cross section and in the assumed counting efficiencies) according to the following convention: A means the results are felt to be reliable to ± 10% or better, B means ± 25%, and a P classification corresponds to order-of-magnitude results.

Table IV. Experimental Cross Sections

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Cross section (mb)</th>
<th>Type of Yield</th>
<th>Reliability</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be$^7$ 24</td>
<td>24</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Na$^{24}$</td>
<td>8.7</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Na$^{22}$</td>
<td>&lt;1</td>
<td>C</td>
<td>F</td>
</tr>
<tr>
<td>P$^{32}$</td>
<td>4.4</td>
<td>I</td>
<td>B</td>
</tr>
<tr>
<td>P$^{33}$</td>
<td>3.5</td>
<td>C?</td>
<td>B</td>
</tr>
<tr>
<td>Mg$^{28}$</td>
<td>2.5</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Ca$^{45}$</td>
<td>1.5</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Ca$^{47}$</td>
<td>0.30</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Mn$^{52}$</td>
<td>0.80</td>
<td>?</td>
<td>B</td>
</tr>
<tr>
<td>Mn$^{54}$</td>
<td>6.3</td>
<td>?</td>
<td>B</td>
</tr>
<tr>
<td>Mn$^{56}$</td>
<td>2.0</td>
<td>?</td>
<td>B</td>
</tr>
<tr>
<td>Co$^{55}$</td>
<td>0.027</td>
<td>?</td>
<td>B</td>
</tr>
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<td>Co$^{58}$</td>
<td>1.4</td>
<td>I</td>
<td>P</td>
</tr>
<tr>
<td>Co$^{61}$</td>
<td>0.67</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Ni$^{57}$</td>
<td>&lt; 0.02</td>
<td>C</td>
<td>P</td>
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(continued)
<table>
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<tr>
<th>Nuclide</th>
<th>Cross section (mb)</th>
<th>Type of Yield</th>
<th>Reliability</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni$^{65}$</td>
<td>0.40</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Ni$^{66}$</td>
<td>0.12</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Cu$^{61}$</td>
<td>1.0</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Cu$^{64}$</td>
<td>3.6</td>
<td>I</td>
<td>A</td>
</tr>
<tr>
<td>Cu$^{67}$</td>
<td>0.64</td>
<td>?</td>
<td>A</td>
</tr>
<tr>
<td>Zn$^{62}$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Zn$^{69m}$</td>
<td>0.016</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Zn$^{72}$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Se$^{72}$</td>
<td>2.1</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Se$^{73}$</td>
<td>1.5</td>
<td>C?</td>
<td>B</td>
</tr>
<tr>
<td>Sr$^{82}$</td>
<td>3.2</td>
<td>(C)</td>
<td>B</td>
</tr>
<tr>
<td>Sr$^{85m}$</td>
<td>0.92</td>
<td>~I</td>
<td>B</td>
</tr>
<tr>
<td>Sr$^{87m}$</td>
<td>0.14</td>
<td>~I</td>
<td>B</td>
</tr>
<tr>
<td>Zr$^{88}$</td>
<td>6.8</td>
<td>C?</td>
<td>B+</td>
</tr>
<tr>
<td>Zr$^{89}$</td>
<td>6.9</td>
<td>(C)</td>
<td>A</td>
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<tr>
<td>Mo$^{90}$</td>
<td>1.5</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Mo$^{93m}$</td>
<td>1.4</td>
<td>I?</td>
<td>B</td>
</tr>
<tr>
<td>Mo$^{99}$</td>
<td>0.11</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Pd$^{100}$</td>
<td>2.3</td>
<td>C</td>
<td>B</td>
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<td>Pd$^{101}$</td>
<td>5.1</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Pd$^{103}$</td>
<td>6.8</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Ag$^{103}$</td>
<td>5.4</td>
<td>~C</td>
<td>A</td>
</tr>
<tr>
<td>Ag$^{105}$</td>
<td>6.5</td>
<td>(C)</td>
<td>B</td>
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<tr>
<td>Ag$^{106}$</td>
<td>0.9</td>
<td>I</td>
<td>P</td>
</tr>
<tr>
<td>Ag$^{112+113}$</td>
<td>$\leq 0.05$</td>
<td></td>
<td>P</td>
</tr>
<tr>
<td>Cd$^{107}$</td>
<td>6.3</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Cd$^{109}$</td>
<td>7.9</td>
<td>(C)</td>
<td>A</td>
</tr>
<tr>
<td>Cd$^{115}$</td>
<td>$\leq 0.004$</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
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<td>7.2</td>
<td>&lt;C</td>
<td>B</td>
</tr>
<tr>
<td>Sb$^{118}$</td>
<td>1.6</td>
<td>I</td>
<td>P</td>
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(continued)
<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Cross section (mb)</th>
<th>Type of Yield</th>
<th>Reliability</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sb 119</td>
<td>0.25</td>
<td>(I)</td>
<td>P</td>
</tr>
<tr>
<td>Sb 120m</td>
<td>≤ 0.5</td>
<td>I</td>
<td>P</td>
</tr>
<tr>
<td>Sb 122</td>
<td>≤ 0.03</td>
<td>I</td>
<td>P</td>
</tr>
<tr>
<td>Te 116 + 117</td>
<td>7.9</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Te 118</td>
<td>8.6</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Te 119</td>
<td>8.7</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Te 119m</td>
<td>&lt; 0.6</td>
<td>I?</td>
<td>P</td>
</tr>
<tr>
<td>Te 121</td>
<td>12.4</td>
<td>(C)</td>
<td>B</td>
</tr>
<tr>
<td>Te 121m</td>
<td>0.54</td>
<td>I?</td>
<td>B</td>
</tr>
<tr>
<td>Cs 131</td>
<td>≤ 0.5</td>
<td>I</td>
<td>B</td>
</tr>
<tr>
<td>Ba 126</td>
<td>6.1</td>
<td>C</td>
<td>B</td>
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<td>Ba 128</td>
<td>7.1</td>
<td>C</td>
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<td>Ba 129</td>
<td>11.2</td>
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<td>B</td>
</tr>
<tr>
<td>Ba 131</td>
<td>13.3</td>
<td>(C)</td>
<td>B</td>
</tr>
<tr>
<td>Ba 131</td>
<td>&lt; 2</td>
<td>(I)</td>
<td>B</td>
</tr>
<tr>
<td>Ce 133</td>
<td>&gt; 3.3</td>
<td>C</td>
<td>B</td>
</tr>
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<td>Ce 134</td>
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<td>Nd 140</td>
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<td>A</td>
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<td>Eu 145</td>
<td>14.9</td>
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<td>B</td>
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<td>Eu 147</td>
<td>18.5</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Eu 147</td>
<td>∼ 4</td>
<td>(I)</td>
<td>P</td>
</tr>
<tr>
<td>Gd 147</td>
<td>14.7</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Gd 149</td>
<td>13.2</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Gd 151</td>
<td>48.6</td>
<td>?</td>
<td>.B</td>
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<td>Gd 153</td>
<td>26.3</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Gd 146</td>
<td>?</td>
<td>8.8</td>
<td>A</td>
</tr>
<tr>
<td>Er 160</td>
<td>25.3</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Tm 165</td>
<td>24.7</td>
<td>~ C</td>
<td>B</td>
</tr>
<tr>
<td>Tm 167</td>
<td>27.3</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Tm 168</td>
<td>1.1</td>
<td>I</td>
<td>B</td>
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(continued)
**Table IV. (continued)**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Cross section (mb)</th>
<th>Type of Yield</th>
<th>Reliability</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lu$^{170}$</td>
<td>28.8</td>
<td>C</td>
<td>A</td>
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<tr>
<td>Lu$^{170}$</td>
<td>19</td>
<td>(I)</td>
<td>B</td>
</tr>
<tr>
<td>Lu$^{171m + 172}$</td>
<td>18.1</td>
<td>I</td>
<td>B</td>
</tr>
<tr>
<td>Hf$^{171}$</td>
<td>~ 13</td>
<td>C</td>
<td>P</td>
</tr>
<tr>
<td>Hf$^{173}$</td>
<td>13.4</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Hf$^{175}$</td>
<td>18.5</td>
<td>C?</td>
<td>A</td>
</tr>
<tr>
<td>Ta$^{176}$</td>
<td>~ 40</td>
<td>C</td>
<td>P</td>
</tr>
<tr>
<td>Ta$^{177}$</td>
<td>31</td>
<td>C</td>
<td>A</td>
</tr>
<tr>
<td>Ta$^{179}$</td>
<td>91</td>
<td>C</td>
<td>B</td>
</tr>
<tr>
<td>Ta$^{176 + 180m}$</td>
<td>129</td>
<td></td>
<td>B</td>
</tr>
<tr>
<td>Ta$^{180m}$</td>
<td>~ 46*</td>
<td>I</td>
<td>B</td>
</tr>
<tr>
<td>W$^{177}$</td>
<td>1.5</td>
<td></td>
<td>P</td>
</tr>
<tr>
<td>W$^{178}$</td>
<td>2.2</td>
<td></td>
<td>A</td>
</tr>
<tr>
<td>W$^{181}$</td>
<td>&lt; 1.5</td>
<td></td>
<td>B</td>
</tr>
</tbody>
</table>

*This cross section is based on beta counting through 20 mg/cm$^2$ of aluminum absorber. Perhaps the reported beta-decay: electron-capture branching ratio is high.*
V. DISCUSSION

A. General Appearance of Results

Inspection of the measured cross sections is facilitated if they are recorded on an N-versus-Z grid, similar to the "G.E. Chart of the Nuclides". Figures 6 to 10 represent sections of such a representation. The following observations may be made when the results are thus examined.

1. In the mass region $A = 22$ to $A = 67$, the cross sections seem to be at a maximum very near to the beta stability line, becoming smaller rapidly as one goes to either neutron-excess or neutron-deficient isotopes.

2. In the mass region $A = 72$ to $A = 153$, the cross sections are mostly of the cumulative chain-yield type, and appear most predominant on the neutron-deficient side of the beta-stability line. In places where several cross sections have been measured in the same region, the cumulative cross sections are highest for isotopes near the beta-stability line, where the beta-decay chains are longest, and become smaller rather slowly as one proceeds to more neutron deficient isotopes. The cross sections for neutron-excess isotopes, when measured, appear to be quite small.

