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Tantalum Spallation and Fission Induced by 340-Mev Protons*

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Nuclides formed as spallation and fission products during bombardment of tantalum metal with 340-Mev protons in the 184-inch Berkeley cyclotron were separated chemically, identified, and their formation cross sections were calculated. A very broad fission peak which extends from mass 20 to mass 132 is observed. The maximum fission yield occurs in the region of the nuclide Kr^{83} and analysis of a set of contour curves fitted to the data indicates that either Hf^{166} or Lu^{166} is "the most probable fissioning nucleus." The total cross section for fission is estimated to be 4.1 mb. Comparison of the fission data of tantalum with those of uranium and bismuth under the same bombardment conditions indicates that asymmetric fission is much more probable in tantalum than in either of the other elements. In the spallation region it is observed that neutron emission is the predominant spallation reaction. Integration under the spallation yield curve indicates that of those tantalum target nuclei which received at least enough excitation energy to reach the region of "the most probable fissioning nucleus" less than 1 percent undergo fission; the remainder undergo spallation reactions.

Activities which have been observed here for the first time include a 136-minute rhodium activity which has tentatively been assigned as an isomer of Rh^{107}; 29.4-hour Er^{160}; 5.0-hour Ho^{160}; and 74-minute Yb^{167}.

I. INTRODUCTION

THE earliest fission product studies and the reaction on which the most complete data are available concern the thermal neutron fission of uranium. Principal features of this fission process are:

1. Predominantly asymmetric splitting of the compound nucleus as shown by the appearance of two peaks in the fission yield *versus* mass curve.

2. Essentially complete absence of fission products on the neutron deficient side of stability.

3. Extremely steep slopes on both "wings" of the fission yield *versus* mass curve, with no fission products having a mass less than 72 or greater than 162 being observed in abundances greater than 10^{-5} percent of the fission events.

As the incident neutron energy is increased, the shape of the fission yield *versus* mass curve begins to change markedly, particularly in the region of symmetrical fission.[1] Engelkemeir, Freedman, Seiler, Steinberg, and Winsberg,[2] and Steinberg and Freedman[3] observed that when Pu^{239} was irradiated with neutrons of approximately 600 kev the yield of Pd^{109} was 50 percent higher than the yield with thermal neutrons. Turkevich and Niday,[4] as the result of bombarding thorium with pile neutrons of 2.6-Mev average energy, also observed an increase in the symmetrical fission region and suggested that the fission process at these energies is a combination of two types, one asymmetric and the other symmetric.

When the neutron energy is increased still further, the probability of symmetrical fission becomes even more pronounced. Spence[5] has shown that when U^{235} is irradiated with 14-Mev neutrons, symmetrical fission becomes one hundred times more probable than with thermal neutrons. This rise in the symmetrical fission yield is accompanied by a decrease in the yields of those nuclides which lie at the peaks of the thermal neutron yield curve; i.e., the yield of Mo^{99} was about 15 percent lower at 14 Mev than with thermal neutrons.

This increase in the symmetrical fission probability at higher excitation energies has also been observed in charged-particle bombardments. Newton,[6] irradiating thorium with 37.5-Mev alpha particles, showed that the symmetrical fission yield is almost equal to that for asymmetrical fission and that a deep minimum of the type which is observed in the yield curve for the thermal neutron fission of U^{235} has practically disappeared. Here the compound nucleus is the same for both particles so that direct comparison of the two yield

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[1] For a review of high-energy fission see R. W. Spence and G. P. Ford, Ann. Rev. Nuc. Sci. 2, 399 (1953).

[2] Engelkemeir, Freedman, Seiler, Steinberg, and Winsberg, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper 204, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, pp. 1331–1333.

[3] E. P. Steinberg and M. S. Freedman, reference 2, Paper 219, pp. 1378–1390.

[4] A. Turkevich and J. B. Niday, Phys. Rev. 84, 52 (1951).

[5] R. W. Spence, Atomic Energy Commission Declassified Report AECD-2625, June 1949 (unpublished).

[6] A. S. Newton, Phys. Rev. 75, 17 (1949).

curves is possible. Jones, Fowler, and Paehler,[7] and Tewes and James[8] found that the bombardment of uranium and thorium with protons of various energies (up to about 20 Mev) leads to increasing symmetrical fission as the proton energy is increased. When bombarding energies are increased to extremely high values (i.e., uranium with 380-Mev alpha particles)[9] symmetrical fission predominates and the yield *versus* mass curve shows a single peak with no noticeable contribution from asymmetric fission.

This single symmetrical fission peak has also been observed when elements lighter than uranium or thorium are bombarded with very high-energy particles. Goeckermann and Perlman,[10] bombarding bismuth with 190-Mev deuterons, observed this type of fission peak. In addition, they noted that the maximum of the symmetric fission peak corresponded to a mass less than half the mass of the target nucleus. Their conclusion was that twelve nucleons had evaporated before fission occurred. Furthermore, the peaks of their isobaric chain yields indicate that most of the chain yield for a given mass number is concentrated in the nuclide having the same neutron to proton ratio as the fissioning nucleus.

Fission has been observed in a number of medium-weight elements farther down the periodic table. Batzel and Seaborg[11,12] irradiated copper, bromine, silver, tin, and barium with very high-energy protons and separated activities which lay in the region of the periodic table corresponding to roughly symmetrical fission for each of these elements. Proof that fission and not spallation of alpha particles is actually occurring in these bombardments is based primarily on the observed thresholds and calculated energetics of the reactions involved. Thus the calculated threshold for the spallation reaction $Cu^{63}(p,p3n9\alpha)Na^{24}$ is about 170 Mev while that for the fission reaction $Cu^{63}+p=Na^{24}+K^{39}+n$ is approximately 50 Mev. Since Na^{24} was produced in 70-Mev proton bombardments of copper, the conclusion was drawn that it must have been formed by a fission process. Greenberg and Miller[13] carried out similar work with similar results by bombarding copper with 370-Mev protons.

Evidence for fission of the medium weight elements in the references quoted was based on radiochemical separation of only a few of the possible activities formed. As part of a program in this laboratory of assembling detailed information in order to learn how a single particle of a single energy reacts with widely differing target elements, iron,[14] nickel,[15] copper,[11] zinc,[16] silver,[17] bismuth,[18] and uranium[19,20] have been bombarded with 340-Mev protons in the Berkeley 184-inch synchrocyclotron. Radiochemical separation and identification of activities formed in both the spallation and fission regions was carried out. These data indicate that for the five light elements spallation is by far the most prevalent reaction, with highest yields on the neutron deficient side of stability near the target nucleus. In the binary fission region the yields are very low and the yield *versus* mass number curves are merely a flattened extension of the spallation portion of the curve. It should be mentioned that although yields in the fission region for these elements are low they are measurable, and every element below the target element is formed in some degree during this type of bombardment.

For bismuth and uranium, fission is the predominant reaction and the characteristic single symmetrical high-energy fission peak is obtained. In each case the fission peak height and the chain yield distributions are such as to indicate initial evaporation of nucleons followed by a fission process in which the neutron-to-proton ratio of the fissioning nucleus is preferentially maintained in the fission fragments.

The present work on tantalum was undertaken as part of this over-all program of 340-Mev proton bombardments with the Berkeley 184-inch synchrocyclotron. Tantalum was of particular interest in this series because of its position in the periodic table. Because of the mass defect, atoms of heavy elements like thorium or uranium are strongly exoergic toward fission. Light elements like copper are thermodynamically much more stable, however, and a larger excitation energy must be imparted to the nucleus for fission to occur. The result is that only a very small fraction of reactions in the copper bombardments yield products which may be considered as fission fragments. Tantalum lies between these two extremes, and while it was expected that the total fission cross section would be low there was some question as to whether or not a fission peak would be discernible.

It should be noted that in previous work in this program spallation data for the five lighter elements were rather complete and the fission data meager, while for bismuth and uranium the opposite was true.

[7] Jones, Fowler, and Paehler, Phys. Rev. **87**, 174 (1952).
[8] H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952).
[9] P. R. O'Connor and G. T. Seaborg, Phys. Rev. **74**, 1189 (1948).
[10] R. H. Goeckermann and I. Perlman, Phys. Rev. **73**, 1127 (1948).
[11] R. E. Batzel and G. T. Seaborg, Phys. Rev. **79**, 528 (1950).
[12] R. E. Batzel and G. T. Seaborg, Phys. Rev. **82**, 607 (1951).
[13] D. H. Greenberg and J. M. Miller, Phys. Rev. **84**, 845 (1951).
[14] Rudstam, Stevenson, and Folger, University of California Radiation Laboratory Unclassified Report UCRL-1586, December 1951 (unpublished).
[15] B. Haldar, University of California Radiation Laboratory Classified Report UCRL-1196, April 1951 (unpublished).
[16] W. J. Worthington, M.S. thesis, University of California Radiation Laboratory Unclassified Report UCRL-1627, January 1952 (unpublished).
[17] P. Kofstad, Ph.D. thesis, University of California Radiation Laboratory Unclassified Report UCRL-2265, June 1953 (unpublished).
[18] W. F. Biller, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-2067, December 1952 (unpublished).
[19] R. L. Folger, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-1195, April 1951 (unpublished).
[20] Folger, Stevenson, and Seaborg, Phys. Rev. (to be published).

It was hoped that the tantalum bombardments would yield enough data on both spallation and fission to enable more accurate conclusions to be drawn on the mechanism of those reactions.

II. EXPERIMENTAL PROCEDURES AND TREATMENT OF DATA

A. Target Arrangements

The target material used in this series of experiments consisted of the purest tantalum foil available. Spectrographic analysis of the metal showed it to be free of any impurities which might interfere with the determination of the fission product yields except for approximately 0.01 percent of niobium. Spallation yields for middle weight elements indicated that this impurity would probably affect the determination of the yields of only the neutron deficient nuclides of molybdenum, niobium, zirconium, and strontium in the fission product region of tantalum. The remaining observed fission product activities and all of the activities in the spallation region must be formed from tantalum in these bombardments.

In order to measure the absolute cross section for the formation of the spallation and fission products, the proton beam monitoring procedure developed by Folger and Stevenson[21] was followed. One-inch diameter disks were cut out of 0.00025-in. aluminum, 0.001-in. aluminum, and 0.002-in. tantalum foil with the same die to ensure that they all had the same area. In order to remove surface impurities from the aluminum disks they were washed in conductivity water and acetone and dried in an oven, while the tantalum disks were cleaned in hot concentrated nitric acid, rinsed in conductivity water and acetone and dried.

