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HIGH ENERGY SPALLATION PRODUCTS OF COPPER

Daniel R. Miller

July 1, 1948

Berkeley, California

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ABSTRACT

About twenty-five radioisotopes, belonging to eleven different chemical elements, have been identified as products of the reaction of 190 Mev deuterons with natural copper; the reaction of high-energy helium ions with copper has also been investigated. The chemical procedures by which the elements were isolated and purified are presented. The observed yields of the various products are discussed with respect to the possible modes of excitation of the target nuclei and the manner by which the excited nuclei might dissipate their energy.

Two previously unreported isotopes have been observed in the course of this work: Zn^{62}, decaying by orbital electron capture with a 9.5-hour half-life; and Fe^{52}, decaying by positron emission with a 7.8-hour half-life.

To be declassified for use as a thesis.

HIGH ENERGY SPALLATION PRODUCTS OF COPPER

Daniel R. Miller

Radiation Laboratory, University of California
Berkeley, California
July 1, 1948

I. INTRODUCTION

The production of beams of deuterons and helium ions with energies of approximately 200 and 400 Mev, respectively, by the 184-inch frequency-modulated cyclotron has made possible investigations concerning the transmutation effects which particles of this high energy cause when they are used to bombard various chemical elements. Previously, the maximum available energies were only about one-tenth those now obtained with the 184-inch cyclotron. The transmutation products produced by bombardment of numerous elements with these lower energy particles have been extensively investigated[1,2]; the compound nucleus formed on consolidation of the target nucleus and bombarding particle possesses up to nearly 40 Mev of excitation energy, which is dissipated by expulsion of a maximum of 6 or 7 nucleons. The resulting product nuclei therefore have a mass and charge of within several units of the original target nucleus.

The manner and products of the transmutation of nuclei excited to the extent of several hundred Mev presents a much more complex problem. As many as about fifty nucleons might be emitted by a nucleus with this amount of excitation, but one might also expect that ejection of only a few nucleons would also occur from nuclei excited to a lesser degree. The anticipated transmutation products would therefore consist of a number of isotopes of numerous masses in each of ten to twenty different chemical elements. The process just described, wherein the target nucleus is transformed to a different product nucleus by the emission of any number of elementary particles, has been termed "spallation". In addition to these spallation reactions, heavier nuclei can also undergo fission by bombardment with high energy

charged particles.

The general program of this laboratory has included an investigation of the spallation and fission reactions induced by the bombardment of a number of chemical elements with 200 Mev deuterons and 400 Mev helium ions; specifically, this program includes the spallation reactions of arsenic[3] and antimony[4], the fission reactions of bismuth, lead, thallium, platinum, tantalum[5,6], and the fission and spallation reactions of uranium[7,8]. This thesis is concerned with a study of the identity and yields of the spallation products resulting from high energy deuteron and helium ion bombardment of copper (stable mass numbers 63 and 65).

Attempts were not made to identify stable products, and in general only those radioisotopes having half-lives of greater than about ten minutes and less than about one hundred days could be investigated. Since product isotopes of many different atomic numbers (Z) and mass numbers (A) are expected and found, identification of the individual products is possible only after rigorous chemical separations and purifications, in order to obtain pure elemental fractions containing isotopes of only one Z, followed by radiometric measurements of half-life and radiation characteristics to identify the mass numbers of the isotopes in these elemental fractions. The following sections will describe these experimental methods and the results which were obtained. These results will then be discussed in terms of their significance with respect to the probable modes of excitation of the target nuclei and transformation of these excited nuclei to give the observed products.

II. EXPERIMENTAL METHODS

A. Bombardments

All the results reported herein were obtained by bombardment with 190 Mev deuterons or 380 Mev helium ions produced by the 184-inch cyclotron of the Radiation Laboratory, University of California[9]. Such high energies are attained by application of frequency-modulation to the established principles of smaller cyclotron operation[10]. The positively charged nuclear particles (deuterons) enter at the center of the vacuum tank and revolve in a uniform magnetic field. As the energy increases, in step with the oscillating voltage on the "dee", the particles spiral outward, just as in the smaller cyclotrons. However, the deuterons, as they increase in velocity, increase in mass due to the relativistic effect; deuterons at 200 Mev have suffered a 10% mass increase. Therefore, to prevent the particles from lagging, the oscillating frequency of the dee voltage in the 184-inch cyclotron is continuously changed. This accelerator is therefore called the "frequency-modulated" cyclotron, or "synchro-cyclotron".

A method for deflecting the beam out of the vacuum tank had not been devised during the course of these bombardments, so the target was bombarded inside the tank. The metallic copper targets used were of two classes of purity: commercial copper and special purity copper. The kind and amount of impurities present in the first type are not known, but ordinary metallic copper is usually greater than 99.5% pure, the main impurities being O, S, Fe, and Ni. The high purity copper metal was analyzed spectrographically by the supplier (American Smelting and Refining Co.) and the maximum amount of any impurity was stated to be 0.0001%; the probable purity was computed by difference to be >99.9995%. The commercial metal was 1/64" sheet, and the high purity metal was 1/100" sheet. A 2 mm. wide strip of the sheet was bombarded, and then the most active 1 cm. length of the strip was subjected to the chemical processing; this amounted to about 70 mg. of the commercial copper and about 45 mg. of the high purity copper.

The cross-sectional area of the synchro-cyclotron beam is about one square centimeter; the 0.2 x 1.0 cm. target therefore intercepted about one-fifth of the total beam. The intensity of the deuteron or helium ion beam varied and moreover has not been accurately measured. Best estimates are that a beam of about one microampere of deuterons or about 0.2 microampere of helium ions was obtained in the later stages of this work. The length of the bombardment also varied, but averaged about 15-30 minutes. A summary of these bombardment details is given in Table 1.

Table 1

Bomb. No.	Bomb. Particle and Energy	Est. Beam Current	Length of Bomb.	Purity Grade of Copper
D-1	190 Mev D^+	0.2 μamp.	15 min.	Commercial
D-2	"	0.5	15 "	"
D-3	"	0.5	15 "	"
D-4	"	0.5	25 "	"
D-5	"	1.	12 "	"
D-6	"	1.	20 "	"
D-7	"	1.	27 "	High
D-8	"	1.	22 "	"
D-9	"	1.	*	"
D-10	"	1.	25 min	"
A-1	380 Mev He^{++}	0.05	40 "	"
A-2	190 Mev He^{++}	0.25	32 "	"

* Consisted of intermittent one-to five-hour bombardments over a period of about two months; total bombardment time about 15 hours.