3. In the mass region $A = 160$ to $175$, the observed nuclides are usually very neutron-deficient and have rather high cumulative cross sections. Above mass 175 the cross sections rapidly increase; more will be said about this later.

Evidently the situation is one in which the independent cross sections form a yield surface in the form of a long, narrow mountain, the crest of which approximately follows the beta-stability line up to $A \sim 70$, and then deviates further and further to the neutron-deficient side, until above $A \sim 150$, it is no longer possible to infer its position by the measurements at hand.
Fig. 6. Experimental cross sections (in millibarns) for the mass region $A = 32$ to 69.

In Figures 6 to 10: independent cross sections are underlined; cross sections in parentheses are estimates. The unbroken curve represents the beta-stability line.
Fig. 7. Experimental cross sections (in millibarns) for the mass region $A = 72$ to 109.
Fig. 8. Experimental cross sections (in millibarns) for the mass region $A = 100$ to $134$. 
Fig. 9. Experimental cross sections (in millibarns) for the mass region $A = 126$ to $160$. 

MU-14103
Fig. 10. Experimental cross sections (in millibarns) for the mass region $A = 160$ to 180.
B. Analytic Treatment of Results

Rudstam\textsuperscript{17} has treated the yield surfaces resulting from spallation reactions on targets of \( A = 51 \) to \( 75 \) by fitting the observed cross sections to the following formula,

\[
\ln \sigma (A,Z) = PA-Q-R (Z-SA)^2,
\]

where \( P, Q, R, \) and \( S \) are adjustable parameters independent of \( A \) and \( Z \), and \( \sigma (A,Z) \) is the measured independent cross section.

The data of this study cannot be readily fitted to Rudstam's formula for two reasons. His formula applies only to independent cross sections, and most of the cross sections in this work are cumulative. Moreover, preliminary study of the data reveals that \( P \) and \( S \) are not independent of \( A \) over the wide range of mass numbers involved in this work.

Still, it is of interest to fit the data to a semi-analytic "yield surface" in order to obtain a more exact idea of the yield distributions, and to estimate unmeasured yields. This was done in two separate sections; the first section included the data from \( A = 32 \) to \( A = 67 \), and the second section included the data from \( A = 72 \) to \( A = 153 \). The manner in which the surface was fitted for each mass range will be briefly described.

The first step in either case was to establish a preliminary curve \( \sigma_A \) versus \( A \) covering the mass range 24 to 170, where \( \sigma_A \) is the summed independent cross sections for all isotopes of mass number \( A \). It was possible to estimate values of \( \sigma_A \) for several values of \( A \) by suitable inspection of the data, and by making reasonable inferences concerning unmeasured yields. When these estimates were plotted on a graph, it proved possible to draw a smooth curve very satisfactorily through the points. This curve was used as a starting point for treating both sections mentioned above.

1. Treatment of Data for \( A = 72 \) to \( A = 153 \).

Twenty-five cumulative cross sections in this region were thought to be sufficiently reliable to include in the surface-fitting treatment. Three others, Ag\textsuperscript{103}, Sb\textsuperscript{117}, and Te\textsuperscript{117}, were rejected because possible
interfering isotopes of nearly the same half lives have been reported in each case. The independent cross sections were too few and too unreliable to do better than serve as checks on the final conclusions.

It was desirable to retain the sense of the Rudstam parameters in order to facilitate comparison with other work, and therefore the following assumptions were made:

(a). The independent cross sections were assumed to obey a modified, Rudstam-type equation,

\[ \sigma(A,Z) = k \sigma_A(A) e^{-R \left[ S - S(A)A \right]^2} \]  

(1)

where \( k \) is a number such that one has

\[ \sum_{Z=\infty} -R \left[ Z - S(A)A \right]^2 = 1. \]

(See below for further remarks.)

(b). \( R \) is assumed to be independent of \( A \). This is a risky assumption, but the data proved to be relatively insensitive to \( R \), because no measurements were available far out on the wings of the isobaric yield curves. Actually, \( R \) is expected to reflect the isobaric curvature of the parabolic mass surface, which varies approximately as \( \frac{1}{A} \).

(c). \( k \) is assumed to be independent of \( A \). Actually, \( k \) varies about as \( \frac{1}{\sqrt{R}} \).

(d). The summation,

\[ \sigma_c(A,Z) = \sum_{Z=\infty} -R \left[ Z - S(A)A \right]^2 \]

where \( \sigma_c(A,Z) \) represents the cumulative chain yield at \( A \) and \( Z \), can be replaced by the integral,

\[ \sigma_c(A,Z) = \int_{\infty}^{Z-1/2} k \sigma_A(A) e^{-R \left[ Z - S(A)A \right]^2} dZ. \]
The limit of integration must be \( Z - 1/2 \) instead of \( Z \), because the nature of the data requires that summation be performed for exact integral values of \( Z \), while if one imposes a smoothed curve for the independent yield distribution, it is necessary, in principle, to calculate the value for \( \sigma(A,Z) \) by integrating the smooth curve from \( Z + 1/2 \) to \( Z - 1/2 \). It should be mentioned, in passing, that the above summations and integrals are set up for electron-capture, positron-decay chains in neutron-deficient nuclides.

\( (e) \), \( S(A) \) and \( \sigma_A(A) \) were assumed to be smooth functions of \( A \), with no sudden breaks or sharp corners. As will be seen in the next section, this assumption is plausible if extensive nucleon evaporation is important in the production of the final distribution of independent cross sections.

Using the above assumptions, it is possible to write

\[
 f(A,Z) = \frac{\sigma_c(A,Z)}{\sigma_A(A)} = k \int_{\infty}^{Z - S(A)A - 1/2} e^{-R[Z - S(A)A]^2} d[Z - S(A)A].
\]

If the function \( S(A) \) is available, one may calculate \( Z - S(A)A \) for each measured \( \sigma_c(A,Z) \), and if the \( Z - S(A)A \) are then plotted against the corresponding values of \( f(A,Z) \), which may be calculated using the experimental \( \sigma_c(A,Z) \) and the \( \sigma_A(A) \) curve already described, the resulting curve should represent a plot of the right side of Equation 2 versus \( Z - S(A)A \).

A preliminary curve of \( S(A) \) versus \( A \) was obtained from the data, using methods of judgment and inference analogous to those used in obtaining the preliminary function \( \sigma_A(A) \), and the above-described plot was constructed. In appearance, it was much like Fig. 11, except that the points were more scattered. Theoretical curves of \( I_R \) versus \( x \), corresponding to the equation
Fig. 11. Graphical presentation of data fit to Equation (2). Experimental points for which A is between 72 and 131 are represented by solid circles. Those for which A is between 134 and 153 are represented by open circles. The smooth curve is a plot of $I_R$ for $R = 0.55$. 
were constructed assuming different values of \( R \), and a "best" value of \( R \) was established by finding that value of \( R \) for which the total deviation of all the points from the constructed curve was at a minimum.

It was now possible to construct a new function \( S(A) \) based on the best \( R \). For each value of \( f(A,Z) \) less than 0.75, a corresponding \( Z - S(A)A \) was read from the theoretical curve. From these values a set of \( S(A) \) could be calculated, the points plotted, and a smooth curve constructed through the points. The resulting function differed somewhat from the preliminary function, but not seriously, and there was much less scatter in the points.

A new function \( \sigma_A^2(A) \) was constructed using the new \( S(A) \) and best \( I_R \). For each nuclide for which there was a measurement, a \( Z - S(A)A \) was calculated based on the new, smooth \( S(A) \) curve, and the corresponding value of \( I_R \) was read off. A set of values of \( \sigma_A \) were then calculated by the formula

\[
\sigma_A^2(A) = \frac{\sigma_{C}(A,Z)}{I_R},
\]

the resulting values were plotted as \( \sigma_A^2 \) versus \( A \), and a smooth curve was drawn through the points. Again, there were some differences between this \( \sigma_A^2 \) curve and the preliminary curve (in this region), but they were not serious.

With these new curves for \( S(A) \) and \( \sigma_A^2(A) \), the whole cycle was repeated, determining a new "best" \( R \), etc. In all, the calculation was repeated three times. The difference between \( R, S(A) \), and \( \sigma_A^2(A) \) in the second and third cycles was so small that further calculation was felt to be unnecessary. The final value of \( R \) was \( R = 0.55 \), and Figs. 12 and 13 illustrate the final functions \( S(A) \) and \( \sigma_A^2(A) \) respectively. Their use allows one to predict 85% of the data in this mass region to within \( \pm 25\% \).
Fig. 12. Graph of the function $S(A)$. The solid circles represent data which are thought to be more reliable than the open circles in determining the course of $S(A)$ [e.g., data for which $f(A,Z) < 0.75$, see Fig. 11]. The smooth curves were drawn "by eye" through the points, and were drawn separately for the regions $A = 32$ to $66$ and $A = 72$ to $153$, because the data in each region were analyzed separately.
Fig. 13. Cross sections as a function of product mass number, for the interactions of tantalum with 5.7-Bev protons. Closed circles represent the data which are thought to be more reliable (e.g., experimental data for which $f(A,Z) > 0.75$, see text). The triangles represent values of $\sigma_A(A)$ which include estimated values for unmeasured yields (see Fig. 10).
2. Treatment of Data for $A = 32$ to $A = 67$.

Fifteen measured cross sections were employed for the surface-fitting treatment in this mass region, all of them representing independent or nearly independent yields. A preliminary examination of the data revealed the following facts.

(a). The data could not be very satisfactorily treated by assuming $S$ independent of $A$.

(b). The $\sigma_A(A)$ function in this region is nearly independent of $A$.

(c). The isobaric yield curve is apparently not quite Gaussian in shape (not quite parabolic on a semilog plot), appearing to skew somewhat to the neutron excess isotopes (Fig. 14).

The data were fitted to Equation (1) using a simplified graphical method analogous to the procedure already described above. Figures 13 and 12 illustrate the functions $\sigma_A(A)$ and $S(A)$ that were obtained. The corresponding value of $R$ was 0.93.

The $S(A)$ functions for the two regions do not join smoothly, although the error of closure is not great. Such a mismatch is not surprising because this treatment causes $R$ to change suddenly from 0.93 to 0.55 in going from $A = 67$ to $A = 72$, which is somewhat unrealistic. The $\sigma_A(A)$ curve shows no definite break, probably because $\sigma_A$ is relatively insensitive to variation in $R$.