Si-Chang Fung[22] has shown that in bombardments with 340-Mev protons a significant fraction of the activity formed in 0.00025-in. aluminum foil may be lost through recoil but that this loss can be minimized by placing 0.001-in. aluminum "guard foils" in front and back of the thinner foil. In addition, since recoiling atoms from the copper target holder might introduce contaminating activities with atomic numbers near that of copper into the tantalum target foils, 0.002-in. tantalum "guard foils" were placed in front and back of the tantalum target foils. Thus the target "sandwich" consisted of a 0.001-in. aluminum guard, a 0.00025-in. aluminum monitor, a 0.001-in. aluminum guard, a 0.002-in. tantalum guard, the tantalum target (usually three 0.002-in. tantalum disks), a 0.002-in. tantalum guard, a 0.001-in. aluminum guard, a 0.00025-in. aluminum monitor, and a 0.001-in. aluminum guard. The sandwiched disks were aligned exactly, clamped firmly in the copper target holder, and trimmed about $\frac{1}{4}$-in. from the face of the target holder. A scale diagram of this "thin target" arrangement is shown in Fig. 1(a).

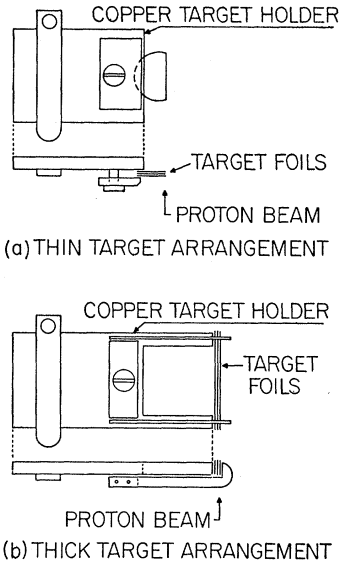

FIG. 1. Target foil arrangements in the 184-inch synchrocyclotron.

After bombardment, the monitor foils were weighed and mounted and their decay followed. The 15-hour activity of Na^{24} could easily be resolved from the decay curve. Since the cross section for the $Al^{27}(p,3pn)Na^{24}$ reaction was known, the cross sections for the formation of the spallation and fission products in tantalum could be calculated.

After several "thin target" bombardments had been evaluated, it became apparent that not enough activity was being formed in the fission product region for accurate resolution of the gross decay curves. Recourse was taken to the "thick target" arrangement [Fig. 1(b)] in which a stack, usually six, of 0.002-in.×0.25-in.×1.75-in. tantalum target foils was wrapped in one thickness of 0.002-in. tantalum guard foil and clamped in a copper target holder in such a manner that the proton beam traversed the 0.25-in. dimension of the target. Radioactive Ni^{66} was used as the "internal monitor" in each of the thick target bombardments. Since the cross section for the reaction $Ta^{181}(p, \text{fission})Ni^{66}$ could be calculated from the thin target bombardments, the cross section for all of the fission products could be obtained by comparing the intensities of their activities to that of Ni^{66}.

B. Treatment of the Target After Bombardment

After data from preliminary experiments had been obtained it became apparent that the choice of chemical treatment procedure for the target after bombardment had to be governed by several equally important factors:

1. The cross sections for the formation of fission products were extremely small, being approximately 10^{-4} of those for the formation of spallation products.

[21] R. F. Folger and P. C. Stevenson, Phys. Rev. (to be published).

[22] Si-Chang Fung, Ph.D. thesis, University of California Radiation Laboratory Unclassified Report UCRL-1465, August 1951 (unpublished).

Thus radiochemical purification (separation) factors of about 10^7 were required between the fission product activities and those in the spallation region (tungsten, tantalum, hafnium, and the rare earth elements).

2. Activities of every element below the region of the rare earth elements were present in comparable yields. Radiochemical purification (separation) factors of approximately 10^4 were required between each of the fission product elements and all of the elements below the rare earth elements.

3. Tantalum, a very difficult metal to put into solution, will dissolve easily only in a mixture of concentrated hydrofluoric acid and concentrated nitric acid. All chemical separation procedures had to begin with a $HF\text{-}HNO_3$ solution of tantalum and its transmutation products.

4. Activity levels of the fission products, even with the thick target arrangements, were so low that the original target solution could not be separated into aliquots. Thus all of the elements which were to be separated in any one bombardment had to come out of the corresponding entire original target solution.

With these factors in mind, the radiochemical separation procedures were developed.[23] In general, after dissolution of the target tantalum in concentrated $HF\text{-}HNO_3$, accurately known quantities, usually 10 mg, of the elements to be separated were added and these were chemically isolated and purified. When there is complete exchange between the active atoms and the inert carrier atoms in the original solution, the weight of the carrier recovered at the completion of the purification will determine the fraction of the original activity recovered. In the case of several elements, special steps had to be taken to ensure complete exchange. In the case of others, where there was danger of the active atoms being adsorbed on the walls of the lusteroid cones in which the target was dissolved before the inert carrier could be added, the carrier had to be added before the target was dissolved. In every case the carriers were added as soon as practicable.

The final purified compound in each case was transferred wet to a tared aluminum dish which had a depression of 1 cm^2 to define the area of the final compound. The precipitate was dried and weighed, then a drop of dilute clear lacquer was placed on it and dried. The lacquer did not contribute significantly to the mass of the sample but served to bind it in place during the subsequent counting operations. Those compounds which could not be dried adequately on an aluminum base were first ignited in porcelain or platinum crucibles and the dried powders transferred to the tared aluminum dishes. For counting purposes the final weighed samples were fastened to 2-mm thick aluminum mounting plates which held the dishes in a fixed position and were constructed to fit the shelving arrangement of the various counters.

[23] Space limitations make it impossible to include a detailed description of the chemical procedures here. For such a description reference should be made to the Ph.D. thesis of Walter E. Nervik, University of California, April, 1954; see also W. E. Nervik, University of California Radiation Laboratory Report UCRL 2542, April, 1954 (unpublished). The description of chemical procedures has also been deposited as Document No. 4440 with the ADI Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington 25, D. C. A copy may be secured by citing the Document number and by remitting $3.75 for photoprints, or $2.00 for 35 mm microfilm. Advance payment is required. Make checks or money orders payable to: Chief, Photoduplication Service, Library of Congress.

C. Treatment of Decay Data

After each element fraction had been separated and purified, the decay of the gross activity was measured by that means which would give the necessary data for the most accurate resolution of the components involved. Wherever possible, samples were counted with a Geiger-Müller counter in which the counting unit itself was an end window, chlorine-argon filled Amperex-type 100 C tube mounted in such a way that samples could be placed in any of five fixed counting positions which ranged from 0.46 to 6.81 cm from the end of the tube. Coincidence counting corrections for this counter were determined empirically by following the decay of a purified sample of 61-hour Y^{90}.

In principle, the decay of the total activity in a given sample may be expressed as a sum of exponential terms and should be subject to resolution into the individual components. In practice, this was usually feasible only in relatively simple mixtures where the half-lives of the activities involved differed by more than a factor of two. For more complex mixtures it was often possible to resolve the components by following decay through absorbers which blocked off the softer radiation. In other cases it was found most practical to obtain the activity of individual nuclides by "milking" their daughter activities chemically and counting them separately.

For samples in which the gamma radiation was of interest, the gamma-ray spectra were obtained through the use of a 50-channel gamma-ray pulse analyzer in which the sensitive unit was a one-inch thick sodium iodide-thallium activated scintillation crystal. Decay of an individual gamma ray could be followed by counting the sample periodically, plotting the gamma-ray spectra, integrating under the desired peak, and plotting integrated counts as a function of time.

In calculating cross sections for the formation of the various product nuclides, the data of interest are the absolute disintegration rates of the nuclides involved. This disintegration rate may be related to the observed counting rate by the equation:

$$D = A_{obs}/(F_a F_{eff} F_g F_{bks} F_{abs} F_{ssa}),$$

where D is the absolute disintegration rate; A_{obs} is the observed counting rate; and the various correction factors are as follows:

F_a, Correction for Abundance of the Radiation Being Counted

For a majority of nuclides observed in this study, the decay schemes were known and abundances for all important modes of decay had been published.[24] In a number of cases, however, where the nuclide had not been studied thoroughly or was discovered in these experiments, such data were not available. When such cases occurred, assumptions about the decay schemes were made which will be stated subsequently in those sections which deal with each nuclide involved. Thus corrections to the calculated cross sections may be made when more complete data on decay schemes become available.

F_{eff}, Correction for Counting Efficiency

The counting efficiency was assumed to be 100 percent for beta particles in the Geiger-Müller tube. When x-rays or gamma rays were present, counting efficiencies in the Geiger-Müller tube were obtained from the work of Studier and James.[25] When gamma-ray spectra were obtained through the use of the 50-channel gamma-ray pulse analyzer, the counting efficiency as a function of gamma-ray energy was obtained from the work of Maguire and O'Kelley,[26] and Kahn and Lyon.[27]

F_g, Geometry Correction

This factor, which is simply the fraction which enters the detection device of the radiation which is being counted, was determined empirically by using known standards for each of the counting arrangements used.

F_{bks}, Backscattering Correction Factor

Empirical data for beta particles were found to agree more closely with Burtt's[28] results than with those of Zumwalt[29]; therefore Burtt's backscattering factors were used throughout. Positrons were assumed to have the same backscattering factors as beta particles since more accurate values were not available.

F_{abs}, Correction for Air and Window Absorption

In the "Shelf 2" geometrical arrangement in which most of the samples were counted, radiation had to pass through approximately 5.8 mg/cm^2 of air and mica before entering the sensitive volume of the Geiger-Müller tube. Correction for the absorption in this material was made wherever possible by making absorption measurements with either aluminum or beryllium absorbers and carefully extrapolating the initial portion of the curves of the plotted data in such a manner as to determine the fraction of soft radiation absorbed by the air and mica window. Where this was not practicable the correction factors were calculated from the beta particle *versus* aluminum half-thickness data of Seelmann-Eggebert.[30]

F_{ssa}, Correction for Self-Scattering and Absorption in the Sample

Data of Nervik and Stevenson[31] were used for all samples in these experiments.

III. RADIOACTIVE NUCLIDES OBSERVED[32,33]

Sodium

Sodium-24 was the only activity observed in the sodium fraction. Starting with approximately 1000 counts per minute, the gross decay curve showed only this 15.1-hour component down to the background counting rate. An aluminum absorption curve indicated the presence of a 1.4-Mev beta particle while analysis of the gamma-ray spectrum showed a gamma ray of 1.36 Mev, both of which have been reported for Na^{24}.

Magnesium

The gross decay curve for the magnesium fraction showed a 21.2-hour component for over eight half-lives, then "tailed" into a long-lived component at 10 counts per minute above the background. Aluminum absorption curve measurements indicated the presence of a 3-Mev beta particle with a softer component also present. This was confirmed by analysis of the beta spectrum on a crude beta-ray spectrometer. The 21.2-hour activity has been identified as Mg^{28}, which decays by emission of a 300–400-kev beta particle. The 2.30-minute Al^{28} is in equilibrium with the Mg^{28} parent and decays by emission of a 3-Mev beta particle. The low long-lived tail may be attributed to a trace of Be^7 which could have followed through with the magnesium in the chemical procedure.