B. Chemical Separations

In all bombardments except D-9, the chemical purification procedure was started 10 minutes or less after the end of the bombardment. The copper target strip was dissolved in about 0.5 ml of hot, concentrated nitric acid, and then by appropriate chemical operations the elemental fractions of interest were purified and isolated. In general, the elements were removed successively from the entire dissolved target solution. Before the chemical fractionations were begun, 2-5 mg each of inactive carrier elements were added. Usually carriers for each

element from $_{21}Sc$ through $_{30}Zn$ were added, even though only several of the included elements were to be isolated; the inactive atoms of the elements not to be removed specifically then acted as "holdback carriers", or diluents, for the active species of that element, and tended to decrease the probability of their appearance in the isolated fractions of other elements. Most of the chemical operations were performed in total volumes of 1-15 ml. Precipitates were removed from solutions by centrifugation.

Since different combinations of elements were removed in each investigation, the chemical procedure varied from bombardment to bombardment. The general principles utilized for the separation and purification of any one element were the same for different runs, however, and the overall procedures varied mainly in the manner in which the individual elemental procedures were joined together. For this reason the fractionation schemes used for each experiment will not be specifically discussed; the vital steps in the purification and isolation of each element will be presented, and one typical procedure joining several of these steps will then be outlined.

The chemical yields, i.e., fraction of the original amount of added element recovered in pure form after the chemical operations, were not determined. It was obvious from qualitative observations, such as size of centrifuged precipitates, etc., that most of the final recoveries were in the range of 50-100%. Any value in this range was deemed satisfactory, since no concerted attempt to increase the accuracy of the final results to better than a factor of two was made.

1. Isolation of Gallium. The main principle of this separation was the extraction of the gallium chloride complex from 6 M HCl into diethyl ether. This is specific against all elements in this region of the periodic table except iron, which also extracts quantitatively under these conditions. Gallium-iron separations were performed in two ways; (a) the iron was reduced to the divalent state in

6 M HCl by $SnCl_2$, the gallium extracted with ether, and the iron then extracted with ether following oxidation with a slight excess of $KMnO_4$; (b) $Fe(OH)_3$ was precipitated from an aqueous solution containing excess NH_4OH or NaOH, leaving the gallium in the supernatant. Gallium was removed from the ether phase either by extraction back into water, or by evaporation of the ether solution and dissolution of the residue in water or acid. The extraction and iron-separation procedures were repeated two or three times in order to insure purity of the final preparation.

2. Isolation of Zinc. Following removal of copper, ZnS was precipitated from a dilute acetic acid solution at 80° C and dissolved in concentrated HCl. After boiling to remove the excess H_2S, iron was added and precipitated as the hydroxide with excess NH_4OH or NaOH. The supernatant was acidified with acetic acid and ZnS again precipitated. This cycle was repeated several times, the hydroxide precipitations being done alternately with NH_4OH and NaOH. The combination of the ZnS precipitation from dilute acid, and the two types of $Fe(OH)_3$ scavenging precipitations, leads to purification of zinc from all other spallation products of copper.

3. Isolation of Copper. Two types of purification by precipitation were used for the removal of pure copper. (a) reduction of Cu^{++} to Cu^{+} by Na_2SO_3 and heat, followed by precipitation of CuSCN from 0.5-1.0 M HCl, using a slight excess of NaSCN as precipitant; (b) CuS precipitation from 3 M HCl or 3 M H_2SO_4, using gaseous H_2S as precipitant. The CuSCN was dissolved in concentrated HNO_3, and the CuS was dissolved in concentrated HCl with the aid of $NaClO_3$. In order to be assured of final purity, at least two precipitations were performed--either all of the same type, or a mixture of the two types. Either type is chemically specific against all other elements below atomic number 31.

4. Isolation of Nickel. The main step in the purification and separation of nickel was the well-known method of precipitation of nickel dimethylglyoxime. These precipitations were carried out from buffered acetic acid solutions. One or two milligrams of La^{+3} were added to the approximately neutral (with NH_4OH) solution before the glyoxime was added; acetic acid was added until the $La(OH)_3$ just dissolved, and the Ni^{++} then precipitated with an alcoholic solution of dimethylglyoxime. This procedure minimized the carrying of hydroxide-insoluble elements on the nickel dimethylglyoxime which would occur if the precipitation were conducted around neutrality. The precipitate was dissolved in a small amount of concentrated HNO_3, and the procedure repeated at least once.

5. Isolation of Cobalt. All cobalt separations included the precipitation of potassium cobaltinitrite, $K_3Co(NO_2)_6$. An excess of K^+ (as KCl or $KC_2H_3O_2$) was added to the dilute acetic acid solution of Co^{++}, and the $K_3Co(NO_2)_6$ precipitated by addition of KNO_2. The precipitate could be dissolved in concentrated HCl and the procedure repeated. Extraction of the cobalt thiocyanate complex into amyl alcohol was also used to a limited extent. A solution of Co^{+2} in about 0.5-1 M HCl was made to contain about 30 weight percent of NaSCN, and shaken two or three times with an equal volume of amyl alcohol. The cobalt was removed from the organic phase by shaking with a dilute NaOH solution, from which the cobalt precipitated as the hydroxide. This hydroxide was dissolved in acetic acid and a $K_3Co(NO_2)_6$ precipitation conducted. The extraction procedure is not adequately specific by itself (e.g., chromium also extracts to some extent), and must be used in conjunction with the $K_3Co(NO_2)_6$ method if used at all.

6. Isolation of Iron. Iron was purified by using the ether-extraction method discussed under gallium. The procedure always involved at least two extractions from 6 M HCl into ether and at least one $Fe(OH)_3$ precipitation by addition of NH_4OH or NaOH. When gallium activities were present in high yields (in the helium

ion bombardments), the procedure also included an iron-gallium separation by the $SnCl_2$ reduction method outlined under gallium.