Before leaving this section, it should be emphasized that the meaning of the various parameters and functions is limited by the assumptions involved. Many different mechanisms must operate to yield products in the reactions of tantalum with Bev protons, and one can easily conceive of situations in which unexpected deviations in the course of $S(A)$ and in the value of $R$ (which governs the width of the isobaric Gaussian) may occur. As an extreme example one may cite the case of the reactions of Bev protons with uranium, where the isobaric yield curve in the mass region $A \sim 140$ seems to display two maxima with a valley between,
Fig. 14. Graphical presentation of the fit of the data, in the mass region $A = 32$ to 69, to Equation (1), assuming that $\sigma_A(A)$ is approximately constant in this region (within the experimental error, this assumption appears to be justified). The smooth curve represents the function expected for $R = 0.93$, with the ordinate adjusted to fit the data.
The $\sigma_A(A)$ function was extended below $A = 32$ to $A = 24$ by utilizing the measured cross sections for Na$^{22}$, Na$^{24}$, and Mg$^{25}$ to estimate the small contribution of Mg$^{24}$. This was added to the cross section for Na$^{24}$ (which is a cumulative cross section) to obtain the total cross section for $A = 24$.

The $\sigma_A(A)$ function was extended above $A = 153$ by estimating reasonable values for unmeasured cross sections. The value for the (p,2p) product, Hf$^{180}$, is based on the ratio of the (p, pn) and (p, 2p) cross sections measured by Caretto and Friedlander$^{42}$ on Ce$^{142}$ at 3 Bev. The appearance of trends in the cross sections for reactions of the type (p, pxn) and (p, 2pxn), together with similar trends observed by Nethaway$^{43}$ with indium at 2 and 6 Bev and Barr$^{49}$ with copper at 5.7 Bev, was made the basis for other estimates. These estimates are included in Fig. 10.

C. Reaction Mechanisms

The various features of the data, which were brought out in the preceding section, can now be made the basis of a discussion of the possible mechanisms involved in the reactions of tantalum with 5.7-Bev protons.

An over-all idea of the pattern assumed by the products of these reactions can be gained from the complete $\sigma_A(A)$ function (Fig. 13), and from Figs. 15 and 16.

Recall that Fig. 13 is a plot of the total reaction yield per mass number versus mass number. Four main features are immediately apparent from this figure: (1) a steep rise within a few masses of the target mass; (2) a hump or step between mass numbers 120 and 175; (3) a broad, shallow valley between mass numbers 24 and 120; and (4) a steep "toe-up" from mass 24 downward.

The unbroken curve in Fig. 15 demonstrates the inferred positions of the isobaric yield maxima of the independent cross sections. It was obtained by using the final function $S(A)$ to solve the
Fig. 15. The relation between the isobaric yield maxima and the beta-stability line (see text).
Fig. 16. Detail of the neutron-deficient aspect of the isobaric maxima of independent yields (see text).
equation $Z = S(A)$. The course of the line of beta stability is represented by the dashed line. The dotted lines indicate the approximate width of the isobaric yield curves at half-maximum, and were calculated from the experimental values of $R$. The most noticeable features of this figure are as follows: (1) as the $Z$ of the products increases, the maximum of the isobaric yield curve becomes more and more neutron-deficient; (2) the width of the isobaric yield curve varies relatively slowly, being, as expected, narrower at low mass numbers than at high mass numbers.

Figure 16 permits a fairly detailed inspection of the neutron-deficient aspect of the inferred isobaric (independent) yield-curve maxima. In the figure, $Z_{\text{max}}(A)$ is that $Z$, for a given mass $A$, at which the isobaric yield curve has its maximum value. $Z_{A}$ is the most stable $Z$ for a given mass $A$, exclusive of odd-even effects but retaining the breaks at the "magic numbers" of protons and nucleons. These values of $Z_{A}$ were taken from Coryell.\textsuperscript{116} The experimental data were used to calculate a set of $Z_{\text{max}}(A)$, using the $\sigma(A)$ curve and the appropriate experimental value of $R$. The corresponding value of $Z_{A}$ was subtracted from each value of $Z_{\text{max}}(A)$, and the points were plotted as in the figure. It is of some interest to compare these results with theoretical predictions based on the assumption of extensive nucleon evaporation from very excited nuclei. Preliminary work with the equations governing nucleon evaporation indicated that such a predicted function would involve rather lengthy calculations, an effort which would be of rather doubtful value because Friedlander et al. are already currently engaged in Monte Carlo calculations of nucleon evaporation at high excitation energies. However, Le Couteur\textsuperscript{14} has performed the calculations for a nucleus of $A = 100$, for various excitation energies. A value derived from his work, representing the average evaporation end product of a nucleus originally excited to ~300 Mev, is plotted in the figure as a four-pointed star. Also, it is not too difficult to compute a function representing an approximate upper limit on the $Z_{\text{max}} - Z_{A}$ which one would expect to fine experimentally, by means of the simplified method outlined by Halpern et al.,\textsuperscript{117}
utilizing the neutron and proton emission rates calculated by Weisskopf and the semi-empirical mass-equation parameters given by Coryell. This function is represented by the unbroken curve, and is that value of $Z - Z_A$ at each $A$ for which the neutron-emission rate is equal to the proton-emission rate, for a nuclear excitation of about 20 Mev. It should be corrected downward because (1) most of the evaporations take place for nuclear excitation energies $>>$ 20 Mev, where the Coulomb barrier against proton emission is much less effective, and (2) alpha particles, deuterons, and other complex particles are also evaporated in addition to neutrons and protons. The dashed curve is merely 0.5 times the unbroken curve, and serves to facilitate the comparison of the data with the functional trend of the calculation.

Above $A \sim 60$ the points seem to follow the trend indicated by the dashed line, except that they appear to fall in clusters alternating above and below. This curious effect is easily explained. The breaks in the experimental function come at just the places where there are breaks in the $Z_A$ curve arising from the closing of the neutron and proton shells at the magic numbers $Z = 28$, $N = 50$, $Z = 50$, $N = 82$. The positions of the isobaric yield maxima are apparently relatively uninfluenced by the irregularities in binding energies found in nuclear ground states. One may draw the expected conclusion that nuclear shell structure has little influence on nuclear evaporation for excitation energies of the order of hundreds of Mev. Whether there is a corresponding lack of even-odd influence cannot be ascertained from the data at hand. The over-all picture in this mass region is consistent with the view that enough nuclear evaporation has contributed to the final observed distribution of products that the original isobaric distribution of evaporation progenitors is nearly obliterated.

In the mass region $A \sim 30$ to 60, the isobaric yield maxima fall very near the line of beta stability, or perhaps very slightly to the neutron-excess side. This may indicate that the evaporation precursors had an excess of neutrons, since one expects, from the figure, to find the maxima in this region to be roughly 0.3 unit of $Z$ to the neutron-deficient side. Since it is difficult to imagine how a pure cascade-evaporation mechanism
could lead to such a product distribution, this observation may provide
evidence that another mechanism is responsible for the evaporation pro-
genitors in this region, as we shall see shortly.

The over-all pattern described by the three figures just examined
is very similar to the one observed by Wolfgang et al.,\textsuperscript{33} in the reactions
of lead with 3-Bev protons, and also there are definite points of simi-
larity between the results of this work and the pattern observed by Barr\textsuperscript{49}
in the reactions of copper with 5.7-Bev protons. In each case the bulk of
products is to be found relatively near the target nucleus in mass number;
there is a prominent toe-up at very low mass numbers; and the mass numbers
in between are represented by a broad distribution of products, with maxi-
mum cross sections of the order of a few millibarns.

The reported mechanisms with which one might try to explain the
observed patterns are (1) the cascade-evaporation process, (2) fission,
and (3) fragmentation.

1. The cascade-evaporation process.

This was the first mechanism that was proposed to describe nuclear
reactions in this energy range. Heisenberg\textsuperscript{3} and Williams\textsuperscript{4}
proposed knock-
on mechanisms that were rather similar to the more recent ideas concern-
ing the cascade event, to account for the main features of the then newly
discovered large "stars" in nuclear emulsions which had been exposed to
cosmic rays, and calculated the bombarding energies beyond which the Bohr
compound-nucleus\textsuperscript{10} theory would fail. Serber\textsuperscript{12} proposed a similar, more
complete theory in order to explain the characteristics of nuclear re-
actions in the hundreds-of-Mev range, soon after the Berkeley 184-inch
synchrocyclotron came into operation.

Briefly, according to this mechanism, the high-energy projectile
interacts with the nucleus by colliding with individual nucleons in its
midst, and the recoiling collision partners also undergo similar collis-
ions, causing a number of "cascade" particles to emerge from the nucleus,
generally in the forward direction. The residual nucleus is, in general,
excited and "evaporates" more nucleons. A wide range is possible in the
number of cascade particles, and in the distribution of excitation energies
left in the residual nuclei, for a given bombarding energy, and this leads to the wide distribution of products that is actually observed. Extensive theoretical work has been done on this mechanism. The cascade process has been treated quantitatively by Monte Carlo calculations, notably by Goldberger,\textsuperscript{13} Turkevich and co-workers,\textsuperscript{45} McManus and Sharp,\textsuperscript{44} and Rudstam.\textsuperscript{17} The evaporation process has been treated analytically by LeCouteur,\textsuperscript{14} Yamaguchi,\textsuperscript{15} Jackson,\textsuperscript{17} and others. Of late, Monte Carlo methods have also been applied to describe the evaporation process, by Rudstam\textsuperscript{17} and G. Friedlander et al.\textsuperscript{46} For bombarding energies below about 500 Mev, where meson production is not yet important, and above about 100 Mev, where the mean free path of the bombarding nucleon in nuclear matter becomes long enough to make the cascade assumption valid, there is good agreement between these calculations and experimental results. The cascade calculations by Turkevich et al.\textsuperscript{45} and the evaporation calculations by Friedlander et al.\textsuperscript{46} comprise the only sets that are really applicable in the Bev region, and conclusions derived from them will be used in connection with various points in the ensuing discussion.

It is probably safe to ascribe the bulk of the observed yield in the mass region $A \sim 120$ to 175 to the cascade-evaporation mechanism. This mechanism must also contribute materially to masses less than 120 and greater than 175, but probably not predominantly, hence the limits are somewhat fuzzily defined. Probably the bulk of products below $A \sim 8$ are also due to the cascade-evaporation process, being the knocked-on and evaporated particles themselves. Here, too, the demarcation is quite indefinite, as we shall see later.