Potassium

The potassium decay curve consists of two components which can be separated into 12.4- and 22.4-hour half-lives and which can be identified as due to K^{42} and K^{43}. Aluminum absorption curves indicate the presence of a beta particle of approximately 3.5 Mev belonging to the short-lived component and a softer beta particle of approximately 0.8 Mev with the longer-lived K^{43}. Potassium-42 decays 75 percent by a 3.6-Mev beta particle and 25 percent by a 2.0-Mev beta particle, while K^{43} has beta particles of reported energies 0.8 and 0.24 Mev. In calculating the cross section for the formation of K^{43}, it was assumed that this nuclide decays 80

[24] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[25] M. Studier and R. James (unpublished data).
[26] P. W. Maguire and G. D. O'Kelley, California Research and Development Corporation Report MTA-40, September 1953 (unpublished).
[27] B. Kahn and W. S. Lyon, Nucleonics **11**, 61 (1953).
[28] B. P. Burtt, Nucleonics, No. 8, 28 (1949).
[29] L. R. Zumwalt, Atomic Energy Commission Declassified Report MDDC-1346, September 1947 (unpublished).
[30] Seelmann-Eggebert, Nature **31**, 201 (1943).
[31] W. E. Nervik and P. C. Stevenson, Nucleonics **10**, 18 (1952).
[32] Unless otherwise noted, disintegration data were obtained from Hollander, Perlman, and Seaborg (reference 24) or the GE Chart of the Nuclides (reference 33).
[33] General Electric Chart of the Nuclides, Knolls Atomic Power Laboratory, revised to November 1952.

percent by emission of the 800-kev beta particle and 20 percent by the 240-kev beta particle, this assumption being based on the shape of the soft components of the absorption curve. Analysis of the gamma-ray spectra showed that there were two gamma rays present, one of 375±10 kev and another of 615±10 kev energy, each of which decayed with approximately a 22-hour half-life. Potassium-43 has a reported gamma ray of approximately 0.4 Mev, but this energy measurement was made by absorbers. It seems probable, therefore, that both the 375- and 615-kev gamma rays may be assigned to K^{43}. When corrected for counting efficiency in the NaI crystal, the two gamma rays have the same abundances within experimental error. No evidence for the 1.51-Mev K^{42} gamma ray was seen in the spectra, probably because of its low abundance and low counting efficiency.

Chromium

Gross decay of the chromium fraction showed the presence of a small amount of a short-lived activity which may have been 42-minute Cr^{49}. The curve then tailed into a long-lived component, part of which may have been 27-day Cr^{51} and part a longer-lived nuclide. The activity level of the chromium fraction, however, was too low for accurate identification of any of these species, although gamma rays of approximately the energies reported for Cr^{51} were observed. Cross sections for these two nuclides would be very uncertain because of this lack of positive identification and therefore have not been included in the tabulated data.

Manganese

The manganese fraction decayed over eight half-lives with a 2.6-hour half-life, then showed a small amount of 6-day activity, and finally tailed into a very long-lived component at approximately 10 counts above background. The gamma-ray spectrum contained a fairly large number of gamma-ray peaks, some of which could be assigned as those reported for 2.6-hour Mn^{56} or 6.0-day Mn^{52}. Since the possibility of the appearance of peaks due to Compton scattering was so great, the complete identification of either spectrum or the unequivocal assignment of all peaks in the observed spectrum was quite impossible. From the peaks identified and the observed half-lives, however, it seems that Mn^{52} and Mn^{56} were present in high purity. The long-lived tail may have been 310-day Mn^{54} or possibly a small amount of impurity in the sample; the activity level was too low for identification.

Iron

The iron fraction decay curve was a straight line corresponding to a half-life of 46 days. Initial activity of these samples was approximately 1500 counts per minute after a two-hour, thick target bombardment. On several bombardments in which special precautions were taken to ensure an iron fraction purified from all possible contamination, no trace was seen of any activity which could be assigned to other isotopes of iron such as Fe^{60}.

Cobalt

An activity which decayed over a period of 9 half-lives with a half-life of 1.65 hours was present in the cobalt fraction. The gross decay curve then tailed over into a very long-lived component (>60 days) at approximately 50 counts above background. Aluminum absorption curves showed a beta particle of approximately 1.1 Mev to be present in the 1.65-hour activity. Cobalt-61 is reported to decay with such a half-life, 55 percent by a 1.45-Mev beta particle and 45 percent by a 1.00-Mev beta particle, which is consistent with the absorption data.

Gamma-ray spectra showed the presence of annihilation radiation and a gamma ray of approximately 830 kev in the long-lived activity of the cobalt fraction. The 72-day Co^{56}, 270-day Co^{57}, and 72-day Co^{58} each decay by positron emission, while Co^{56} has a gamma ray of 845 kev and Co^{58} a gamma ray of 810 kev. Therefore abundances of these nuclides could not be calculated from the intensities of the gamma-ray spectra. Since it also was impractical to resolve these activities in the low count Geiger-Müller decay curve, their cross sections have not been included in the final tabulation.

Nickel

A two-component decay curve of 2.56-hour and 56-hour activities was obtained in the nickel fraction, corresponding to Ni^{65} and Ni^{66}, respectively. Aluminum absorption curves taken after the short-lived Ni^{65} had decayed showed a hard component of approximately 2.7 Mev and a softer component of approximately 0.3 Mev. A 2.63-Mev beta particle is reported for the 5.1-minute Cu^{66} daughter of Ni^{66}. In calculating cross sections, it was assumed that Ni^{66} decayed 100 percent by emission of the 300-kev beta particle.

Copper

Nuclides with 12.9- and 60-hour half-lives were the only ones detectable in the copper fraction. These were assumed to be Cu^{64} and Cu^{67}, respectively. No evidence was seen of the neutron deficient 3.3-hour Cu^{61} although purified samples were counted less than 6 hours after the end of the bombardment.

Gallium

The gallium decay curve contained two activities with 5.0-hour and 14.3-hour half-lives, which definitely could be resolved. In addition, there was a small amount of a short-lived activity which may have been 20-minute Ga^{70} but which could not be identified accurately because of the relatively large amounts of other activities present. Since purification of the gallium fraction

took slightly more than two hours (or six half-lives of Ga^{70}) following the end of bombardment, a large part of this original activity decayed before the fraction could be counted. Aluminum absorption curves confirmed the presence of a beta particle of approximately 1.5 Mev which was associated with the 5-hour activity. The two activities seen are assumed to be 14.3-hour Ga^{72} and 5.0-hour Ga^{73}.

Arsenic

After decay of short-lived activities was complete, the arsenic fraction decayed to background with a 17.5-day half-life. When this activity was subtracted from the gross curve a curve was obtained which could not be visually resolved into straight components. If, however, it is assumed that the second curve consisted of 26.8-hour As^{76} and 39-hour As^{77}, an analytical treatment could be used to establish the intensities of the activities. Aluminum absorption curves could not be used in this case to identify individual nuclides but did show beta particles of approximately the energies reported for As^{74}, As^{76}, and As^{77}.

Bromine

Resolution of the gross decay curve of bromine gave activities of three half-lives; 2.4 hours, 4.4 hours, and 35 hours. The decay was followed from an initial count of approximately 16 000 counts per minute down to 10 counts above background. Aluminum absorption curves showed the presence of beta particles of approximately 1.8 and 0.6 Mev. No gamma-ray spectra were taken since equipment was not available when these bombardments were made. On the basis of these data, the activities were assumed to be 4.58-hour Br^{80} in equilibrium with the 18-minute Br^{80}, 35.9-hour Br^{82}, and 2.4-hour Br^{83}.

Rubidium

The decay curve of the rubidium fraction consisted of a small amount of activity of approximately 5 hours half-life, and a mixture of activities having half-lives of approximately 20 days. An analytical treatment of the data was used to identify the intensities of the long-lived components assuming that they were due to the 19.5-day Rb^{86} and 34-day Rb^{84}. Since there was a good possibility that 4.7-hour Rb^{81} and 6.3-hour Rb^{82} could have been formed largely by spallation of the niobium impurity in the target, the short-lived component of the rubidium curve was ignored in calculating cross sections. Using a value of 0.1 percent as niobium impurity in the tantalum target, and estimating a cross section of 5 millibarns for the reaction $Nb^{93}(p,5p5n)Rb^{84}$, it is estimated that niobium could have contributed no more than 5 percent of the cross section actually observed for the production of Rb^{84} from tantalum. The contribution of niobium impurity to the production of Rb^{86} should have been even smaller.

Strontium

The strontium fraction decay curve was resolved into 9.7-hour and 38-hour activities and a long-lived component which had a half-life greater than 30 days. Aluminum absorption curves showed the presence of hard beta particles such as those reported for 9.7-hour Sr^{91}. The 38-hour activity was presumed to be Sr^{83}, but since this nuclide is in the region of highest yields in the spallation of niobium, its cross section is not included in the tabulated data. When all of the short-lived components had had time to decay, yttrium was separated from the strontium sample, the strontium was remounted, and decay of both samples was followed. The repurified strontium fraction decayed with a 54-day half-life and aluminum absorption curves also showed the presence of the 1.5-Mev beta particle reported for Sr^{89}. The yttrium activity, which decayed with approximately 60 days half-life, contained a beta particle of 1.5 Mev and was identified with 61-day Y^{91} which should have grown in from 9.7-hour Sr^{91}. Cross sections for Sr^{91} were calculated on this basis.

Zirconium

Activities with half-lives of 17 hours, 80 hours, and approximately 65 days could be resolved in the zirconium decay curve. Aluminum absorption curves showed the presence of beta particles of approximately 0.96 Mev. These were probably due to the 0.91-Mev positrons of 78-hour Zr^{89}. This nuclide undoubtedly has one of the higher cross sections for formation in the spallation of niobium; therefore, it is not included in the tabulated data. The 17-hour activity could be due either to Zr^{86} or Zr^{97}, while the long-lived component might have been either 85-day Zr^{88} or 65-day Zr^{95}. The neutron deficient isotopes, however, would be expected to have very much lower counting efficiencies than the beta particle emitting neutron excess nuclides. The assumption has been made, therefore, that all of the observed 17-hour and 65-day activities were due to Zr^{97} and Zr^{95}, respectively. The cross sections obtained on this basis then represent upper limits for the true cross sections.