7. Isolation of Manganese. Precipitation of manganese dioxide from strong HNO_3 yielded radiochemically-pure manganese fractions. A solution of Mn^{++} in 10-15 M HNO_3 containing $NaClO_3$ was boiled vigorously for 2-4 minutes. Further purification was performed by dissolving the MnO_2 in concentrated HNO_3 containing a small amount of H_2O_2, boiling to destroy the excess H_2O_2, and then repeating the MnO_2 precipitation as above. In one experiment further purification was attempted by precipitation of tetraphenyl arsonium permanganate from dilute H_2SO_4 solution, following oxidation to MnO_4^- by $NaBiO_3$. Since tetraphenyl arsonium permanganate was difficult to dissolve, this procedure was not regularly employed, especially since several MnO_2 precipitations as above gave satisfactorily pure fractions.

8. Isolation of Chromium. The common step in the isolation of all chromium fractions was the extraction of peroxychromic acid from dilute acid solutions into diethyl ether. Trivalent chromium was oxidized to $Cr_2O_7^=$ by the action of ClO_3^- in hot 10-15 M HNO_3, usually as a part of the MnO_2 precipitation step. After oxidation (and after removal of the MnO_2), the solution was made slightly acid (0.05 - 0.2 M) in HNO_3, by dilution and/or neutralization. The diluted solution was cooled in an ice bath, and an equal volume of ether and sufficient H_2O_2 to make the aqueous phase about 0.3% in H_2O_2 were then added. After agitation, the ether was removed and a second volume of ether added in order to remove completely the blue peroxychromic acid. The ether extracts were evaporated to dryness and the oxidation and extraction procedures repeated at least one additional time. An H_2O_2 oxidation in alkaline solution followed by acidification and extraction were attempted in one bombardment (D-10), but an extremely low chemical yield was obtained. The alkaline oxidation procedure is basically sound and would be more convenient in

many fractionation schemes.

9. Isolation of Vanadium. The only useful procedure found to have any degree of specificity for vanadium was the precipitation of the alpha-benzoinoxime compound of pentavalent vanadium from about 0.5 M HCl. The only possible contaminant among the spallation products of copper is hexavalent chromium. The chromium-vanadium separation was performed by first insuring the presence of all chromium in the trivalent form by the addition of a small amount of Na_2SO_3; this also reduced the vanadium, which was then reoxidized to V^{+5} with a small excess of NaOCl. The chromium was now trivalent and the vanadium pentavalent; addition of an alcoholic solution of alpha-benzoinoxime precipitated the vanadium and not the chromium. The difficulty with this precipitation procedure was the difficulty of dissolving and re-precipitating the vanadium oxime, which was necessary to obtain a radiochemically pure fraction. The dissolution was attempted by boiling the precipitate with concentrated HCl and concentrated HNO_3; eventually an almost clear solution was obtained. After boiling to dryness several times and making up to 0.5 M HCl, however, addition of alpha-benzoinoxime led to precipitate formation only occasionally. The reasons for the usual failure to obtain a precipitate are not known. The precipitate could probably be brought into solution by fuming with H_2SO_4 or $HClO_4$, although this method might be too time-consuming to allow observation of the shorter-lived activities.

10. Isolation of Titanium. The precipitation of potassium titanium iodate from 6 M HNO_3 was used for the purification and removal of titanium. Insufficient work was done on titanium activities to be absolutely certain that radiochemically pure fractions were obtained by this method; this precipitation should be specific against possible contaminating elements, however, judging by the known properties of the elements with atomic numbers below 31. The procedure used was as follows:

a large excess of solid KIO_3 was added to a 6 M HNO_3 solution of Ti^{+4}; the precipitate was separated after about 5 minutes and dissolved in 6 M HCl through which gaseous SO_2 was bubbled. Titanium hydroxide was precipitated, after dilution, by the addition of excess NH_4OH. The hydroxide was dissolved in 6 M HNO_3 and the cycle repeated once or twice more. In one bombardment (D-10) these cycles (two) were preceded by precipitation of titanium phosphate from 0.5 M HCl by addition of solid Na_2HPO_4; the phosphate was dissolved by heating with concentrated HCl, which was then diluted to about 2 M and KIO_3 added to precipitate the double iodate. In the other bombardment (D-9) three iodate-hydroxide cycles were followed by precipitation of titanium phenylarsonate from ca. 0.5 M HCl.

11. Isolation of Chlorine. As previously stated, the metallic copper target was dissolved in concentrated HNO_3. When an investigation of radiochlorine was desired, the target was dissolved in a small distilling flask with an air-cooled condenser arm, the end of which extended just below the surface of about 2 ml of H_2O in a tube placed in a beaker of cold water. The copper was dissolved by heating the distilling flask containing the target and 1-2 ml of HNO_3 (plus about 10 mg Cl^-), and the chlorine simultaneously evolved into the H_2O in the receiving tube. After the residue in the flask was heated to near dryness, the solution in the receiving tube was diluted to 5-10 ml and a small excess of Ag^+ added, as $AgNO_3$. The AgCl was digested for several minutes at 90° C, separated, dissolved in NH_4OH, and reprecipitated by addition of HNO_3 in slight excess. Up to three such AgCl precipitations were conducted. A rough chemical yield determination through the distillation portion of this procedure indicated an 85-90% recovery of the added chlorine.

12. Isolation of Phosphorus. A phosphorus fraction was removed from only one bombardment (D-9); the procedure follows. Ammonium phosphomolybdate was precipitated from ca. 2.5 M HNO_3 under the standard analytical conditions[11].

This precipitate was washed and dissolved in NH_4OH. The solution was made slightly acid with HCl, an excess of Mg^{++} added, and $MgNH_4PO_4$ precipitated by addition of excess NH_4OH. This precipitate was dissolved in 6 M HNO_3 and the cycle repeated three more times. An aliquot of the final solution was prepared for counting. An additional three cycles were then performed on the remainder of the solution; in the first and third of these cycles, the ammonium phosphomolybdate precipitations were performed in the presence of about 0.5% H_2O_2, and in the second cycle in the presence of $SO_3^{=}$. These last three cycles were specifically designed to free the phosphorus of titanium and vanadium, which might have carried through the procedure as titanium phosphate and vanadomolybdate. Judging by relative sizes of the precipitates through the entire procedure, a chemical yield of about 70% was obtained.