The Monte Carlo calculations predict a broad maximum in the yield distribution, occurring at about 15 mass numbers less than the target mass, for a bombarding energy of 1.8 Bev, and one would expect that at 5.7 Bev the calculated maximum would be even further from the target mass. No such maximum is discernible in the data. Barr\textsuperscript{47} reports a similar non-observation of the expected maximum in the reactions of copper with 5.7-Bev protons. Since the calculations were performed assuming a square-well potential containing a degenerate Fermi gas, the contribution from
the smaller cascades and lower-energy transfers may have been underestimated. If a more realistic picture of the nucleus were adopted, in which the nucleon density tapers off gradually with increasing radial distance, agreement with experiment should be improved. Another reason for the nonagreement with theory could lie in the competitive "fragmentation" process, which is discussed in the next section.

2. The fission and fragmentation processes.

The long, shallow valley in the mass range A ∼ 24 to 120 includes the products that are expected to be due either to fission, or to the recently identified fragmentation. As before, the exact boundaries between this region and the cascade-evaporation regions on either side are rather ill-defined.

A process identified as fission has been observed by Nervik and Seaborg in the reactions of tantalum with 340-Mev protons, and confirmed by Kruger and Sugarman with 450-Mev protons. In addition,Perfilov et al. have observed fission events in tungsten-loaded nuclear emulsions exposed to proton beams of 460 and 660 MeV. Nervik and Seaborg report the total fission cross section of tantalum at 340 Mev to be about 4 millibarns. Their fission-yield distribution forms a definite, if broad, peak with the maximum occurring at about mass number 83.

The same mass region at 5.7 Bev reveals a totally different distribution of products (Fig. 17). The integrated cross section of this mass region is 800 mb (or if it is binary fission, 400 mb), which is 100 times as large as the corresponding cross section at 340 MeV. There is no peak discernible; mass number 83 is actually near the bottom of a valley, instead. Also, the bulk of the products in this region are considerably more neutron-deficient than was the case at 340 MeV.

Wolfgang and co-workers observed a very similar distribution in the reactions of lead with 3-Bev protons, and were led to postulate that a process other than fission contributes materially to the observed yield pattern in this region. This mechanism, which they have named
Fig. 17. Comparison of product distributions in the reactions of tantalum with 5.7-Bev protons, and with 0.34-Bev protons (from Nervik and Seaborg).
"fragmentation," is thought to be associated with the increasing probability for the production and reabsorption of mesons as the bombarding energy is raised. Briefly, the mechanism is one in which the bombarding proton enters the nucleus, and while initiating the usual cascade event undergoes one or more proton-nucleon collisions which create mesons; the mesons in turn have short mean free paths and are usually reabsorbed, perhaps having undergone some scattering collisions beforehand. The result is that a limited zone of the nucleus becomes highly excited while the rest of the nucleus remains relatively cool. It seems reasonable to expect that under such conditions the nucleus can break into two fragments, the relative sizes of which depend on the location and size of the excited zone. Obviously this must happen very quickly, before the excitation energy can be dissipated to the rest of the nucleus. The fragments would be expected to have about the same neutron-to-proton-ratio as the target nucleus and to be highly excited. The slight neutron-excess character of the products in the mass region $A \sim 22$ to 60, which was observed in this work (Fig. 16), and the corresponding effect seen in the 3-Bev proton bombardments of lead, are consistent with this view.

Other evidence pertaining to such a process has been put forward in the recent work of Porile and Sugarman. They analyzed excitation functions up to 3 Bev for the formation of various products appearing in the proton bombardment of bismuth, in terms of the deposition-energy spectrum predicted by the Monte Carlo calculations of Turkevich et al. One of the results they obtained was that a typical fission product, such as Cu$^{67}$, has a probability of formation as a function of deposition energy which attains a maximum at 150 to 200 Mev and becomes vanishingly small above $\sim 300$ Mev, when the excitation function is analyzed for bombarding energies up to 450 Mev. When this formation-probability function is then used to predict the excitation function up to 3 Bev, the results are too low. This observation was interpreted to mean that a new mode of formation of Cu$^{67}$ appears at Bev bombarding energies. Its excitation function rises rapidly from threshold at bombarding energies of about 500 Mev to become equal to the contribution from fission at about 1.3 Bev, and to surpass it thereafter. They have identified this process with the same
mechanism that Wolfgang et al. have termed "fragmentation." From an analysis of several formation products across the fragmentation region, they have concluded that the cross sections from fragmentation are largest at the higher and lower mass numbers in this region, and reach a minimum for mass numbers of about half the target mass.

The possibility must not be dismissed, however, that fission also contributed materially to the formation of the products appearing in this region. The $\frac{Z^2}{A}$ for excited nuclei will not increase much beyond a certain value at any given mass number, determined by the point at which the proton- and neutron-emission rates are equal, and this consideration might lead one to suspect that there is a certain maximum total fission cross section for tantalum. However, it is to be expected that the average excitation energy of a given group of fissioning nuclei will, in general, increase with bombarding energy. The depression of nuclear surface tension will then become important in lowering the threshold for fission, and, according to Morrison, whose discussion is based on the calculations of Fujimoto and Yamaguchi, and Bagge, for really great excitation energies where the surface tension becomes small and the amplitude of surface distortions becomes very large, fission may readily occur in a wide variety of modes almost uninfluenced by the small surface energy, and one would expect a nearly flat distribution of fission-fragment masses. He goes further to state that one might preferably expect non-equilibrium local heating which would "pinch off" relatively small pieces of nuclear matter. This would increase the contribution to the lower and higher portions of the fission region at the expense of the middle, and lead to a distribution not unlike that reported for fragmentation. Thus, one sees that these ideas allow a rather smooth transition from the fragmentation mechanism at one extreme to fission at the other, with a corresponding time scale governing the events ranging from the time required to develop a nuclear cascade and initiate fragmentation, through the time required to distribute the nuclear excitation energy throughout the nucleus, to be associated with the events characteristic of low surface tension, to events which compete with or follow nucleon evaporation.
3. Low-energy processes.

Mention should be made of the anomalous magnitude of the results in the mass region 175 to 180, representing products in the immediate neighborhood of the target nucleus. The integrated cross section for this region appears to be about 500 mb, although much of this figure comes from estimates of unmeasured cross sections. This is to be compared with the cosmic-ray results of Sinha and Das,\textsuperscript{125} who measured total nuclear cross sections with respect to both high-energy and high-energy-plus-low-energy interactions, using penetrating charged secondaries with an average energy of 4 Bev. They interpret their results by stating that when the bombarding particle comes within about \(0.15A^{1/3} \times 10^{-13}\) cm of the equivalent square-well radius of the nucleus, a low-energy interaction will take place. This corresponds to a cross section of about 400 mb for tantalum, which is of the same order of magnitude as the experimental integration. However, since the cosmic-ray experiment excludes both the evaporation of protons, and very small deflections of the bombarding particle, in measuring the low-energy interaction, its meaning in terms of subsequent nuclear reactions is not too well defined, but it does serve to indicate the order of magnitude of the extent of low-energy reactions.

There seems to be a distinct tendency for yields in this region to be substantially higher than simple cascade-evaporation theory would predict, not only in this work but in the work of others. Markowitz\textsuperscript{1} has done a systematic study of \((p,pn)\) cross sections at energies up to 3 Bev, and on target nuclei as heavy as tantalum, and reports that the cross sections he obtains are higher by a factor of two than his calculations, based on a simple knock-on model, would predict. Barr's\textsuperscript{49} measurements of the reaction products of copper with 5.7-Bev protons reveal the same trend when compared to the Monte Carlo calculations done by Turkevich et al.\textsuperscript{45} on copper.

To account for these results, it may be necessary only to do the theoretical calculations over, employing a diffuse nuclear edge. On the other hand, the corrections necessary seem larger than can be accounted for.
for in this way, especially with tantalum. It might be that important new reaction mechanisms that transfer only a small amount of energy to the target nucleus await identification.

Several possible mechanisms for such low-energy transfer have been mentioned. Markowitz suggests, but rejects, the possibility that inelastic meson-producing reactions, such as \((p,2n^+1)\), may contribute materially to cross sections of products in the region of the target nucleus. The mean free path of mesons in nuclear matter is so short that the meson would have to be formed very near the edge of the nucleus in order to escape.

Also it may be possible that Coulomb excitation contributes to the cross sections in this region. The level densities become very high in the region 6 to 30 Mev of nuclear excitation, while the energy of the passing proton is very much greater than this, so that it seems possible that perceptible reactions of the type \((p,p'xn)\) could occur this way. However, since the interaction time of the passing proton is very short, tending to offset the effect of the high level densities and available energy, one might not expect very high cross sections from this reaction.

Perhaps a more sensitive indicator of the high probability of despositions of small amounts of energy in heavy nuclei when they interact with Bev protons is to be found in the results of Shudde, who studied the interaction of uranium-238 with 5.7-Bev protons. His data, although fragmentary, display not only the typical broad, single hump one has come to expect from high-energy fission, but also, somewhat more to the neutron-excess side than this hump and in about the position at which one finds the yield distribution of low-energy fission, there appear in high yield the typical product nuclides one expects of low-energy fission. Shudde's results in the barium-lanthanum region are especially suggestive, because one sees a high yield for the very-neutron-deficient \(^{128}\text{Ba}\), a very small independent cross section for \(^{140}\text{La}\), but again high cumulative cross sections for the neutron-rich \(^{140}\text{Ba}\) and \(^{141}\text{La}\). The reason one expects from uranium fission somewhat greater sensitivity to the acquisition of small amounts of energy from the 5.7-Bev proton is that the threshold for fission in \(^{238}\text{U}\) is only
about 5 Mev where one ordinarily needs about 7 Mev to bring about a single neutron-out reaction.

Appropriate comparisons of $^{238}\text{U}$ fission induced by bombardments with Bev protons, and Bev neutrons, might prove very revealing to this aspect of the problem of the interactions of Bev particles with nuclei.