Molybdenum

The molybdenum decay curve could be resolved into two activities with half-lives of 6.7 and 67 hours. The 6.7-hour isomer of Mo^{93} would be formed in very high yield as a spallation product of niobium, so its cross section has not been included in the tantalum data. Beta-spectrograph curves confirmed the presence of the 1.2-Mev beta particle of 67-hour Mo^{99}.

Ruthenium

Ruthenium, which was separated immediately after bombardment, showed a decay curve which could be resolved into activities having apparent half-lives of 4.5 hours, 36 hours, and 41 days. Aluminum absorption

curves taken 2.5 hours after the end of bombardment showed the presence of the 1.1-Mev beta particle of 4.5-hour Ru^{105}. Cross sections for the formation of Ru^{105} were calculated both on the basis of its identification by resolution of the decay curve and resolution of its 36-hour Rh^{105} daughter. The 41-day activity was presumed to be Ru^{103} although its activity level was not high enough for accurate identification with absorption curves or with the beta spectrometer.

Following bombardments in which all of the 4.5-hour Ru^{105} was allowed to decay before the ruthenium was purified, the ruthenium decay curve could be resolved into components of 40 days and 2.8 days half-life. Presumably the 2.8-day activity was due to Ru^{97}. Since this nuclide decays by electron capture, however, its counting efficiency with the Geiger-Müller counter would be both low and uncertain. Therefore decay of these samples was also followed through the use of the gamma-ray pulse analyzer. The 217-kev gamma ray reported for Ru^{97} showed up quite clearly in the spectrum and it decayed with approximately a 2.8-day half-life. No abundances for the 217-kev gamma ray have been reported. In calculating the cross section for the formation of Ru^{97}, it was assumed that 100 percent of the decay was accompanied by the emission of a 217-kev gamma ray and that there was no conversion. Thus the value obtained should represent a lower limit to the cross section for the formation of Ru^{97}.

Rhodium

The decay curve of rhodium could be separated into components of 136-minute and 36-hour half-lives. Beta-spectrograph curves showed that a beta particle of approximately 1 Mev was associated with the short-lived activity while a beta particle of 0.6 Mev was seen after all of this short-lived activity had decayed. The 36-hour 0.6-Mev beta particle emitter was presumed to be Rh^{105}. No 136-minute beta particle emitter had previously been reported for rhodium. A consideration of the chemical separation and purification procedure used for rhodium showed that no element other than iridium could have been present in the final sample. Iridium activities could not have been formed in any significant amounts in the tantalum bombardments. Comparison of formation cross sections for this activity with those for Rh^{105} indicated that this new nuclide probably has a mass greater than 105.

In order to investigate this new activity further, a series of bombardments was made in which uranium was bombarded with 340-Mev protons. In one of these the target material was bombarded for four minutes. Ruthenium was separated within three minutes of the end of bombardment and allowed to decay. Rhodium was then separated from both the original target solution and the ruthenium fraction. A 25-minute Rh^{107}, 36-hour Rh^{105}, and the 136-minute activity were seen in the rhodium removed from the original target solution while only the 25-minute and 36-hour activities

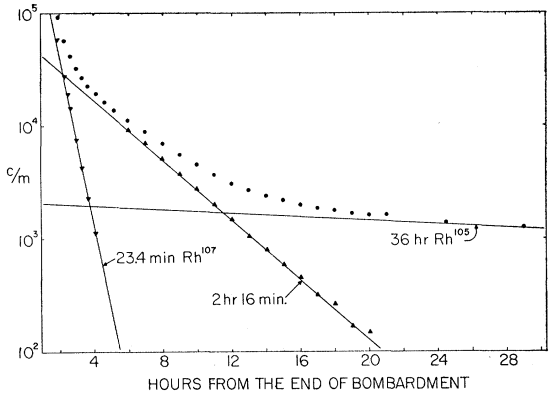

FIG. 2. Decay curve of the rhodium fraction separated from uranium.

grew from ruthenium parents. The three-component decay curve obtained in these bombardments is shown in Fig. 2. Special steps were taken in these separations to ensure that no iridium was present. Assuming the new activity to have the same counting efficiency as Rh^{105}, these decay curves showed the new nuclide to have a higher formation cross section than Rh^{105} in the uranium bombardments.

Gamma-ray spectra were obtained on rhodium samples separated from both tantalum and uranium and indicated a large number of gamma rays associated with decay of the 136-minute activity. The difficulty of resolving the pulse analyzer curves makes abundance data very uncertain, but it seems probable that gamma rays of 195, 225, 510, 630, 715, 1060, 1200, 1260, and 1500 kev are present in the decay scheme of the new activity.

Assignment of a mass to this new activity cannot be made unequivocally. The yields from both tantalum and uranium indicate that the activity is heavier than Rh^{105}. The ruthenium-rhodium experiment shows that it is not the daughter of a ruthenium parent which has a half-life greater than 1 minute. Several of the gamma rays have energies close to those reported for 30-second Rh^{106}, but if we take the maximum gamma-ray energy and the beta particle energy we obtain a total beta disintegration energy of only 2.5 Mev, while the reported value for Rh^{106} is 3.53 Mev. On the basis of the present data, it seems probable that the 136-minute activity is an isomer of Rh^{107} which is not involved appreciably in the decay of 4-minute Ru^{107}. Attempts have been made to separate this nuclide on a time-of-flight isotope separator but they were not successful because of difficulties in ionizing the rhodium samples.

As part of the results from the uranium bombardments, spectra of gamma rays associated with the 25-minute Rh^{107} were obtained. These were superimposed on those from the 136-minute and 36-hour activities so that accurate abundances could not be calculated. Gamma rays of 95 ± 5, 145 ± 5, 305 ± 5, 390 ± 10, and

475±10 kev were observed to decay with an approximately 25-minute half-life.

Decay curves of rhodium samples which had been separated from the tantalum targets after all the 136-minute activity had decayed could be resolved into three components: a large amount of 36-hour Rh^{105}, a small amount of 4.5-day activity, presumably Rh^{101}, and a long-lived component of approximately 200-day half-life, probably Rh^{103}. Decay of gamma-ray spectra was followed in these samples. Although both Rh^{105} and Rh^{101} have known gamma rays of approximately 300 kev, resolution of the decay curve of the 300-kev gamma ray made it possible to obtain the cross sections for the formation of Rh^{101}. No abundance has been reported for the 300-kev gamma ray in the Rh^{101} decay scheme, however, so in these calculations it was assumed that 100 percent of the decay included the 300-kev gamma ray and that no conversion took place. The value obtained is therefore a lower limit to the actual cross section.

Palladium

The only activities observed directly in the palladium fraction were the 13.1-hour Pd^{109} and a small amount of activity which could have been the 17-day Pd^{103}. Palladium 103 decays by electron capture with no gamma rays reported. The amount of 17-day activity present was too low to allow counting of x-rays through absorbers with the Geiger-Müller tube and attempts to follow decay of the x-rays in the pulse analyzer also were not successful.

No evidence was seen in the gross palladium decay for 5.5-hour Pd^{111m}, 22-minute Pd^{111}, or 21-hour Pd^{112}. The 7.5-day Ag^{111} was observed to grow into a target solution from which all silver had been removed immediately after bombardment. On the basis of this information the total cross section for the formation of Pd^{111} could be calculated; but since both of the Pd^{111} isomers decay to Ag^{111}, calculations of cross sections for the formation of each of the Pd^{111} isomers could not be made without more information.

Silver

The activities of 5.3-hour Ag^{113}, 3.2-hour Ag^{112}, 7.5-day Ag^{111}, and 210-day Ag^{110} could be resolved in the silver decay curve. The 7.5-day and 270-day activities were easily resolved, but analytical treatment was necessary to establish the intensities of the 3.2- and 5.3-hour activities. Absorption curves taken when the activity was primarily Ag^{111} showed a 1-Mev beta particle with no soft component which could be attributed to conversion electrons from 8.3-day Ag^{106}.

Gamma-ray spectra taken after almost all of the 7.5-day activity had decayed showed the presence of gamma rays of 63, 154, 280, and 350 kev, all of which have been reported for 40-day Ag^{105}. Cross sections for the production of this nuclide were based on the intensity of the 280-kev peak, assuming that the 280-kev gamma ray was 25 percent abundant in the decay scheme and unconverted.

Gamma-ray spectra taken after most of the 40-day Ag^{105} had decayed showed peaks at 660, 760, 935, and 1400 kev, all of which have been reported for 270-day Ag^{110}. Calculations of the cross section for the formation of Ag^{110} were based on the intensity of the 660-kev gamma radiation; its reported abundance of 100 percent and its e/γ ratio of 0.0025 were used.

Cadmium

Activities with half-lives of 43 days, 53 hours, and 6.7 hours were resolved in the gross cadmium decay curve, corresponding to Cd^{115m}, Cd^{115}, and Cd^{107}, respectively. The Cd^{115} and Cd^{115m} isomers decay by emission of beta particles of known energies so cross sections can be calculated for the formation of these nuclides fairly easily. Cadmium-107 decays more than 99 percent by electron capture and emission of a 94-kev gamma ray so that the cross section for the formation of Cd^{107} is based on gamma-ray pulse analyzer data and the reported e/γ ratio of 16.

The 57-minute Cd^{105} could not be identified in the cadmium fraction because of the large abundance of other activities present. In order to determine whether any of this activity had been formed, an experiment was done in which silver was removed from the target solution immediately after bombardment. Cadmium-105 was allowed to decay for two days, then silver was again removed and purified. Decay of this silver sample was followed to see whether or not Ag^{105} had grown in from the Cd^{105} parent. While the gross Geiger-Müller counter decay curve did seem to show an activity of approximately 40-day half-life at about 10 counts above background, none of the Ag^{105} gamma-ray peaks could be identified in the pulse analyzer equipment. Therefore, it must be concluded that Cd^{105} is not formed in sufficient quantities to be detected by this method.

Gamma-ray spectra taken after most of the 43-day Cd^{115m} had decayed confirmed the presence of the 87-kev gamma ray associated with 470-day Cd^{109}. The activity level was so close to background, however, that no very accurate value for the abundance of this gamma ray could be determined. Cross sections for this nuclide are therefore not included in the final tabulation.

Indium

A long-lived component of 50-day half-life was present as a tail on the indium decay curve. Aluminum absorption curves showed the 2-Mev beta particle of 49-day In^{114} to be in this activity. Shorter-lived components were also present in the sample, but since 4.3-hour In^{109}, 5.0-hour In^{110m}, 1.74-hour In^{113m}, and 4.5-hour In^{115m} were all expected to be formed in some degree they could not possibly be separated in the gross Geiger-Müller decay curve. The decay of indium samples was followed in the gamma-ray pulse analyzer and cross

sections for the formation of those nuclides that could be identified were based on the gamma spectra.