13. Typical Fractionation Procedure. Since in any one bombardment practically all elemental fractions were successively removed from the same dissolved target solution, several group separations were performed during the course of the removal of the individual elements by the methods just outlined. Hydroxide precipitation was the main method for separating the elements into two groups. Considering the elements from Ti through Zn, only Fe, Mn, Cr^{+3} and Ti are precipitated by excess NH_4OH; V^{+5} is not precipitated when it alone is present, but is partially precipitated in the presence of other hydroxides. Cu, Ni, Co, Fe, Mn, Cr^{+3}, and Ti are precipitated by excess NaOH or KOH, although Cu and Cr^{+3} are amphoteric to a limited extent; the same remarks as above about V^{+5} apply here. Sulfide precipitations from NH_4OH solutions were used occasionally as a volume reduction step. All elements under consideration except perhaps gallium and vanadium are precipitated by this method. In order to illustrate the use of the group separations and the joining of the various procedures for the individual

elements, the procedure used for bombardment D-7 is reproduced in Figure I. The details of this procedure are those given in the preceding paragraphs. The chemical operations of this procedure were completed in about 3.5 hours.

The various elements which were isolated from each of the several bombardments are listed in Table 2.

Figure I.

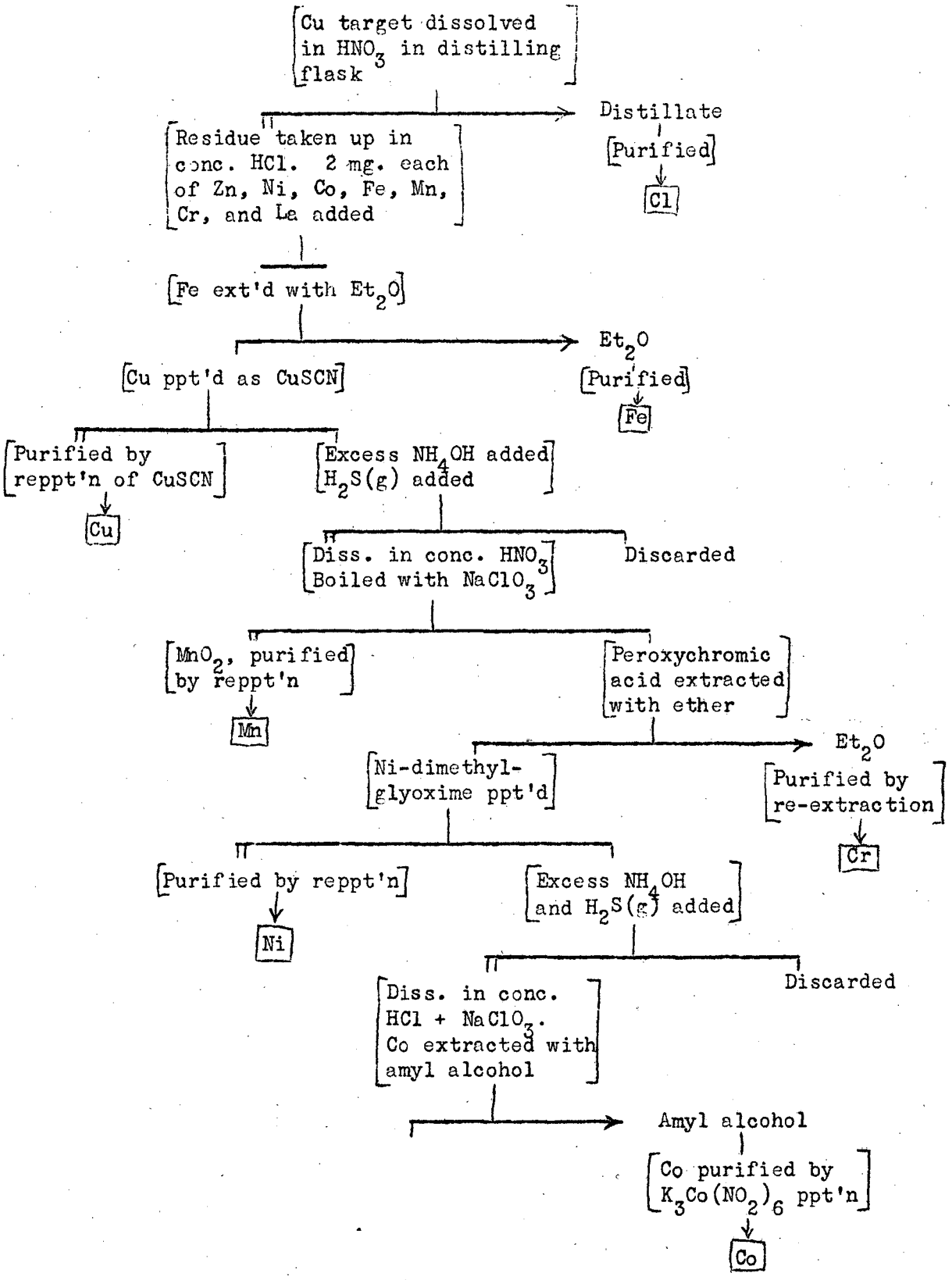
Cu target dissolved in HNO_3 in distilling flask
Distillate
Purified
Cl
Residue taken up in conc. HCl. 2 mg. each of Zn, Ni, Co, Fe, Mn, Cr, and La added
Fe ext'd with Et_2O
Et_2O
Purified
Fe
Cu ppt'd as CuSCN
Purified by reppt'n of CuSCN
Cu
Excess NH_4OH added $H_2S(g)$ added
Diss. in conc. HNO_3 Boiled with $NaClO_3$
Discarded
MnO_2, purified by reppt'n
Mn
Peroxychromic acid extracted with ether
Et_2O
Purified by re-extraction
Cr
Ni-dimethyl-glyoxime ppt'd
Purified by reppt'n
Ni
Excess NH_4OH and $H_2S(g)$ added
Discarded
Diss. in conc. HCl + $NaClO_3$. Co extracted with amyl alcohol
Amyl alcohol
Co purified by $K_3Co(NO_2)_6$ ppt'n
Co

Table 2

Bombardment Number	Elements Isolated and Purified
D-1	Zn, Cu, Ni, Co, Fe, Mn, Cr
D-2	Zn, Cu
D-3	Zn
D-4	Cu, Fe, Cr
D-5	Ga, Cu
D-6	Cu, Fe
D-7	Cu, Ni, Co, Fe, Mn, Cr, Cl
D-8	Cu, Cl
D-9	Zn, Fe, Mn, V, Ti, P
D-10	Cu, V, Ti
A-1	Ga, Cu, Ni, Fe, Mn, Cr, Cl
A-2	Cu, Cl

C. Radiometric Identification

Aliquots of the purified elemental fractions were mounted and dried on one-inch discs of platinum (D-1 through D-9 and A-1) or of stainless steel (D-10 and A-2). The aliquots were of such a size that the initial activity in them, as determined in the arrangement to be described, was 5000-8000 counts per minute. If the dried samples were of a flaky nature, they were covered with a sheet of thin cellophane (3-5 mg/cm^2). The samples thus prepared were mounted on an aluminum sheet (D-1 through D-9 and A-1), or on a cardboard card(D-10 and A-2), and the intensity and properties of the radioactivity in them determined by the following methods and instruments.