4. Total cross section.

The total reaction cross section was estimated to be 2.0 barns by integrating the $\sigma_A(A)$ curve (see Fig. 13) from $A = 90$ to $A = 180$, with the expectation that most of the products below $A = 90$ are fragmentation, fission, or evaporation products that have corresponding complementary products at $A > 90$. The resulting estimate actually represents a lower limit, although it is expected to be fairly close to the truth. The total cross section interpolated from the work of Sinha and Das $^{125}$ is 1.8 barns, which agrees to within the experimental error with this work.

5. Special reactions.

The formation cross sections for tungsten are of some interest because they can be the result of such special processes as charge exchange ($p, p'n'\alpha$) reactions, and the like. However, crude calculations indicate that even with the relatively thin targets (32 mg/cm$^2$ of tantalum and 28 mg/cm$^2$ of aluminum) used in the measurement of the tungsten isotopes, a total formation cross section for tungsten from secondaries could be appreciable. In particular, assuming that an average of 10 secondary protons is produced per primary interaction, the expected total production cross section for tungsten isotopes works out at be about 3 mb, which is comparable to the observed cross sections. Much thinner tantalum targets will be necessary to make a valid study of the direct formation of tungsten isotopes.

In conclusion, it can be said that at least two and very likely three or more processes can be identified among the reactions of 5.7-Bev protons with tantalum:
(a) the cascade-evaporation process, which accounts for more than half of the reaction products, including most of those that have $A < 7$ and $A > 120$;

(b) the fragmentation process and (or) other processes such as fission which account for the broad distribution of products between mass numbers 24 and 120 (it seems likely that the evaporation precursors of this distribution are highly excited and evaporate enough nucleons to render the cascade-evaporation products indistinguishable from the fragmentation and fission products in the mass region $A \sim 100$ to $140$);

(c) some unknown process or processes to account for the unexpectedly high yield of reaction products with mass numbers near that of the target nucleus; these must be processes in which the 6-Bev proton transfers only a very small amount of energy to the target nucleus.
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The radiochemical purification procedure adopted for each element is outlined below. With a few exceptions, these procedures are usually adaptations or modifications of those to be found in the widely available compendia by M. Lindner and J. Kleinberg. In all cases, references are given to indicate the sources of the procedures that were actually used.

The initial treatment of the target solution to bring about first separations of the elements to be purified has already been described in Section II, C. The following outlines begin with the crude preparations obtained at that point. The details of mounting the purified samples for counting have also been discussed in Section II, C, and will be omitted here.

**Beryllium**

Beryllium hydroxide was precipitated by adding ammonium hydroxide to the aqueous solution resulting from the tantalum-extraction procedure. The precipitate was dissolved in hydrochloric acid, and a drop of ferric nitrate solution was added. Ferric hydroxide was precipitated by pouring the solution into sodium hydroxide solution, and after a 5-min digestion, the ferric hydroxide was centrifuged. The ferric hydroxide precipitation was repeated twice, by adding a drop of ferric nitrate to the supernatant solution each time, digesting, and centrifuging. The resulting solution was acidified with hydrochloric acid, and beryllium hydroxide was precipitated by adding ammonium hydroxide. The precipitate was dissolved in 20% sodium hydroxide and beryllium hydroxide was precipitated by adding saturated ammonium chloride. The washed beryllium hydroxide was dissolved in hydrochloric acid, and put into a buffer solution 0.5 M in sodium sulfite and 1 M in sodium bisulfite. The beryllium was then extracted into 0.4 M thenoyl trifluoracetone in benzene. The organic phase was washed with water (twice), 8 M nitric acid (twice), water (once),
1 M sodium hydroxide (twice, ten seconds each), and water (once, briefly). The beryllium was back-extracted into a mixture of two parts concentrated formic acid to one part of concentrated hydrochloric acid. Beryllium hydroxide was precipitated by adding ammonium hydroxide, washed, and dissolved in nitric acid. The resulting solution was evaporated to dryness and dried for several hours at 225°C. The remaining solid was slurried in ethanol, and mounted for counting using the filter-chimney setup.

Sodium

The aqueous solution from the tantalum-extraction procedure was chilled in an acetone-and-ice bath, and sodium precipitated as sodium chloride by saturating the solution with hydrochloric acid gas. The sodium chloride was centrifuged, dissolved in water, cupric nitrate solution was added, and cupric sulfide was precipitated by passing in hydrogen sulfide. The supernate was boiled, ferric nitrate solution was added, and ferric hydroxide precipitated by passing in ammonia gas. Nickel nitrate and manganous nitrate solutions were added and nickel and manganese sulfides were precipitated by passing in hydrogen sulfide. A few drops of lanthanum nitrate solution were added and the suspension was centrifuged. A few drops each of calcium, zinc, and magnesium nitrate solutions were added to the supernate and the corresponding phosphates were precipitated by adding 0.1 M ammonium phosphate. The phosphate-precipitation step was repeated by adding barium and strontium nitrate solutions. The volume of the solution was reduced to a few ml by boiling, and a few drops each of potassium, rubidium, and cesium nitrate solutions were added. The solution was cooled in an ice bath and saturated with hydrogen chloride gas to precipitate sodium chloride. The sodium chloride was centrifuged, redissolved in ~3 ml of water, and reprecipitated by cooling in an ice bath and saturating with hydrogen chloride gas. The sodium chloride precipitation was repeated twice more. The final sodium precipitate was slurried in ether and mounted for counting.
Magnesium

The mixture of rare earth and alkaline earth fluorides and sulfates, precipitated directly from the target solution, was treated with 8 N nitric acid and centrifuged. The supernatant solution, containing the magnesium, was made ammoniacal, and disodium phosphate solution was added to precipitate magnesium phosphate. The precipitate was dissolved in a few ml of concentrated hydrochloric acid, cooled in an acetone and ice bath, and saturated with hydrogen chloride gas. The resulting precipitate (which contains the magnesium) was centrifuged, dissolved in a few ml of water, cooled in the acetone and ice bath, and the magnesium was again precipitated by saturating with hydrogen chloride. The magnesium chloride was dissolved in water, made 1.5 N in hydrochloric acid, and copper and antimony sulfides were precipitated by adding the appropriate carrier solutions and saturating with hydrogen sulfide.

Nickel and lanthanum nitrate solutions were added to the supernate, which was then made ammoniacal and saturated with hydrogen sulfide; the resulting precipitate was discarded. The acidic and basic sulfide precipitations were repeated once more. Ammonium acetate was added to the supernatant solution and the pH was adjusted to 4 with hydrochloric acid. Four percent ammonium oxalate solution was added, followed by a few drops each of calcium, strontium, and barium nitrate solutions. After centrifuging, the precipitate was discarded and the oxalate precipitation was repeated in the supernate. The final supernate was evaporated to dryness a few times with concentrated nitric acid and the residue was taken up in water. A few drops each of sulfuric acid and strontium nitrate solution were added, the pH was adjusted to ~6, and the resulting precipitate was centrifuged after standing a few minutes. The supernate was made ammoniacal, 0.1 M ammonium phosphate was added, and after standing a few minutes the resulting precipitate was filtered, washed, and mounted for counting.
The phosphorus was recovered from the aqueous solution resulting from the tantalum-extraction procedure by adding zirconium chloride solution to precipitate zirconium phosphate. The zirconium phosphate was dissolved in a few drops of hydrofluoric acid, 4 N nitric acid and ammonium molybdate reagent were added, and, after digestion in a hot water bath, the resulting precipitate was centrifuged and washed with water. The precipitate was dissolved in concentrated ammonium hydroxide, diluted, and a few drops of 30% hydrogen peroxide were added. The solution was acidified with concentrated hydrochloric acid and zirconium chloride was added to precipitate zirconium phosphate. The precipitate was dissolved in a few drops of hydrofluoric acid, 3 N hydrochloric acid, arsenic carrier and a few drops of aerosol solution were added, and the mixture was heated in the hot water bath and saturated with hydrogen sulfide for 15 minutes. The resulting suspension was centrifuged, lanthanum carrier was added to the supernatant solution, and the precipitated lanthanum fluoride was centrifuged. The supernate was made 4 N in nitric acid, ammonium molybdate reagent was added, and the mixture was heated in the hot water bath for a few minutes. The precipitated ammonium phosphomolybdate was centrifuged and washed with water. The zirconium phosphate precipitation, arsenic sulfide scavenge, and ammonium phosphomolybdate precipitation cycle was repeated once. The final ammonium phosphomolybdate precipitate was dissolved in concentrated ammonium hydroxide, 50% citric acid solution and magnesia mixture were added, and the solution was made alkaline with concentrated ammonium hydroxide. After standing, the magnesium ammonium phosphate was centrifuged, slurried with dilute ammonia, and mounted for counting using the standard filter-chimney setup.

It may be of interest at some time to remove phosphorus much more quickly from the target solution than the tantalum-extraction procedure permits. This may be done by making the target solution strongly acid with nitric acid immediately after dissolving the target foil, followed by the addition of an overwhelming excess of zirconium chloride solution. The phosphorus is precipitated incompletely as zirconium phosphate.
Calcium

The combined precipitate of fluorides and sulfates obtained from the target solution contains the calcium. Strontium and barium were also precipitated in the same step to help promote a more complete recovery of calcium. The precipitate was treated with a mixture of saturated boric acid, concentrated nitric acid, and several drops of sulfuric acid, and the suspension was diluted with water and centrifuged. The precipitate was washed with dilute sulfuric acid, and then metathesized twice for several minutes with strong sodium carbonate solution. The resulting precipitate was washed with water, dissolved with a few drops of hydrochloric acid, and iron carrier was added. Ferric hydroxide was precipitated by making the solution basic with ammonium hydroxide. The ferric hydroxide precipitation was repeated twice, by adding a few drops of iron carrier each time to the supernatant solution. The final supernate was made 0.1 M in hydrochloric acid, palladium and copper carriers were added, and hydrogen sulfide was passed through the heated solution to precipitate the acid insoluble sulfides. After boiling, nickel and cobalt carriers were added, the solution was made slightly basic with ammonium hydroxide, and the basic sulfides were precipitated by passing in hydrogen sulfide. Ammonium oxalate was added to the supernate and the resulting precipitate was washed with water. The oxalate precipitate was dissolved in a few ml of 3 M nitric acid, a large excess of white fuming nitric acid was added, and the solution was cooled in an ice bath to precipitate strontium and barium nitrates. The volume of the supernate was decreased to a few ml by boiling, more strontium and barium carriers were added, and the strontium and barium nitrate precipitation was repeated, and then repeated once more. The final supernate was reduced in volume to a few ml by boiling; diluted, made ammoniacal, and calcium oxalate was precipitated by adding ammonium oxalate. The calcium oxalate was dissolved in concentrated nitric acid and 1 M sodium bromate, boiled to near dryness, diluted with water, made ammoniacal, and calcium carbonate was precipitated by adding ammonium carbonate solution. The calcium carbonate
was dissolved in a small amount of nitric acid, diluted, lanthanum carrier was added, and lanthanum hydroxide precipitated by making the solution ammoniacal. Finally, the supernate was heated, and ammonium oxalate was added to precipitate calcium oxalate, which was filtered using the usual filter-chimney setup, washed with water and alcohol, and mounted for counting.