The 205-kev gamma ray reported for In^{109} was observed to decay with a 4.3-hour half-life. No decay scheme abundance data were available for this gamma ray; in the calculations it was presumed to be present in 100 percent abundance and to be unconverted.

The 935- and 661-kev gamma rays of In^{110m} were seen to decay with approximately a 5-hour half-life. Both are reported to be in 100 percent abundance in the decay scheme. The e/γ ratio is 0.005 for the 660-kev gamma ray and it was assumed that the 935-kev line was also essentially unconverted.

An activity of 2.8 days half-life could be separated in the gross Geiger-Müller counter decay curve, and was presumed to be 2.8-day In^{111}, but since this nuclide decays essentially 100 percent by electron capture, the Geiger-Müller data were not used in calculating this cross section. Gamma rays of 247 and 172 kev were seen in the gamma-ray spectra and decayed with approximately a 2.8-day half-life and the intensities of these were used in the calculation on the assumption that they occur once in each decay process and are largely unconverted. In the decay of In^{111} the 172- and 247-kev gamma rays are 100 percent abundant and have conversion coefficients of 0.12 and 0.064, respectively.

Tellurium

The decay curve of the tellurium fraction could be separated into component activities having half-lives of 2.8 hours, 4.5 days, and 17 days, corresponding to Te^{117}, Te^{119}, and Te^{121}, respectively. In the absence of data on the abundance of the 2.5-Mev positron in the decay scheme of Te^{117} it was assumed that this nuclide decays 100 percent by the emission of positrons, with the 28-hour Sb^{117} daughter decaying 100 percent by electron capture.

Cesium

Activities having half-lives of 6.2 hours, 31 hours, and 34 days were the only ones detectable in the cesium decay curve. These would correspond to Cs^{127}, Cs^{129}, and Xe^{127}, respectively.

Gamma-ray spectra showed the presence of the 125- and 416-kev gamma rays reported[34] for Cs^{127} and they were seen to decay with an approximately 6-hour half-life. The 57-kev gamma ray reported for Xe^{127} was observed as part of the 34-day component. Since the positron abundances in Cs^{127} have not been reported, the cross section calculations for Cs^{127} were based on the 57-kev gamma ray in its Xe^{127} daughter. Here the nuclide decays 100 percent by electron capture and it was assumed that there was one unconverted 57-kev gamma ray per disintegration.

Since the 31-hour Cs^{129} decays completely by electron capture, the Geiger-Müller counter data could not be used effectively in calculating the cross section for its formation. Wapstra, Versta, and Boelhouwer[35] have reported gamma rays of 385 and 560 kev as belonging to this nuclide. Gamma-ray spectra taken on the cesium fraction after all of the 6-hour Cs^{127} had decayed showed the presence of gamma rays of 375 ± 10 and 585 ± 20 kev, each of which decayed with a 31-hour half-life. In addition, the 375-kev peak was asymmetric in such a way as to indicate the presence of a small amount of a gamma ray of approximately 420 ± 10 kev. Correcting the 375-, 420-, and 585-kev gamma rays for counting efficiencies of 42, 34, and 17 percent, respectively, the gamma-ray abundances are in the approximate ratio 1.0, 0.092, and 0.233. Xenon-129 has a reported excited state of 40 kev as deduced from beta decay of I^{129}, and it is possible that the 40-kev and the 375-kev gamma rays are in coincidence while the 420-kev gamma ray is a crossover transition. The 40-kev gamma ray is very highly converted, however, so that it would be easily missed in the gamma spectrum. The 375- and 420-kev peaks could not be easily resolved in the gamma spectrum and therefore in calculating cross sections for the formation of Cs^{129} the combined peak was used. It was assumed that these gamma rays were unconverted and were present in 82 percent of the disintegrations of Cs^{129}.

No evidence could be seen of either the 9.6-day Cs^{131} or 7-day Cs^{132} in the gross Geiger-Müller decay curve. Each of these nuclides decays by electron capture, Cs^{131} with no appreciable gamma rays, and Cs^{132} with a gamma ray of 685 kev.[33] These nuclides would therefore be detected with very low counting efficiency in the Geiger-Müller counter. A gamma peak was observed at approximately 680 kev, but it was not in sufficient abundance to identify the half-life.

Barium

Activities corresponding to 2.0-hour Ba^{129}, 2.4-day Ba^{128}, and 12-day Ba^{131} were observed in the gross barium decay curve. Aluminum absorption curves taken when the 2.4-day activity was predominant showed the presence of a beta particle of approximately 3 Mev, presumably the 3.0-Mev positron emitted by the cesium daughter of Ba^{128}. Gamma-ray spectra taken when the 12-day activity was predominant showed the 122, 214, 241, 370, and 494 kev gamma rays reported for Ba^{131}.

In calculating cross sections for the formation of Ba^{128} the decay scheme of Lindner[36] was used. According to these data, 2.4-day Ba^{128} decays 100 percent by electron capture while its 3.8-minute Cs^{128} daughter decays 71 percent by emission of a 3.0-Mev positron and 29 percent by electron capture.

Barium-129 was assumed to decay 100 percent by the 1.6-Mev positron reported by Fink and Wiig.[37]

Barium-131 decays completely by electron capture; cross sections for the formation of this nuclide were

[34] H. B. Mathur and E. K. Hyde, Phys. Rev. **95**, 708 (1954).

[35] Wapstra, Versta, and Boelhouwer, Physica **19**, 138 (1953).

[36] M. Lindner (private communication).

[37] F. W. Fink and E. O. Wiig, Phys. Rev. **91**, 194 (1953).

calculated on the basis of the number of cesium x-rays seen in a single-channel gamma pulse analyzer using a NaI crystal.

Cerium

Activities having half-lives of 6.3 hours, 16 hours, and 72 hours were resolved in the cesium decay curve. These were assumed to be Ce^{133}, Ce^{134}, and Ce^{135}, respectively. There was also a long-lived tail of very low abundance (less than 10 counts per minute) which may have been due to 36-day Ce^{141}, but the activity level was too low for an accurate half-life to be determined. Aluminum absorption curves taken five hours after bombardment showed the presence of beta radiation of approximately 2.4 Mev, probably the 2.7-Mev positron reported for the 6.3-minute La^{134} daughter of Ce^{134}. In calculating cross sections for the formation of cerium isotopes, the data on decay energies and abundances reported by Stover[38] were used.

Rare Earth Elements

After several bombardments in which the heavy rare earth elements were separated and their decay followed, it became apparent that published data on the neutron deficient isotopes of these elements were insufficient to enable correlation of the data with known nuclide decay schemes. A program was then begun to get more accurate mass assignments, half-lives, and decay characteristics of the isotopes of these elements formed in the tantalum bombardments. In general, the procedures used in this program were as follows: (1) The heavy rare earth elements were separated after a low proton intensity bombardment and their gross decay measured. This gave an indication of the half-lives and approximate abundances of the activities present in each element. (2) High intensity bombardments were made in which it was attempted to have at least 10^8 counts per minute of each nuclide present after chemical purification. The chemical steps were carried out essentially carrier-free (usually about 20 micrograms of carrier were added). After the elements had been chemically separated the activities present were mass separated on a time-of-flight isotope separator.[39] Thus accurate mass assignments could be made for the activities seen in the gross decay of each element. However, it was found that only nuclides of the four heaviest rare earth elements could be made from tantalum in sufficient quantities for mass identification by this method. (3) When possible, decay characteristics were obtained by measurements on isotopically separated samples. Otherwise they were measured in the gross mixture under the most favorable conditions. In those cases where a daughter activity of sufficiently long half-life was present a second chemical separation was made to confirm the presence of the daughter.

When little or no information on abundances of gamma rays, positrons, or conversion electrons (i.e., the decay scheme) was available, calculation of a cross section for formation of the rare earth nuclide was undertaken in the following very approximate manner. Geiger counter data were used with the measurements made under conditions where the radiations passed through sufficient absorber (972.6 mg Al/cm^2) to block out all beta particles, positrons, and conversion electrons but not sufficient to absorb a significant fraction of the gamma radiation. Cross section calculations for the formation of these nuclides were then made by using an assumed counting efficiency of 1 percent in each case (or 2 percent for a parent and daughter in equilibrium).

Erbium

Neutron deficient activities of erbium reported in the literature at the time the rare earth program was begun were ~17-hour Er^{161}, ~65-hour Er^{163}, and 10-hour Er^{165}. The gross decay curve of erbium from tantalum bombardment could be resolved into activities with 183-minute and 29.4-hour half-lives. No evidence was seen for any of the three activities previously reported. Separation on the time-of-flight instrument[40] showed the 29.4-hour activity to be due to Er^{160}, while the 183-minute activity is due to Er^{161}.

Handley and Olson[41] have recently reported Er^{161} as having a 3.6-hour half-life and a holmium daughter of 2.5 hours half-life. They report gamma rays of 195, 824, and 1120 kev for the parent and 90 kev for the daughter. They also report no annihilation radiation and say that positron emission, if present, constitutes only a small fraction of the decay. Examination of the erbium fraction for beta radiation on a crude beta-ray spectrometer showed the presence of positrons of 1.2 ± 0.1 Mev which decayed with approximately a 3-hour half-life. No attempt was made to separate the holmium daughter of Er^{161} so that the positron may belong to either Er^{161} or Ho^{161}.

The 29.4-hour Er^{160} is a new activity which has not previously been observed. Neither the decay curve of the erbium fraction separated immediately after bombardment, nor that of isotopically separated Er^{160} showed the growth of any short-lived daughter activity. In the erbium fraction this growth would easily have been obscured by the presence of relatively large amounts of 3.6-hour Er^{161} and its 2.5-hour Ho^{161} daughter. The isotopically separated Er^{160} could also have been contaminated by the activity of the short-lived mass-161 nuclides. When an erbium fraction which had been separated immediately after bombardment had been allowed to decay for several days, however, and a second erbium-holmium separation was made,

[38] B. J. Stover, Phys. Rev. **81**, 8 (1951).

[39] M. C. Michel, Ph.D. thesis, University of California Radiation Laboratory Unclassified Report UCRL-2267, 1953 (unpublished).

[40] M. C. Michel and D. H. Templeton, Phys. Rev. **93**, 1422 (1954). Where the values for half-lives given by Michel and Templeton differ from those given in the present paper, the latter, more recent values are probably to be preferred.

[41] T. H. Handley and E. L. Olson, Phys. Rev. **93**, 524 (1954).

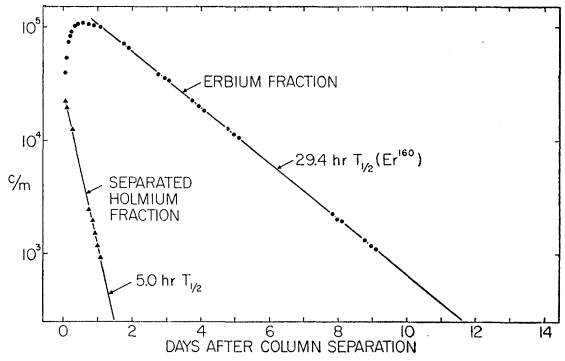

FIG. 3. Er^{160}-Ho^{160} decay curve.