The disintegration rate was measured with the aid of an end-window Geiger-Müller counting tube connected to a scale-of-64 counting circuit. The windows of the tube were mica, of an average thickness of about 3 mg/cm^2, with an "Aquadag" coating. The copper or brass shell tubes were filled with 9 cm of argon and 1 cm of ethyl alcohol vapor; the tungsten or Kovar central wire is insulated from the shell and placed at a high positive potential with respect to it. Tubes of this type showed the typical Geiger "avalanche action" starting at about 900-1000 V. applied potential, and had satisfactorily constant plateaus for 200-300 V. A shelf-like arrangement was used so that the sample to be counted could be placed in any of five positions of varying distance below the window of the Geiger-Müller tube: 3, 19, 35, 51, and 67 mm. The geometrical efficiencies of these positions are about 30%, 10%, 4%, 2.5%, and 1.5%, respectively. Two corrections were made to the recorded counting rate: (a) a coincidence correction of 1.2% for every thousand counts per minute; and (b) a background correction, determined periodically and averaging about 25 counts per minute.

The disintegration rate of each sample was followed as a function of time, and the decay curves obtained were analyzed for the component activities by the standard procedure of successively subtracting off the longest-lived member. These half-life determinations were the main evidence in the identification of the radioactive spallation products.

Confirmatory evidence for the presence of a certain radioisotope was also obtained in some cases by a determination of the energy of the radiation. This was done by counting through varying thicknesses of Al or Be for beta particles, and Pb for gamma rays[12]. The range of the beta particles was determined from these absorption curves either visually or by Feather analysis[12]; the range in Al or Be was converted into energy in Mev by use of Feather's relation[12]. The half-thicknesses of the gamma rays in lead were converted to energies with the aid of the theoretical relationship between the mass absorption coefficient for photons in lead and the energy of the photon[12].

In addition to the above identification methods, a crude beta-ray spectrometer was used to determine the sign of the beta radiation. A variable magnetic field is imposed to bend either the positive or negative particles through 180° into a Geiger-Müller counter tube described above. By varying the strength of the magnetic field and noting the counting rate as a function of the field strength, the approximate maximum energy could also be determined.

D. Calculations

The yield of a radioisotope was calculated by first determining, from the decay curve, the counting rate at the end of the bombardment, and then converting this to the disintegration rate by the appropriate geometrical efficiency for the counting position used. By correcting this by the chemical aliquot factor, the number of disintegrations per minute of the product which was present in the 70 or 45 mg. of target at the end of the bombardment was known. If the isotope decayed

by orbital electron capture, the counting efficiency of the radiation was also considered; a counting efficiency of 10% was assumed, quite arbitrarily, for any isotope decaying purely by orbital electron capture. If the half-life of the isotope was short compared to the length of the bombardment, the following expression was used to give the total disintegrations per minute of the isotope produced by the bombardment:

$$N_T = N_o \frac{t}{T_{1/2}} \cdot \frac{0.693}{1-e^{-0.693t/T_{1/2}}}$$

where N_T and N_o are the total number of product nuclei formed and the number of product nuclei present at the end of the bombardment, respectively, t is the length of the bombardment, and $T_{1/2}$ is the half-life of the isotope. N_T and N_o are proportional to the disintegration rates; the latter can be converted to the former by use of the well- known decay law.

The yields of the various product isotopes were expressed relative to the 3.4-hour Cu^{61}, on an atom basis. A relative yield of 2.0 for isotope X would therefore signify that two atoms of X were produced for every one atom of Cu^{61}. The results were originally expressed in this way because the uncertainty in the beam current made cross-section calculations of doubtful value. Since the beam current is still somewhat uncertain, it is still felt desirable to express the results as atom yields relative to Cu^{61}. However, cross-section calculations using the thin-target formula were made for several bombardments, using the estimated beam currents listed in Table 1. The results of these calculations for Cu^{61} (relative yield of one) are given in Table 3. These figures indicate that the relative yield (Y) and cross section (σ), in units of 10^{-24} cm^2, are related by the formula

$$\sigma = 0.06\,Y$$

The uncertainty in the absolute value for the cross section calculated by this relation is about a factor of two.

Table 3

Bombardment Number	Cross Section (in 10^{-24} cm^2) for formation of Cu^{61}
D-5	0.085
D-7	0.055
D-10	0.028
	Ave.= 0.056
A-1	0.057
A-2	0.069
	Ave.= 0.063

III. EXPERIMENTAL RESULTS

A. New Isotopes

1. Zinc-62. The zinc fraction from the first bombardment (D-1) showed a growth in activity for about one hour after isolation, and then decayed with a 9.5-hour half-life; the initial growth appeared to correspond to about a 10-minute half-life (Figure II). (The zinc fraction was not removed until about 4 hours from the end of the bombardment, and the 38-minute Zn^{63} was not observed.) These facts indicated that the 9.5-hour period belonged to a previously unreported zinc isotope of mass 62, and that the 10-minute growth was due to its daughter isotope, the well-known 10.5-minute, positron-emitting Cu^{62} *. Experiments in which copper was separated from the zinc fraction were conducted in order to confirm this interpretation. In bombardment D-2, several $Fe(OH)_3$ precipitates were separated from the Zn fraction by addition of KOH, in order to remove the copper activity; the copper fraction showed an activity with half-life of 10-minutes. In bombardment D-3, more specific and quantitative removal of copper was performed by precipitating CuSCN from the isolated zinc fraction. Five such separations were made over a period of about 30 hours; all separated copper fractions showed an activity of 9.5 to 11 minutes in addition to a small amount of the 9.5 hour zinc present as a contaminant. The decay curve of a typical copper fraction removed from the zinc fraction is reproduced in Figure III. The number of counts per minute of the 10-minute copper activity removed from the zinc was extrapolated back to the time of separation, and this figure was compared with the total counts per minute in the zinc fraction, calculated from its decay curve. The ratios of these figures were 1.14, 0.98, 1.05, 1.04 and 1.07 in the five separations.