### Manganese

The manganese was recovered initially as precipitated manganous hydroxide by adding ammonium hydroxide to the aqueous solution resulting from the tantalum-extraction procedure. The precipitate was dissolved in dilute nitric acid and manganese dioxide was precipitated by adding an excess of solid potassium bromate and heating in the hot water bath. The manganese dioxide precipitate was dissolved in a few ml of concentrated hydrochloric acid and boiled to remove chlorine. The solution was then passed through a column, 6 mm by 10 cm, packed with Dowex A-2 anion-exchange resin. The eluate was boiled to reduce its volume, made 1 N in hydrochloric acid, silver carrier was added, and the precipitated silver chloride was removed by centrifugation. Acid-insoluble sulfides were precipitated by adding copper carrier to the solution, and saturating with hydrogen sulfide. The supernatant solution was evaporated to dryness a few times with concentrated nitric acid, water was added, and the solution was made 1 N in nitric acid. An excess of solid sodium bismuthate was added to oxidize the manganese to permanganate. The solution was adjusted to a pH of 10 with sodium hydroxide and iron carrier was added to precipitate ferric hydroxide, which was removed by centrifugation. The solution was made 1 N in nitric acid and a few drops of saturated oxalic acid solution were added to reduce the permanganate to the manganous state. Excess solid potassium bromate was introduced, and the mixture heated until manganese dioxide precipitated. The precipitate was washed with water, slurried with more water, and mounted for counting.
Cobalt

The aqueous solution resulting from the tantalum-extraction procedure was made ammoniacal and the cobalt was precipitated by passing in hydrogen sulfide. The precipitate was dissolved in nitric acid, and the solution was diluted, and cobalt hydroxide was precipitated by adding 10 M potassium hydroxide. The precipitate was washed with water and dissolved in 6 M acetic acid. The solution was diluted and potassium cobaltinitrite was precipitated by adding a few ml of a solution made by saturating 3 M acetic acid with potassium nitrite. The precipitate was washed with water, dissolved in concentrated hydrochloric acid, and boiled to remove oxides of nitrogen. Nickel carrier was added, followed by 10 M potassium hydroxide to precipitate nickel and cobalt hydroxides. The potassium cobaltinitrite precipitation was repeated. The precipitate was dissolved in concentrated hydrochloric acid, boiled nearly to dryness, and a few ml of 0.1 N hydrochloric acid were added. Copper and palladium carriers were added and the acid-insoluble sulfides were precipitated by passing in hydrogen sulfide. After centrifugation, the solution was boiled and the acid sulfide precipitation was repeated. The resulting supernatant solution was boiled to remove hydrogen sulfide, and iron carrier was added followed by a moderate excess of ammonium hydroxide to precipitate ferric hydroxide. After centrifugation the solution was acidified, and the ferric hydroxide precipitation was repeated twice more. The final supernatant solution was made 0.4 N in hydrochloric acid, and 15 grams of ammonium thiocyanate was added and dissolved (total volume, 35 ml). The cobalt thiocyanate complex was extracted into a mixture of amyl alcohol and ethyl ether, and the organic phase was washed twice with concentrated ammonium thiocyanate solution. The cobalt was back-extracted into 3 N ammonium hydroxide. The solution was saturated with hydrogen sulfide to precipitate cobalt sulfide. The precipitate was washed with water, slurried in more water, and mounted for counting using the filter-chimney arrangement.
The aqueous solution resulting from the tantalum-extraction procedure was made ammoniacal, and 5% dimethylglyoxime was added to precipitate the nickel. The nickel dimethylglyoxime was filtered, washed with water, and dissolved in a few drops of concentrated nitric acid. The solution was boiled and diluted with water, silver carrier was added, and a few drops of hydrochloric acid were added to precipitate silver chloride. Copper and palladium carriers were added to the supernatant solution, which was then heated in the hot water bath and saturated with hydrogen sulfide to precipitate the acid-insoluble sulfides. After centrifugation the solution was boiled to expel hydrogen sulfide, iron carrier was added, and excess ammonium hydroxide was added to precipitate ferric hydroxide. Dimethylglyoxime was added to the supernatant solution to precipitate nickel dimethylglyoxime, which was separated by filtration. The whole purification cycle was repeated once. The final nickel dimethylglyoxime precipitate was filtered directly in the filter chimney, washed with water and alcohol, and mounted for counting.

Copper

Copper sulfide was precipitated by passing hydrogen sulfide through the target solution. After washing with water, the copper sulfide was dissolved in nitric acid and diluted with water. Silver carrier was added, followed by a few drops of hydrochloric acid to precipitate silver chloride, which was removed by centrifugation. Iron carrier was added and ferric hydroxide was precipitated when an excess of ammonium hydroxide was added. After centrifugation to remove the ferric hydroxide, the solution was made slightly acidic with hydrochloric acid and saturated with sulfur dioxide. Cuprous thiocyanate was precipitated by cooling the solution in ice and adding potassium thiocyanate. The precipitate was dissolved in a few drops of nitric acid, diluted, and made slightly ammoniacal. Solid potassium cyanide was added, followed by arsenic, tellurium, and cadmium carriers, and
the solution was saturated with hydrogen sulfide. The precipitated sulfides were removed by centrifugation and the solution was made acidic with hydrochloric acid, boiled to expel hydrogen cyanide, and saturated with hydrogen sulfide. The precipitated copper sulfide was dissolved in nitric acid and the purification procedure was repeated through the precipitation of cuprous thiocyanate. The final cuprous thiocyanate precipitate was filtered, washed, and mounted for counting.

Zinc$^{55}$

The aqueous solution resulting from the tantalum-extraction procedure was made feebly basic with ammonium hydroxide, and zinc sulfide was precipitated by saturating the solution with hydrogen sulfide. The washed precipitate was dissolved in a few drops of hydrochloric acid, diluted with water, and the resulting solution was made 2 M in hydrochloric acid. This solution was passed through a column, 6 mm by 10 cm, packed with Dowex A-2 anion-exchange resin. The column was washed with 2 M hydrochloric acid and the zinc was removed by passing 0.001 M hydrochloric acid through the column. Two drops of hydriodic acid (without preservative) were added to the eluate, which was then passed through another 6-mm-by-10-cm column packed with Dowex-50 cation-exchange resin. The resin was washed with 0.5 M hydrochloric acid containing a few drops of hydriodic acid and the zinc was removed by passing 2 M hydrochloric acid through the column. Iron and cadmium carriers were added to the eluate, together with a drop of sodium nitrite solution, and the mixture was made strongly basic with sodium hydroxide. The precipitate was removed by centrifugation and the supernatant solution was made 2 M in hydrochloric acid. Both column purification steps were repeated, except that the final Dowex-50 column was washed also with 0.1 M sulfuric acid and the zinc was eluted with 6 M sulfuric acid. Iron carrier was added to the eluate, and ferric hydroxide was precipitated by adding ammonium hydroxide. After centrifugation the solution was adjusted to the methyl red end point with sulfuric acid and a few ml of 0.1 M ammonium phosphate buffer (pH = 8)
were added. The solution was heated in a hot water bath until a precipitate formed. The precipitate was filtered, washed with water and alcohol, and mounted for counting.

Selenium

The radiochemical purification of selenium was almost identical to that for tellurium (which see). The difference was that whenever selenium and tellurium were separated, the tellurium, which was introduced as carrier to aid in the purification, was discarded and the selenium was retained for further purification.

Strontium

The radiochemical purification of strontium was identical to that of calcium (which see) up to the point at which strontium was separated from calcium by precipitating strontium nitrate with fuming nitric acid. The strontium nitrate was taken up in water, iron carrier was added, and ferric hydroxide was precipitated by adding a slight excess of ammonium hydroxide. After centrifuging to remove the precipitate, ammonium carbonate was added to the supernatant solution to precipitate strontium carbonate. The strontium carbonate was dissolved in a few drops of nitric acid, and the strontium nitrate precipitation, ferric hydroxide scavenge, and strontium carbonate precipitation were repeated. The strontium carbonate was dissolved in dilute nitric acid, barium carrier was added (if not already present), the solution was made about 1.5 M in ammonium acetate buffered to pH 5, 1.5 M sodium chromate solution was added, and the precipitated barium chromate was removed by centrifugation. The barium chromate precipitation was repeated twice. Strontium carbonate was precipitated by making the solution basic with ammonium hydroxide and adding ammonium carbonate solution. The strontium carbonate was dissolved in dilute nitric acid and the strontium carbonate precipitation repeated. The strontium carbonate was filtered in the filter chimney, washed with water and alcohol, and mounted for counting.
Zirconium

The radiochemical purification of zirconium is identical in every respect to that of hafnium (which see). The only difference is that in the final column separation step, zirconium is eluted first and hafnium second.