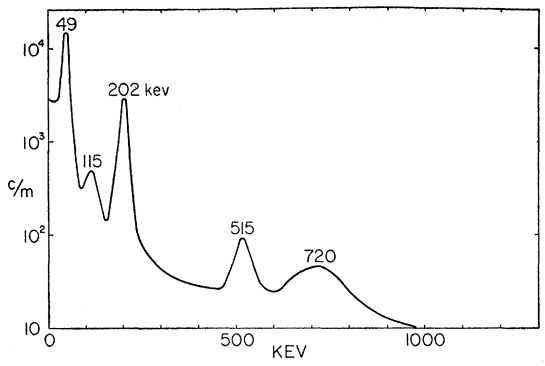

FIG. 4. Gamma spectrum of Tm^{167}.

the erbium fraction showed a decided growth before decaying with a 29-hour half-life, while the holmium fraction decayed with a half-life of 5.0 hours. (See Fig. 3.) Gamma rays of approximately 87, 194, 650, 730, 890, and 970 kev plus x-rays of 46 kev were seen in the holmium fraction. They also were seen to grow into the erbium sample. No gamma rays or annihilation radiation were observed which could be assigned to Er^{160}.

Handley[42] has recently reported a 5.0-hour holmium activity with gamma rays of 190, 710, and 950 kev which was formed by proton bombardments on Dy_2O_3. The activity was assigned to Ho^{162} on the basis of abundance measurements relative to the 2.5-hour Ho^{161} formed in the same bombardments. From the half-life and gamma-ray spectra measurements, it would seem that this is the same activity as that which grew from Er^{160} and that it actually is Ho^{160} rather than Ho^{162}.

Handley and Olson[43] have reported a 75-minute activity for Er^{163}. No evidence of this activity was seen in erbium fractions upon which measurements were made within six hours of the end of bombardment, although the activity may have been obscured by the 183-minute Er^{161}. Attempts to separate Er^{163} on the time-of-flight instrument and to extract it from any possible long-lived Tm^{163} parent were unsuccessful.

No evidence of 10-hour Er^{165} was seen in the erbium decay curve and attempts to separate it isotopically were unsuccessful. For reasons which will be explained later, Er^{165} should not be formed from tantalum in large enough abundance to be separated isotopically. As mentioned above, the erbium decay curve included a 2.5-hour daughter growing from a 3.1-hour parent and a 5-hour daughter growing from a 29-hour parent, so it is not surprising that a relatively small amount of a 10-hour activity could not be identified by resolution of the curve.

Thulium

Neutron deficient isotopes previously reported for thulium are 7.7-hour Tm^{166}, 9.6-day Tm^{167}, and 85-day Tm^{168}. In the decay of the thulium fraction, the 7.7-hour and 9.6-day activities could be resolved. In addition, an activity of approximately 29 hours half-life was seen. The activities were separated isotopically[40] and the mass assignments of the 7.7-hour and 9.6-day activities were found to be correct. The 29-hour activity was similarly[40] identified as Tm^{165}. As a further check on this latter mass assignment a sample of thulium which had been separated immediately after bombardment was allowed to decay for three days. A thulium-erbium separation was then made. The erbium fraction decayed with the known 10.2-hour half-life of the Er^{165} daughter of Tm^{165}. Handley and Olson[43] have recently reported Tm^{165} to have a half-life of 24.5 hours. The discrepancy between these half-lives probably results from the difficulty of resolution of the multicomponent decay curves in each case.

Unequivocal assignment of gamma-ray energies was not made for these nuclides in the thulium fraction. Thulium-167, however, was obtained in pure form by extracting it from its Yb^{167} parent. The 7.7-hour Tm^{166} daughter of Yb^{166} was also present initially in this experiment. After the 7.7-hour Tm^{166} had disappeared the sample decayed with a pure 9.6-day half-life. A gamma spectrum taken on this activity showed the presence of gamma rays of 49, 115, 202, 515, and 720 kev energies in relative abundances of 1, 0.02, 0.29, 0.09, and 0.18, respectively (corrected for counting efficiency in the NaI crystal). This spectrum is shown in Fig. 4.

Thulium-166 and Tm^{167} both have parent activities of moderate half-lives. Since the chemical purification takes approximately six hours measured from the end of the bombardment, cross sections for the formation of these nuclides could be seriously affected by decay of the parents. In those bombardments from which cross sections for the formation of thulium isotopes were obtained, the rare earth fraction was allowed to decay overnight before the chemical separations were made. Cross sections for the formation of 29-hour Tm^{165} and 9.6-day Tm^{167} then represent total chain yields for their mass numbers.

No 85-day activity was seen in the thulium samples upon which cross section determinations were made. In

[42] T. H. Handley, Phys. Rev. **94**, 945 (1954).
[43] T. H. Handley and E. L. Olson, Phys. Rev. **92**, 1260 (1953).

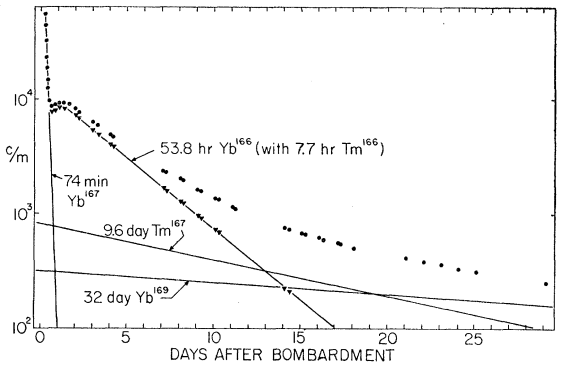

FIG. 5. Gross ytterbium decay curve.

one of the survey bombardments, however, when decay of a very active thulium sample was measured, the 9.6-day Tm^{167} decay curve was seen to tail into a small amount of activity which decayed with approximately an 85-day half-life. This could have been either 85-day Tm^{168}, 129-day Tm^{170}, or a mixture. Assuming that this was all due to 85-day Tm^{168} and that the radiations from this nuclide are counted with the same efficiency as those from Tm^{167}, the cross section for the formation of Tm^{168} was calculated to be less than 2 percent as large as that for Tm^{167}. It should be noted that Tm^{168} is a shielded nuclide and may be formed only directly while Tm^{167} lies at the end of a chain of relatively short-lived activities.

Ytterbium

The 62-hour Yb^{166} and 32-day Yb^{169} were the only neutron deficient ytterbium activities known when the present research was begun. The ytterbium fraction separated in these bombardments gave a decay curve which could be separated into five components: an initial 74-minute activity, growth of a daughter into a parent having a 53.8-hour half-life, a 9.6-day activity, and a 32-day activity. A copy of this decay curve is shown in Fig. 5. Time-of-flight separation[40] of the ytterbium activity showed the 53.8-hour activity to be due to Yb^{166} with the 7.7-hour Tm^{166} daughter growing into this sample. The 32-day activity was separated[40] at mass-169.

The short-lived 74-minute activity decayed too rapidly to allow isotopic separation to be carried out. In an experiment where ytterbium was chemically separated immediately after bombardment, allowed to decay for 24 hours, and a second ytterbium-thulium separation made, the only activities present in the thulium fraction were 7.7-hour Tm^{166} and 9.6-day Tm^{167}. On the basis of this extraction experiment, the 74-minute activity is assigned to Yb^{167}. No gamma-ray data were obtained on this nuclide because of the presence of so much other activity. Analysis of data taken with a crude beta spectrometer showed the presence of a positron of 2.4 ± 0.2 Mev which decayed with an approximately 70-minute half-life. The fraction of decay via the positron route was not determined.

Gamma spectra taken on Yb^{166} using the ytterbium-thulium equilibrium mixture showed the presence of gamma rays of 80, 112, 140, 180, 670, 800, and 1320 kev energy as well as the 47-kev x-rays.

Lutetium

The gross decay curve of the activity in the lutetium fraction could be resolved into components with half-lives of 1.7 days, approximately 7.5 days, 32 days, and a small amount of a long-lived tail. Attempts to separate these activities on the time-of-flight isotope separator were unsuccessful. In the calculations of the cross sections for the formation of the lutetium isotopes, published data on the half-lives were used. The 1.7-day activity was assumed to be due to Lu^{170}.

Half-lives of 8.5 days and 6.7 days have been reported for Lu^{171} and Lu^{172}, respectively, but activities with half-lives as similar as these could not be resolved in the gross decay curve. Since both of these isotopes are essentially shielded nuclides, the cross sections for their formation are of particular interest to the over-all picture. Thus, in order to get something to use for these cross sections, their combined decay curve was treated as a single activity having a half-life of 7.5 days and the resulting cross section value divided by two in order to get the individual values. This is admittedly only a rough approximation to the actual cross section in each case, but since the two nuclides have adjacent mass numbers the values should not be too poor.

The presence of a 32-day activity is somewhat puzzling. Since the lutetium-ytterbium separation is a difficult one, it is possible that this activity is due to an impurity consisting of unseparated 32-day Yb^{169}. In these separation experiments, however, the lutetium and ytterbium apparently were well separated, and since only the leading edge of the lutetium peak was used (in the ion exchange resin adsorption-elution method of separation), the contamination due to ytterbium should be negligible. It seems more probable that 32-day Yb^{169} is growing into the lutetium fraction as a daughter activity of Lu^{169}. From the relative magnitude of the 32-day and 1.7-day activities, the 1.7-day activity could be the Lu^{169} parent. Further work will have to be done before any more definite mass assignments can be made.

The very long-lived activities in the tail of the decay curve could have been 1.6-year Lu^{171}, 1.4-year Lu^{173}, 165-day Lu^{174}, or a mixture of these activities. Samples from which cross sections were calculated have not been measured for decay long enough for accurate identification of these species so the cross sections for their formation are not included in the tabulated areas.

Hafnium

Five active neutron deficient isotopes of hafnium have been previously reported: 1.9-hour Hf^{170}, 16-hour

Hf^{171}, ~5-year Hf^{172}, 24-hour Hf^{173}, and 70-day Hf^{174}. Activities with half-lives of ~70 days, 8.5 days, 44±2 hours, and 12±1 hours were resolved from the decay curve of the hafnium fraction. Analysis of the gamma-ray spectra showed gamma rays of 122±10 and 275 ±10 kev energy as well as 55-kev x-rays associated with the 44-hour activity, while a gamma ray of 175±10 kev energy seems to be present with the 12-hour activity.