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* The properties of all previously known isotopes are quoted from Seaborg[1], unless otherwise specified.

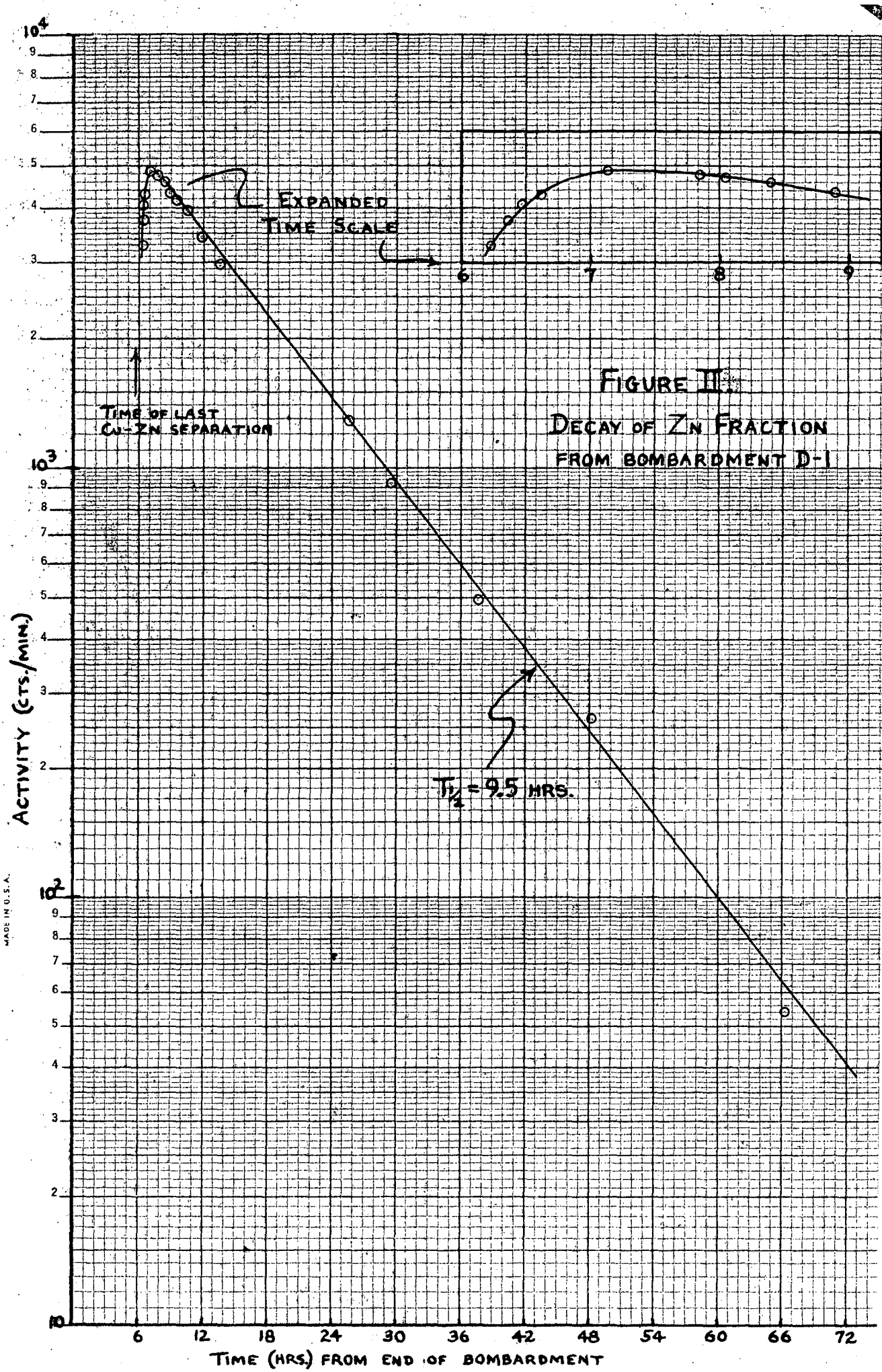


FIGURE II.
DECAY OF ZN FRACTION
FROM BOMBARDMENT D-1

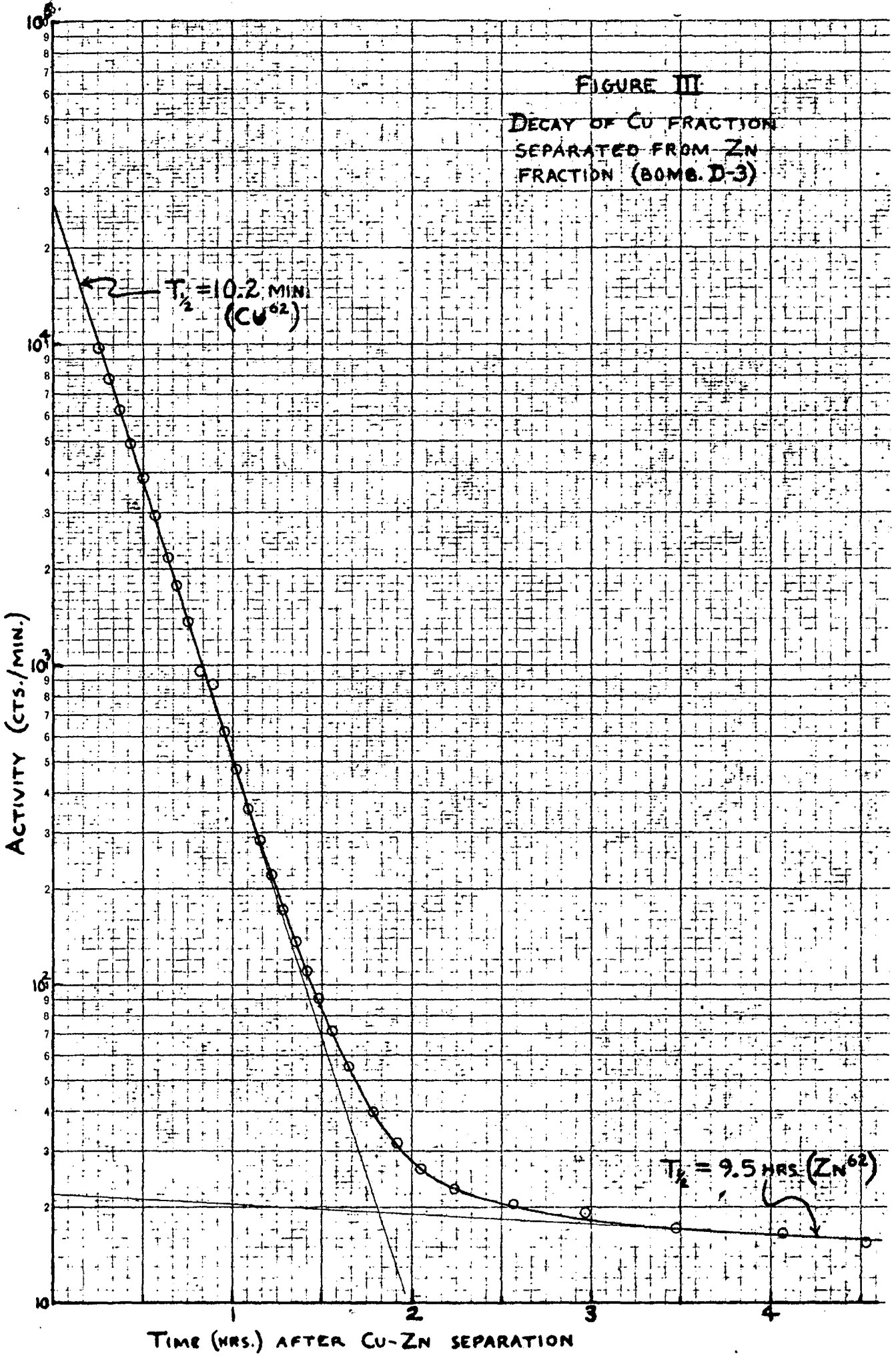


FIGURE III
DECAY OF CU FRACTION SEPARATED FROM ZN FRACTION (BOMB. D-3)
$T_{1/2}$ = 10.2 MIN. (Cu^{62})
$T_{1/2}$ = 9.5 HRS. (Zn^{62})
ACTIVITY (CTS./MIN.)
TIME (HRS.) AFTER CU-ZN SEPARATION
10^6
10^5
10^4
10^3
10^2
10
1
2
3
4

The fact that these ratios were approximately one indicates the relative absence of detectable radiation attributable to the 9.5-hour zinc parent; a ratio of 2.0 would have indicated that both parent and daughter decayed with radiations of equal counting efficiency. An aluminum absorption curve was taken on the zinc fraction from D-2 when it was showing a pure 9.5-hour decay (Figure IV); this curve indicated that only one particle was present, having a maximum energy of about 2.5 Mev. This corresponds well to the known energy of the Cu^{62} positron, 2.6 Mev.

These results all show that the radiation of Zn^{62} is either of energy too small to have been detected efficiently by the counting tubes used, or that its counting efficiency must be so low as to be negligible in comparison with its daughter. The latter possibility seems more probable, and Zn^{62} is presumed to decay by orbital electron capture.

2. Iron-52. A 7.8-hour activity was observed in the iron fractions of all bombardments from which they were removed; the decay curve of the iron fraction from bombardment D-4 is reproduced in Figure V. No particles of negative sign were observed at a time when the radiation from the 47-day β^--emitting Fe^{59} was in low relative concentration, indicating that the 7.8-hour isotope decayed by positron emission or orbital electron capture. To ascertain its mass number, several manganese separations were made from the iron fractions over a period of about five hours. These separations were carried out by extracting the manganese from the ether solution of iron by shaking with 6 M HCl. All manganese fractions removed from the iron showed small amounts of the 47-day and 7.8-Hour iron activities, but the main component was one of 21-minute half-life, corresponding to the positron-emitting Mn^{52}. The decay of one of these separated Mn fractions (bombardment D-6) is shown in Figure VI.

An aluminum absorption curve of the iron fraction from D-4 is reproduced in Figure VII; this curve was taken when the 7.8-hour activity represented