Molybdenum\(^{55}\)

When hydrogen sulfide was passed through the target solution for twenty minutes or so, molybdenum was found in the precipitate. It was considered advisable to add some copper carrier to the target solution, in order that the precipitated copper sulfide would help bring down the molybdenum, which tends to form a colloidal suspension. The precipitate was dissolved in concentrated nitric acid and subsequently evaporated to near dryness a few times with concentrated hydrochloric acid. The residue was taken up in 6 M hydrochloric acid and passed through a column 6 mm by 10 cm packed with Dowex A-2 anion-exchange resin. The column was washed, first with 0.1 M hydrochloric acid (the copper appears in the eluate), and then with 3 M ammonium hydroxide. The molybdenum was removed by passing 6 M sodium hydroxide through the column. The eluate was made just acidic with concentrated nitric acid, a few drops each of ferric, zirconium, and tellurium carriers, 0.3% aerosol, and 1 drop of 1 M sodium nitrite solution were added, and the solution was adjusted to pH 10 with concentrated ammonium hydroxide. The resulting suspension was centrifuged, discarding the precipitate, and the supernate was re-acidified with concentrated nitric acid. Ferric and zirconium carriers were again added, and the precipitation of hydroxides was repeated. The supernatant solution was acidified to pH 5.5 with concentrated acetic acid, warmed in a hot water bath, and 0.1 M lead nitrate was added. The precipitated lead molybdate was centrifuged, washed with water, slurried with more water and mounted for counting using the standard filter-chimney setup.
Palladium

The mixed sulfides of palladium and antimony which were precipitated from the target solution (see Section II, C) were dissolved in the minimum amount of a mixture of hydrochloric and nitric acids. A small amount of sulfuric acid was added and the mixture was evaporated until dense fumes of sulfur trioxide appeared. After cooling, the residue was taken up in water and slightly more than enough potassium iodide solution was added to precipitate all of the palladium as palladous iodide. The palladous iodide was boiled with concentrated hydrochloric acid, to which concentrated nitric acid had been added, until the solid material was dissolved and no more purple iodine fumes were emitted. This solution was then evaporated to dryness twice with concentrated hydrochloric acid, and finally the residue was taken up in 1 M hydrochloric acid. Silver carrier was added and the precipitated silver chloride was centrifuged. Iron carrier was added, the solution was made alkaline with ammonium hydroxide, and the precipitate was centrifuged. The supernatant solution was made 0.4 M in hydrochloric acid and a few ml of a saturated solution of dimethylglyoxime in ethanol was added. After digesting the suspension for a short time in a hot water bath, the palladium dimethylglyoxime was filtered. The precipitate was dissolved in a small amount of concentrated nitric acid, boiled nearly to dryness, boiled nearly to dryness again after adding concentrated hydrochloric acid, and finally taken up in 1 M hydrochloric acid. The silver chloride, ferric hydroxide, and dimethylglyoxime precipitations were carried out once again, and the final palladium dimethylglyoxime precipitate was washed with water and ethanol and mounted for counting using the standard filter-chimney setup.

Silver

Silver chloride was precipitated directly from the target solution upon the addition of hydrochloric acid. The precipitate was washed with water and dissolved in ammonium hydroxide. Iron carrier was added and the resulting suspension was agitated vigorously, then centrifuged, discarding the precipitate. The supernatant solution was saturated with hydrogen
sulfide, and the precipitated silver sulfide was washed with water and dissolved in hot, concentrated nitric acid. The solution was diluted with water, and silver chloride precipitated by adding hydrochloric acid. The entire purification cycle was repeated. The final silver chloride precipitate was filtered, washed with water and ethanol, and mounted for counting.

Cadmium

Cadmium sulfide was precipitated, together with copper sulfide, by passing hydrogen sulfide through the target solution (the presence of copper appears to be necessary to the precipitation of cadmium sulfide from concentrated hydrofluoric acid). The precipitate was dissolved in nitric acid and diluted with water, and silver carrier was added followed by hydrochloric acid to precipitate silver chloride. The precipitate was removed by centrifugation. Iron and indium carriers were added to the solution, which was then made ammoniacal; the resulting precipitate was discarded. Solid potassium cyanide was added to complex the copper, and cadmium sulfide was precipitated by saturating the solution with hydrogen sulfide. The cadmium sulfide was washed with water, dissolved in hydrochloric acid, boiled, and diluted with water. Acid-insoluble sulfides were precipitated by adding palladium and antimony carriers and saturating with hydrogen sulfide. After centrifuging, the supernatant solution was made ammoniacal to precipitate cadmium sulfide. The entire purification procedure was repeated. The final cadmium sulfide precipitate was washed with water, slurried with more water, and allowed to stand quietly in the filter chimney for 15 minutes (to allow most of the cadmium sulfide to settle out onto the filter paper disk) before the vacuum was applied to start actual filtration. The final cadmium sulfide deposit was then mounted for counting.
Antimony

The initial steps in the radiochemical purification of antimony were the same as those for palladium (which see), up to the point at which palladium was separated from antimony by precipitation as palladous iodide. After centrifugation, the supernatant solution was saturated with hydrogen sulfide to precipitate orange antimony sulfide. The antimony sulfide was dissolved in concentrated hydrochloric acid, boiled very briefly, and a few drops of each of the following carriers were added: arsenic, selenium, copper, molybdenum, and iron. The solution was diluted to 6 M hydrochloric acid, and saturated with hydrogen sulfide for several minutes, first in the hot water bath, and then as it cooled. The precipitated sulfides were removed by centrifugation. The supernatant solution was boiled briefly to expel hydrogen sulfide, diluted to 2 M in hydrochloric acid, silver carrier was added, and the silver chloride precipitate was centrifuged. The supernate was saturated with hydrogen sulfide, and the precipitated sulfides were centrifuged and washed with 2 M hydrochloric acid. The precipitate was slurried with concentrated ammonium hydroxide, a few drops of lanthanum carrier were added, and the suspension was saturated with hydrogen sulfide and centrifuged. The supernate was acidified to 2 M in hydrochloric acid, centrifuged, and the precipitate was washed with 2 M hydrochloric acid. (At this point the sulfide precipitate should be orange, except that in this work it was discolored a dark brown from traces of platinum which were picked up from the platinum utensils used in parts of the work. The platinum can be removed by leaching the precipitate in concentrated hydrochloric acid—the platinum dissolves much more slowly than the antimony—centrifuging, diluting the supernatant solution to 2 M in hydrochloric acid, and saturating with hydrogen sulfide.) The precipitate was dissolved in concentrated hydrochloric acid; tungsten, palladium, and molybdenum carriers were added; 2 ml of concentrated sulfuric acid were cautiously added; and the mixture was warmed in the hot water bath to expel most of the hydrogen chloride. The mixture was then taken to the first dense fumes of sulfur trioxide over a burner, cooled, and diluted with water.
After adding one drop of concentrated nitric acid, the solution was boiled, diluted to 24 ml with water, cooled in an ice bath, and 6 ml of 2% alpha-benzoinoxime in ethanol were added. Bromine water was added until the suspension was definitely colored, another 3 ml of the alpha-benzoinoxime were added, the mixture was allowed to stand in the ice bath for 15 minutes, and was then filtered. The filtrate was saturated with hydrogen sulfide and centrifuged. The precipitate was digested with concentrated hydrochloric acid and the resulting suspension centrifuged. The supernate from the digestion was diluted to 2 M in hydrochloric acid, saturated with hydrogen sulfide, centrifuged, and the precipitate was washed with 2 M hydrochloric acid. At this point, tin would be the only contaminant remaining. The antimony sulfide was dissolved in one ml of concentrated hydrochloric acid, a few drops of tin carrier were added, the solution was boiled briefly, and bromine water was added to slight excess; 0.5 ml each of concentrated sulfuric acid and concentrated hydrofluoric acid were added, and the solution was diluted to 15 ml and saturated with hydrogen sulfide for a few minutes. One ml of concentrated hydriodic acid was added, the solution was boiled for one minute, and again saturated with hydrogen sulfide. The resulting suspension was centrifuged, and the precipitate was put through the concentrated hydrochloric acid leach, dilution to 2 M hydrochloric acid, and reprecipitation with hydrogen sulfide as before. The final orange antimony sulfide was mounted for counting using the standard filter-chimney setup.

Tellurium

Tellurium sulfide was precipitated from the target solution by saturating with hydrogen sulfide. The tellurium sulfide was dissolved in concentrated nitric acid, selenium and arsenic carriers were added, and the mixture was evaporated nearly to dryness over a flame. The residue was dissolved in 8 M sodium hydroxide, iron carrier was added, and the precipitated ferric hydroxide was removed by centrifugation. The supernatant solution was made 3 M in hydrochloric acid, and saturated with sulfur dioxide gas to precipitate elemental tellurium, which was
centrifuged and washed with water. The precipitate was dissolved in concentrated nitric acid, evaporated to dryness, and taken up in concentrated hydrochloric acid. The solution was saturated with sulfur dioxide gas to precipitate the selenium, which was centrifuged. The supernate was diluted with water until it was 3 M in hydrochloric acid, again saturated with sulfur dioxide gas, and the precipitated tellurium was centrifuged. The entire procedure described above was repeated twice, except that the second time the selenium and arsenic carriers were omitted. The final tellurium precipitate was mounted for counting using the standard filter-chimney setup.