The 12-hour activity is presumably to be identified with the previously reported 16-hour Hf^{171} while the 8.5-day activity is due to its Lu^{171} daughter. The 12-hour half-life was obtained after resolution of three other components in the gross decay curve so that this would be subject to greater errors than any of the others. All of the hafnium samples showed the same half-life within the error reported, however, so a value of 12 hours was used in the calculation of the cross section for the production of Hf^{171}.

The presence of a 44-hour activity is more puzzling. It is uncomfortably close to the 52.8-hour half-life reported for Ta^{177} and makes one suspect that tantalum may have been incompletely removed from the hafnium fraction. If this were the case, however, the two 8-hour tantalum activities should also have been seen, and in estimated abundance approximately thirty times that of Ta^{177}. No evidence was seen of any 8-hour component. Further, if a mixture of 24-hour (previously reported Hf^{173}) and 52.8-hour activities were present, even in comparable abundances, the decay curve would show a decided bend. This was not the case; the resolved 44-hour component was a straight line over approximately five half-lives. Although further bombardments will have to be made in order to clear up this matter, for the purpose of these experiments it was concluded that the 44-hour activity is actually due to Hf^{173} and cross sections were calculated on that basis.

Tantalum

Seven activities of neutron-deficient tantalum isotopes have been reported in the literature. The gross decay curves of tantalum samples separated immediately after bombardment could be resolved into activities having half-lives of approximately 100 minutes, 8 hours, 2.2 days, and a small amount of a long-lived activity which had a half-life greater than 50 days. No detailed gamma-ray spectra for these nuclides were available from the literature for identification by comparison. In the cross section calculations the 2.2-day activity was assumed to be due to Ta^{177} and the 100-minute activity was assumed to be due to Ta^{178}. The 8-hour decay was undoubtedly due to a mixture of 8.0-hour Ta^{176} and 8.1-hour Ta^{180}. Since these could not be resolved from each other in the gross decay curve, the combined total cross section for their formation is tabulated. Decay of the long-lived tail has not been followed long enough to identify it with the 2-year half-life reported for Ta^{179} so data for this activity have not been included with the cross section figures.

TABLE I. Cross sections.[a]

Nuclide	Cross section (mb)	Nuclide	Cross section (mb)
Na^{24}	0.006	Sr^{89}	0.036
Mg^{28}	0.035	Sr^{91}	0.016
K^{42}	0.0066	Zr^{95}	0.027
K^{43}	0.017	Zr^{97}	0.0042
Mn^{52}	0.0018	Mo^{99}	0.046
Mn^{56}	0.033	Ru^{97}	0.0015
Fe^{59}	0.050	Ru^{103}	0.032
Co^{61}	0.054	Ru^{105}	0.0032
Ni^{65}	0.038	Rh^{101}	*0.0023
Ni^{66}	0.015	Rh^{105}	0.022
Cu^{64}	0.065	Rh^{107m}	(0.009)
Cu^{67}	0.054	Pd^{109}	0.0052
Ga^{72}	0.064	Ag^{105}	*0.027
Ga^{73}	0.044	Ag^{110m}	0.029
As^{74}	0.021	Ag^{111}	0.018
As^{76}	0.056	Ag^{112}	0.0064
As^{77}	0.030	Ag^{113}	0.0029
Br^{80m}	0.095	Cd^{107}	0.012
Br^{82}	0.080	Cd^{115}	0.0016
Br^{83}	0.050	Cd^{115m}	0.0027
Rb^{84}	0.078	In^{109}	*0.0062
Rb^{86}	0.047	In^{110m}	0.019
In^{111}	0.013	Tm^{167}	50.7
In^{114m}	0.021	Yb^{166}	76
Te^{117}	*0.0045	Yb^{169}	32.6
Te^{119}	*0.011	Lu^{170}	218
Te^{121}	0.028	$Lu^{171,172}$	63.7
Cs^{127}	*0.0047	Hf^{171}	67
Cs^{129}	*0.0022	Hf^{173}	130
Ba^{128}	0.0041	Hf^{175}	45
Ba^{129}	*0.0025	Ta^{177}	27
Ba^{131}	0.013	Ta^{178}	152
Ce^{134}	0.019	$Ta^{176,180}$	265
Ce^{135}	0.22	W^{176}	65
Er^{160}	31.3	W^{177}	30
Tm^{165}	48.6	W^{178}	65

[a] Cross sections for nuclides whose decay schemes are not accurately known, and about which assumptions were made, are marked with an asterisk (*).

Tungsten

Five radioactive neutron deficient tungsten isotopes have been previously reported, and four of them have radioactive tantalum daughters. In the decay curves for the tungsten fraction, activities with half-lives of 21.5 days, 53 hours, 8 hours, and approximately 130 minutes were resolved. The 130-minute activity was probably due to a mixture of W^{176} and W^{177} (80 minutes and 130 minutes, respectively). In the calculations, cross sections for the formation of these nuclides were based on the measured yields of their 8.0-hour Ta^{176} and 53-hour Ta^{177} daughter activities.

The 21.5-day activity was identified with the 21-day W^{178} which has 9.35-minute Ta^{178} daughter in equilibrium with it.

No evidence was seen of the 140-day W^{181}, although the tungsten decay was followed as close to background as was practicable. In the gross decay curve a 140-day activity could not have been present with an intensity greater than 0.5 percent of that of the 21.5-day W^{178}.

IV. RESULTS AND DISCUSSION

A summary of the cross sections for the production of spallation and fission products formed in the 340-Mev

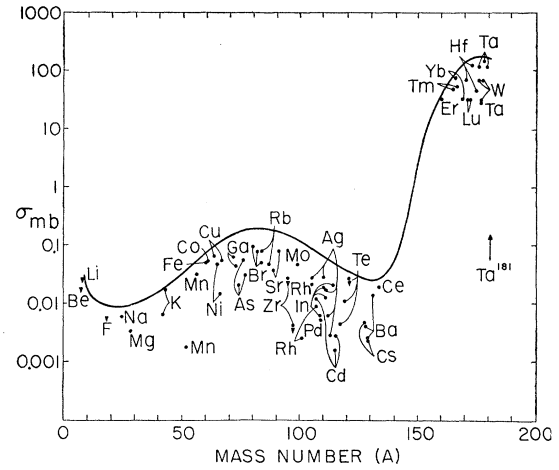

FIG. 6. The distribution of spallation and fission products as a function of mass. Solid line depicts the total chain yield curve obtained by integration of cross sections in Fig. 7.

proton bombardment of tantalum is presented in Table I. Cross sections for individual nuclides represent average values taken from a series of bombardments and are calculated on the basis of a 10.0-mb cross section for the reaction $Al^{27}(p,3pn)Na^{24}$ in the aluminum monitor foils.[21]

The error in these cross sections will depend on the type of decay involved in the individual cases. For beta-emitting activities with an excess of neutrons, cross sections should be reproducible to ± 5 percent. In view of the large number of corrections involved in the calculations, however, an average error of no better than ± 15 percent can be claimed. For neutron-deficient isotopes whose decay schemes, including abundances of positrons, gamma rays, and conversion coefficients are known accurately, this same limit of error should apply. For those nuclides whose decay schemes are not accurately known, and about which assumptions were made, the cross sections will obviously be no more accurate than the accuracy of the assumptions. Cross sections for nuclides in this last category are marked with (*) in Table I and must be regarded as only approximations. Neutron-deficient nuclides which decay by the electron capture process, whose cross sections are based on x-ray and gamma ray counting in which the radiations pass through absorbers, will have values which are not too much affected by lack of information concerning detailed decay schemes. The counting efficiency for a mixture of gamma and x-rays differing widely in energy can be somewhat uncertain when samples are counted on the Geiger-Müller counter, however, so that an accuracy of no better than ± 25 percent may be claimed for samples counted in this way.

A plot of the cross sections from Table I *versus* mass number is shown in Fig. 6. In addition, cross sections for the formation of Be^7, Li^8, and F^{18} which were obtained by interpolation in curves given by Marquez[44] are included for reference. Several features of this plot are apparent even at first glance. First, cross sections for the formation of fragments in the fission product region differ from those in the spallation product region by factors as high as 10^4. Second, although the cross sections for the formation of fission products are low, a peak which can be attributed to the distribution of fission products is discernible. Third, although there is such a peak due to fission products, it is very small and relatively flat and indicates that fragments due to fission may be spread over a very wide range of atomic numbers.

When the cross sections are plotted on a "chart of nuclides," i.e., on a graph of atomic number *versus* neutron number for all nuclides (*Z versus N* plot), this spread-out quality of the peak due to fission products becomes much more apparent. Maximum cross sections for a given mass lie fairly close to the main line of beta stability for all elements from sodium to indium and these maximum cross sections do not differ by much more than a factor of ten. This compares with ratios of approximately 10^3 for the total chain yield cross sections at the fission peak to those at the minimum in the region between fission and spallation for the products formed in the bombardment of both bismuth and uranium with 340-Mev protons. Attempts to make a detailed analysis of the peak due to fission products are hampered by the fact that it is so broad and low. If, on the *Z versus N* plot, a line is drawn between all nuclides having the same cross sections, reasonable interpolation being made between adjacent nuclides, curves such as those shown in Fig. 7 are obtained. If estimates of cross sections for the formation of stable nuclides are now made by interpolation between these isobarn lines and summations are made for each mass number, the solid curve of Fig. 6 may be drawn. This line will then represent the total chain yield at each mass number.

Cross sections for the formation of nuclides in the spallation region of tantalum are shown in the *Z versus N*

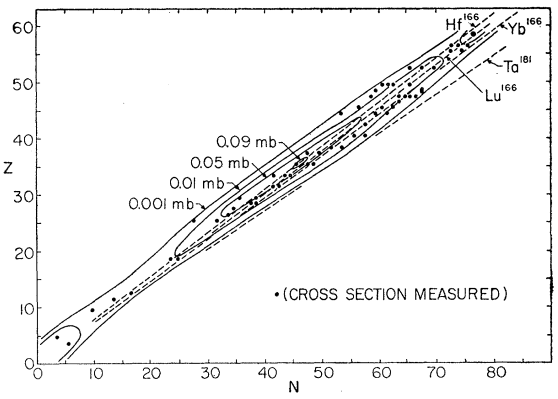

FIG. 7. Cross-section contour plot of the fission area.

[44] Luis Marquez, Phys. Rev. **86**, 405 (1952).

chart of Fig. 8. Mass numbers for which the plotted cross section represents total yield for the given nuclide plus all of its precursors are designated by arrows pointing down the chain. In the case of $Lu^{171+172}$ and $Ta^{176+180}$ the summed cross section listed in Table I has been divided by two and plotted in brackets.

Inspection of these figures leads to several very interesting conclusions. The peak of the fission product distribution curve in Fig. 6 lies at approximately mass number 83, indicating a "most probable fissioning nucleus" with a mass number of 166. From the contour lines of Fig. 7 it is not possible to say whether the peak cross section for the formation of the nuclide with mass number 83 lies at atomic number 35 or 36, either one of which seems probable. Biller, in his work on the reactions of bismuth[12] with 340-Mev protons, came to the conclusion that the peaks of his chain yields fell on a straight line having the same neutron to proton ratio as his most probable fissioning nucleus. Lines drawn with the neutron-to-proton ratios of $_{72}Hf^{166}$, $_{71}Lu^{166}$, and $_{70}Yb^{166}$ are included in Fig. 7. From the fit of these lines to the isobarn lines plotted it is apparent that either Hf^{166} or Lu^{166} is equally possible as the most probable fissioning nucleus while Yb^{166} has too low an atomic number to fit the data. Thus it may be said that the most probable fission process is that in which the Ta^{181} target atom loses sixteen nucleons (counting the incident proton) before the fission process takes place, and that these nucleons are boiled off in the ratio of 2.5 protons to 13.5 neutrons. These data are not inconsistent with Biller's results on bismuth, where two protons and 19 neutrons must be boiled off to give the most probable fissioning nucleus, or with the results of Folger, Stevenson, and Seaborg[20] and Douthett and Templeton[45] with uranium and 340-Mev protons, where two protons and approximately 20 neutrons are emitted prior to the most probable fission event.

From a study of the shape of the total chain yield *versus* mass number curve of Fig. 6 three separate areas may be delineated: first, the area of very light masses (and probably relatively high cross sections for formation) which represent small fragments emitted as part of the spallation process; second, the fission area corresponding to fragments having comparable sizes, or sizes larger than those normally considered as being involved in spallation reactions; and third, the area involving end products of spallation reactions in which only relatively small particles are ejected from the excited nucleus. It is evident that these areas are not very sharply defined for this type of bombardment; one cannot say precisely where the spallation process ends and fission begins, or where fission ends and the small spallation fragments begin. As a matter of fact, such distinctions are largely a matter of nomenclature and definition. For the sake of argument, however, let us take the area under the total chain yield curve between

[45] E. M. Douthett and D. H. Templeton, Phys. Rev. **94**, 152 (1954).

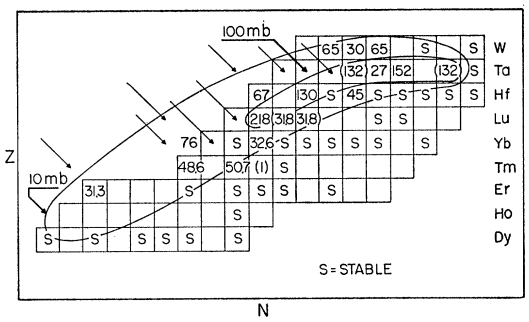

FIG. 8. Cross sections in the spallation region of tantalum.

the two minima in Fig. 6 (mass numbers 20 through 132 inclusive) as a measure of the total fission cross section. Upon integrating this portion of the curve and dividing by two for a binary fission process, the total fission cross section obtained for 340-Mev protons on tantalum is 4.1 ± 1.5 mb. This compares with 2000 mb for uranium,[20] 239 ± 30 mb for bismuth,[18] and approximately 10^{-1}–10^{-2} mb for silver[17] under the same bombardment conditions.

Note should be taken of trends that are apparent in the distribution of fission products as the atomic number of the target material is increased. In silver, and to a lesser extent copper, indications are that in the fission region no symmetrical fission peak is detectable. Fission data for these elements are meager (i.e., five measured cross sections between mass-24 and mass-56 in the silver bombardments) so that no detailed information concerning the fission region is available. Even if such information were known, however, it seems probable that the indefinite extent of the spallation process would tend to make interpretation of the fission data difficult. These total reaction cross section curves may be considered as a superposition of spallation and fission cross section components and if spallation extends into the fission area the effect will be to raise the "wings" of the fission curve, i.e., make it appear as though asymmetric fission were more probable than is actually the case. About all that can be done in the lighter elements is to set an upper limit to the total fission cross section, in the absence of any detailed knowledge about the distribution of the mass numbers of the fission fragments.

This "contamination" of the fission peak by products which may be due to spallation is negligible for bismuth and uranium. Here fission is a predominant reaction and the fission cross section peak height is such that the ratio between cross sections at the fission peak and those at the minimum in the region between fission and spallation is roughly 10^3. For these elements the conclusion has been drawn that binary fission into roughly equal sized fragments is the most probable type of fission reaction.

The tantalum fission data seem to indicate a picture which lies between these two extremes. From the shape

of the total chain yield *versus* mass number curve in Fig. 6 we may consider that the fission region extends roughly from mass number 20 to mass number 132. There will be overlapping between spallation and fission curves at the two extremes of this region, of course, but it is difficult to see how spallation, i.e., emission of a long series of small particles, will extend to products far enough into the fission region to change the shape of the fission curve significantly. It would appear, therefore, that the spread-out quality of the tantalum fission peak is real and that asymmetric fission is relatively more probable than in either bismuth or uranium. As a quantitative measure of this tendency toward asymmetric fission, let us compare the ratio of the cross section for symmetrical fission to that for a process in which the fissioning nucleus splits into fragments having masses in the ratio 70:30. Consideration of the data leads to a value of 5 for this ratio in tantalum fission. In uranium[20] the 50:50 split is approximately 50 times more probable than the 70:30 process and in bismuth[18] this ratio is about 90. The latter two ratios are made somewhat uncertain by the steep slopes of the fission product distribution curves for bismuth and uranium, but they do indicate a trend which suggests for their explanation an extension of the line of reasoning used to explain the change in shape of the double humped thermal neutron uranium fission product distribution curve as the energy of the bombarding particle is increased. Included in this reasoning is the thought that at the higher energies the nuclear shell effects which probably contribute to asymmetric fission with thermal neutrons are overshadowed as a result of large nuclear excitation and therefore symmetrical fission becomes a favored reaction. The fact that the above-mentioned ratio is lower for uranium than for bismuth may indicate that the low-energy asymmetric fission process is still important enough with 340-Mev protons to broaden somewhat the uranium fission peak while low-energy fission does not occur with bismuth. This interpretation seems reasonable in view of the fact that a very wide range of excitation energies may be imparted to the target nucleus and that even in uranium nuclei excited to about 40 Mev the peaks corresponding to asymmetric fission may be seen.

Since tantalum, like bismuth, does not undergo fission with low-energy particles, the question arises as to why the asymmetric fission of tantalum is so much more probable than that of bismuth. The small magnitude of the tantalum total fission cross section and the large energy required to effect the fission reaction indicate that a larger excitation energy is required to get tantalum into a fissionable state than is necessary for either bismuth or uranium. Perhaps it may be inferred that these very high excitation energies tend to destroy the fissioning nuclides' preference for roughly symmetrical fission. Thus we have asymmetric fission favored at low energies and symmetric fission at high bombardment energies with asymmetric fission becoming important again in those cases where the nucleus must take on high excitation energies as a prerequisite to undergoing the fission process.

Incident protons with 340 Mev of energy may impart a very wide range of excitation energies[46] to a target nucleus such as tantalum. It is interesting to try to determine the relative probabilities of the various type spallation reactions and the fission reaction in tantalum at the various excitation energies. The spallation yield data as summarized in Fig. 8 indicate that spallation reactions, with neutron emission favored over proton emission, are by far the most important of the competing reactions. The fission process does not seem to be significant. At higher excitation energies, after a fairly large number of neutrons and possibly one or two protons have been evaporated, emission of charged particles and fission both become increasingly important. Even under these circumstances, however, it is interesting to note that the fission process does not have nearly as large a yield as the spallation process. This can be deduced from the yield data by the following approximate method of reasoning. Let us assume that most of the spallation products below mass number 166 are formed by a spallation process in which they pass through intermediate nuclides with mass number 166 or near 166. Integration of the spallation product yield curve below mass number 166 and comparison with the total fission cross section should then give an indication of the relative probabilities for spallation and fission for those tantalum atoms which had at least sufficient excitation energy to evaporate sixteen nucleons. Treatment of the data in this manner shows a total spallation cross section of about 600 mb for the formation of products with mass number less than 166 as against a total fission cross section of 4.1 mb. In view of the fact that we are considering a region of intermediate fissioning nuclei rather than a single nuclide, and that this region should extend to mass numbers higher than 166, this ratio of fission to spallation yield must be regarded as only very approximate. As an order of magnitude, however, it shows that even when tantalum nuclei receive sufficient initial excitation energy to enable them to undergo the fission reaction, less than 1 percent of the nuclides actually do split; the remainder dissipate their excitation energy by the spallation process.

The spallation yield data of Fig. 8 indicate that the highest cross sections are observed for those reactions in which the incident proton is not captured and only a few neutrons are evaporated. The relative probability of a proton being emitted with these first few neutrons could not be determined because the hafnium isotopes in this region which would be used for detection are stable rather than radioactive as required by the technique used in this research. However, from the magnitude of the cross section for the production of Hf^{175} it may be inferred that this probability is small compared

[46] R. Serber, Phys. Rev. **72**, 1114 (1947).

to that for neutron evaporation. These data also indicate that spallation of as many as 21 nucleons has a yield lower than the maximum by a factor of only 7.

In general it is possible by radiochemical techniques to detect only the end products of spallation reactions and not possible to establish the mechanism by which such products are formed. Thus a reaction in which four alpha particles are emitted leads to the same nucleus as one in which eight deuterons or eight protons and eight neutrons are emitted. However, some interesting conclusions can often be drawn by comparing the yields of neighboring shielded and unshielded nuclides. For example, some information concerning the mode of formation of the spallation products may be inferred from a comparison of the cross sections for the formation of Tm^{167} and Tm^{168}. In this case Tm^{168} is a shielded nuclide and may be formed only by spallation of five protons and nine neutrons (or combinations of these) from the target tantalum nucleus. Thulium-167, however, in addition to the possibility of formation by direct spallation of five protons and ten neutrons (or combinations of these) from tantalum, can be formed by beta decay (positron emission or electron capture) of members of the mass number 167 chain of higher atomic number, i.e., Yb^{167}, Lu^{167}, Hf^{167}, Ta^{167}, and W^{167}. Since Tm^{167} and Tm^{168} are neighboring isotopes, it is reasonable to assume that their cross sections for direct formation by spallation are comparable. The observed cross section for the formation of Tm^{167} is approximately fifty times that for Tm^{168}. The conclusion naturally follows that most of the observed cross section for the formation of Tm^{167} must be due to contributions of its precursors and that the major portion of the 167 chain yield lies in nuclides having atomic numbers greater than 69.

V. ACKNOWLEDGMENTS

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