Cesium

Cesium was obtained from the target solution as a precipitate of cesium silicotungstate (Section II, C). The precipitate was washed with cold 6 M hydrochloric acid and dissolved in a few ml of 6 M sodium hydroxide. The resulting solution was warmed, cautiously acidified by adding an equal volume of 70% perchloric acid, and evaporated to the first dense fumes of perchloric acid. The mixture was cooled, diluted with water, and the precipitated silica and tungsten oxide were removed by centrifugation. The supernatant solution was again evaporated to fumes of perchloric acid, cooled in ice, and diluted with ice-cold absolute ethanol. The precipitated cesium perchlorate was recovered by centrifugation, and washed several times with ice-cold absolute ethanol to remove sodium. The cesium perchlorate was taken up in about 30 ml of cold 8 M hydrochloric acid, and 2 ml of 1/8 M silicotungstic acid was added. The mixture was allowed to stand for a few hours with occasional stirring, and was finally centrifuged to recover the precipitated cesium silicotungstate. The recovery of cesium as cesium perchlorate, as described above, was repeated. The final cesium perchlorate precipitate was slurried in absolute alcohol and mounted for counting.
The mixed precipitate of fluorides and sulfates obtained from the target solution was treated with several ml of a mixture of concentrated nitric acid and saturated boric acid containing a few drops of concentrated sulfuric acid. The resulting suspension was centrifuged, and the barium sulfate precipitate was washed with dilute sulfuric acid. The barium sulfate was metathesized to barium carbonate by heating in the hot water bath with a mixture of 8 M sodium hydroxide and saturated ammonium bicarbonate for several minutes. The metathesis was repeated once, and the barium carbonate was washed with dilute ammonium bicarbonate solution. The barium carbonate was then dissolved in 2 drops of concentrated hydrochloric acid, and strontium carrier was added, followed by 30 ml of ice-cold hydrochloric-acid-ether reagent (5 parts, by volume, of concentrated hydrochloric acid mixed with 1 part of ethyl ether). This mixture was kept in an ice bath with constant stirring for more than one minute, centrifuged, and the precipitate was washed with a few ml of the same ice-cold hydrochloric-acid-ether reagent. The precipitate was dissolved in a few ml of water, a few drops of iron carrier were added, ammonium hydroxide was added dropwise until the ferric hydroxide just persisted, four more drops of concentrated ammonium hydroxide were added, and the mixture was centrifuged. To the supernate was added another 30 ml of the hydrochloric-acid-ether reagent, and the entire procedure from that point was repeated twice, except that after the third precipitation of barium chloride, lanthanum carrier was used instead of the iron carrier. The final supernatant solution was carefully neutralized with hydrochloric acid, 2.5 ml of 6 M acetic acid and 10 ml of 3 M ammonium acetate were added, and the solution was heated in the hot water bath. To the hot mixture, 2 ml of 1.5 M sodium chromate were added dropwise, and the mixture was kept in the hot water bath, with stirring, for several minutes, after which it was cooled. The precipitated barium chromate was washed with water and alcohol, and mounted for counting using the standard filter-chimney setup.
The mixed precipitate of fluorides and sulfates from the target solution was leached with a mixture of concentrated nitric acid and saturated boric acid containing a few drops of sulfuric acid. The precipitate was removed by centrifugation, and the supernatant solution was diluted with water and made basic with ammonium hydroxide to precipitate cerium hydroxide. The cerium hydroxide was dissolved in a few drops of concentrated nitric acid and the solution was diluted with water. Lanthanum carrier was added, followed by a few drops of concentrated hydrofluoric acid, and the insoluble fluorides were recovered by centrifugation and dissolved in concentrated nitric acid and saturated boric acid. The solution was diluted, and made alkaline with ammonium hydroxide. The insoluble hydroxides were centrifuged and washed with dilute ammonium hydroxide. The precipitate was then dissolved in 10 ml of 6 M nitric acid, 0.5 gram of solid ammonium persulfate and several small crystals of solid silver nitrate were added, and the mixture was stirred until it became dark brown. The dark brown mixture was transferred to a separatory funnel and the cerium was extracted into a mixture of 30% (by volume) tributyl phosphate and 70% carbon tetrachloride, the extraction being done three times with 10-ml portions of the organic phase. The combined organic extraction phases (which should be distinctly yellow) were washed with a solution made by adding solid ammonium persulfate and several small crystals of silver nitrate to 20 ml of 6 M nitric acid and stirring until brown. Finally, the cerium was extracted back into an aqueous phase which was composed of about 0.5 gram of solid sodium bisulfite dissolved in about 30 ml of 1 M hydrochloric acid. A few drops of silver carrier were added to the aqueous layer after the extraction, and the precipitated silver chloride was centrifuged. The supernate was made alkaline with ammonium hydroxide and the cerium hydroxide was centrifuged and washed with dilute ammonium hydroxide. This entire procedure was repeated once more. Finally the cerium hydroxide was dissolved in a few drops of concentrated hydrochloric acid, diluted to a few ml with water, saturated oxalic acid was added, and the mixture was cooled for a
few minutes in an ice bath. The precipitated cerium oxalate was washed with water and mounted for counting using the standard filter-chimney setup.

Rare Earths

A radiochemical purification procedure identical to that described for cerium was followed as far as the second precipitation of the rare earth hydroxides. The washed rare earth hydroxide precipitate was dissolved in a drop or so of concentrated hydrochloric acid, and the solution was diluted with water and heated in a hot water bath; 1/4 ml of wet Dowex-50 cation-exchange resin was added to the solution, and the resulting suspension was agitated for a few minutes and allowed to settle. The resin was then drawn directly into a syringe-operated glass pipette ("spitzer"), leaving most of the supernatant liquid behind. The pipette was used to distribute the resin evenly, in a thin layer, over the top of a heated bed of Dowex-50, 7 mm in diameter by 55 cm long. The column was maintained at a constant temperature by means of refluxing trichloroethylene. The rare earths were eluted by passing 1 M ammonium lactate through the column at a rate of about six drops per minute. The pH of the eluting agent was 3.20 at the beginning of the run and was increased at a rate of about 0.1 pH unit per hour throughout its duration. The change of pH was accomplished by employing a pH changer nearly identical to the one described by Nérvik. The drops were allowed to collect in small tubes mounted in an automatic turntable; usually 5 drops per tube were collected in the early part of the run and this was later increased to about 15 or 20. The rare earths were located by introducing two or three drops of oxalic acid into each tube. Usually, thulium appeared at about drop 500 and neodymium at about drop 3000.

The rare earth oxalate precipitate generally appeared in a zone of from three to five tubes near thulium to around ten tubes near neodymium. Only the central highest-yield tubes were chosen for any particular rare earth sample. The tubes on either side of the center section (containing perhaps 20% to 40% of the total rare earth) were
rejected, particularly on the trailing side of the peaks, because of the danger of contamination by adjacent rare earths. The contents of the selected tubes for a given rare earth were quantitatively transferred to a long-taper centrifuge cone, more saturated oxalic acid was added, and the rare earth oxalate was allowed several minutes to form completely. The cone was then centrifuged, the supernate poured off, and the precipitate slurried with a few drops of oxalic acid solution and transferred directly, using a syringe-operated pipette, to a damp filter paper disk mounted in the filter chimney. The precipitate was cautiously washed with water, employing the same pipette, and mounted for counting.

Hafnium

Barium fluohafnate and barium fluozirconate were precipitated together when ice-cold, saturated barium nitrate solution was added to the chilled target solution. The precipitate was quickly washed with ice water and then slurried for several minutes with an ice-cold mixture of concentrated nitric acid and saturated boric acid. The slurry was diluted with water, to dissolve the sediment, and made basic with ammonium hydroxide. The precipitated hydroxides were washed with water and dissolved in 1 M hydrofluoric acid. Cerium carrier was added, and the precipitated fluoride was removed by centrifugation. The solution was chilled in an ice bath, and ice-cold saturated barium nitrate was added to precipitate the hafnium and zirconium. The precipitate was dissolved and the hydroxides were precipitated in the manner described above. This time the hydroxide precipitate was dissolved in 3 M perchloric acid. The zirconium and hafnium were extracted from this solution into 0.4 M thenoyltrifluoroacetone in benzene, and back-extracted into 5 percent ammonium bifluoride. Boric acid was added to complex the fluoride, and the solution was made basic with ammonium hydroxide. The precipitated hydroxides were washed with water and dissolved in 3 M perchloric acid. The solution was heated in the hot water bath and 1/2 ml of wet Dowex-50 cation-exchange resin was introduced. The hot suspension was allowed to stand with occasional stirring for a few
minutes, then allowed to settle, and the resin was transferred to the top of a Dowex-50 resin bed contained in a column 7 mm by 55 cm. This was done in the same way as is described in the rare earth section. Zirconium was eluted by passing 0.5 M sulfuric acid through the column. After the zirconium was completely eluted, that is, after the eluate failed to show a precipitate when made basic with ammonium hydroxide, the hafnium was eluted with 3 M sulfuric acid. The hydroxides were precipitated by making the respective eluates basic with excess ammonium hydroxide. The hydroxides were washed with water, slurried with more water, and mounted for counting employing the filter-chimney setup.

This separation is very clean. No trace of the very prominent K x-rays occurring in the decay of hafnium could be found in the zirconium, and spectrographic analysis of the hafnium sample proved the absence of zirconium.

**Tantalum**

Tantalum was separated from the target solution only after the insoluble fluorides and sulfates and zirconium and hafnium had been removed. Concentrated potassium fluoride was added to the target solution to precipitate potassium fluotantalate. The precipitate was washed with water and dissolved in a mixture of saturated boric and saturated oxalic acids. The solution was made slightly alkaline with ammonium hydroxide and the precipitate was washed with water. The precipitate was dissolved in a solution 6 M in sulfuric acid and 0.4 M in hydrofluoric acid, from which the tantalum was extracted into diisopropyl ketone. The organic phase was washed with two small portions of the 6 M sulfuric acid and 0.4 M hydrofluoric acid mixture. The tantalum was back-extracted into water and precipitated with ammonium hydroxide. The precipitate was washed by boiling with concentrated nitric acid. The organic extraction and boiling nitric acid wash were repeated. The final precipitate was washed with water, slurried with more water, and mounted for counting.
Tungsten was recovered from the target solution after the insoluble fluorides and sulfates, zirconium and hafnium, and tantalum, had already been precipitated. The final supernatant solution resulting from the above-mentioned separations, was evaporated nearly to dryness in a platinum vessel. The residue was slurried with concentrated ammonium hydroxide and centrifuged. Solid cobalt chloride was added to the supernatant solution, and then concentrated hydrochloric acid was added, adjusting the solution to a pH of ~6. The resulting precipitate was heated with 6 M hydrochloric acid for several minutes, then centrifuged. The precipitate was dissolved in a few drops of concentrated ammonium hydroxide, water was added, and ferric hydroxide was precipitated by adding a drop or two of iron carrier, agitating vigorously. To the supernatant solution was added 1/2 ml of saturated tartaric acid, 1/2 ml of concentrated sulfuric acid, and a few drops each of bismuth and molybdenum carriers. The solution was heated and hydrogen sulfide was passed in for several minutes, until the precipitated sulfides were coagulated, after which the suspension was centrifuged. Tungsten oxide was precipitated from the supernatant solution by adding several ml of concentrated nitric acid and heating in the hot water bath. The sulfide precipitation and tungsten oxide recovery was repeated. The tungsten oxide was dissolved in a few drops of concentrated ammonium hydroxide, and 3/4 ml of saturated tartaric acid were added, followed by 10 ml of water, 1/2 ml of concentrated hydrochloric acid, and niobium carrier. The mixture was shaken briefly with chloroform, 5 ml of 6% cupferron reagent was added, and the mixture was again shaken. The chloroform layer was discarded, and the extraction was repeated once with clean chloroform. To the aqueous layer was added 1/2 ml of concentrated sulfuric acid, and bismuth and molybdenum carriers. The acid sulfide precipitation and subsequent precipitation of tungsten oxide was repeated. The final tungsten oxide precipitate was washed with water, slurried with more water, and mounted for counting.
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