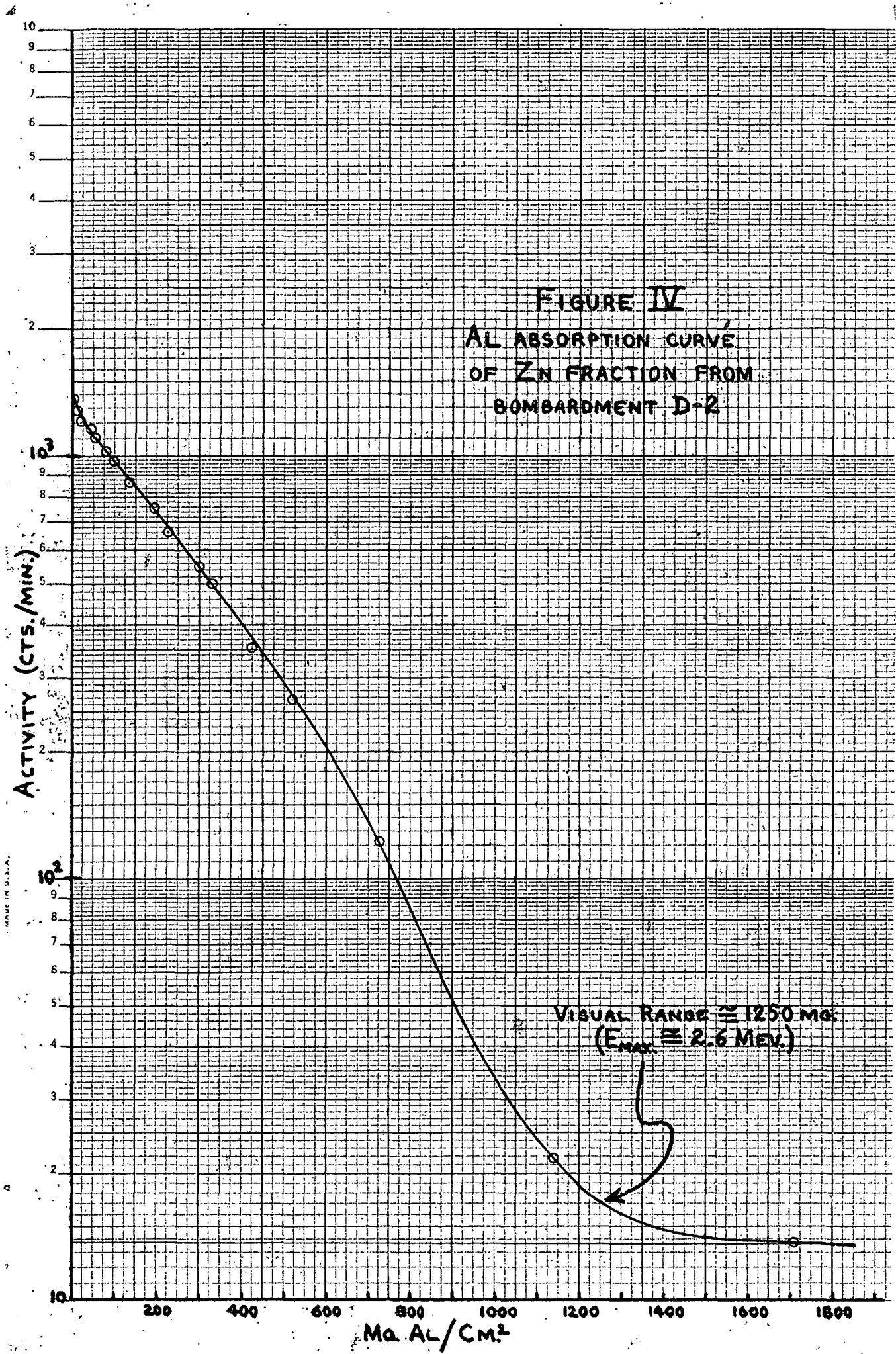


FIGURE IV
AL ABSORPTION CURVE
OF ZN FRACTION FROM
BOMBARDMENT D-2
VISUAL RANGE ≅ 1250 MG.
($E_{MAX.}$ ≅ 2.6 MEV.)
ACTIVITY (CTS./MIN.)
MG. AL/CM.²
10
10^2
10^3
200
400
600
800
1000
1200
1400
1600
1800

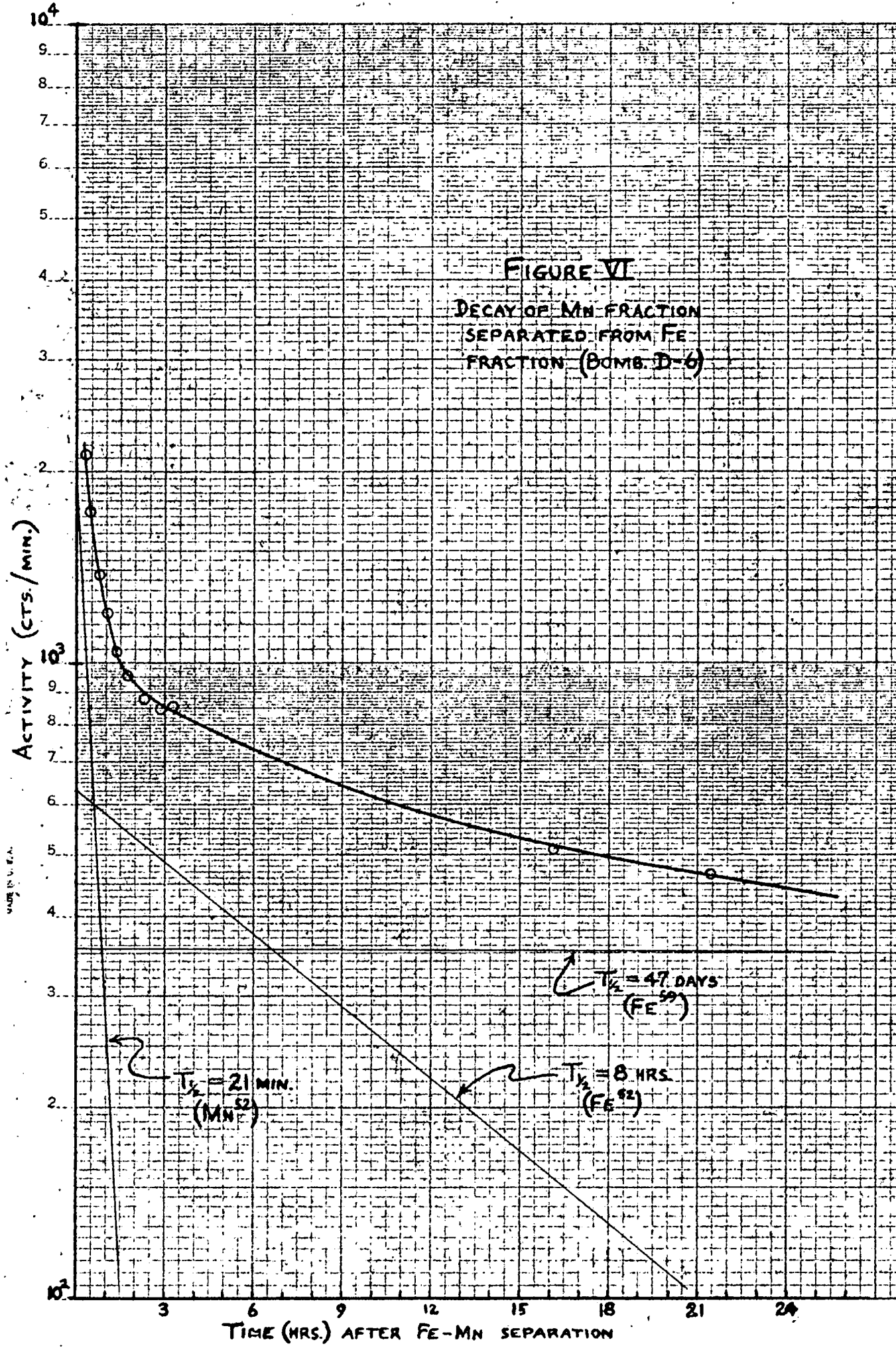


FIGURE VI
DECAY OF MN FRACTION SEPARATED FROM FE FRACTION (BOMB. D-6)
ACTIVITY (CTS./MIN.)
TIME (HRS.) AFTER FE-MN SEPARATION
$T_{1/2}$ = 47 DAYS (FE^{59})
$T_{1/2}$ = 8 HRS. (FE^{52})
$T_{1/2}$ = 21 MIN. (MN^{52})

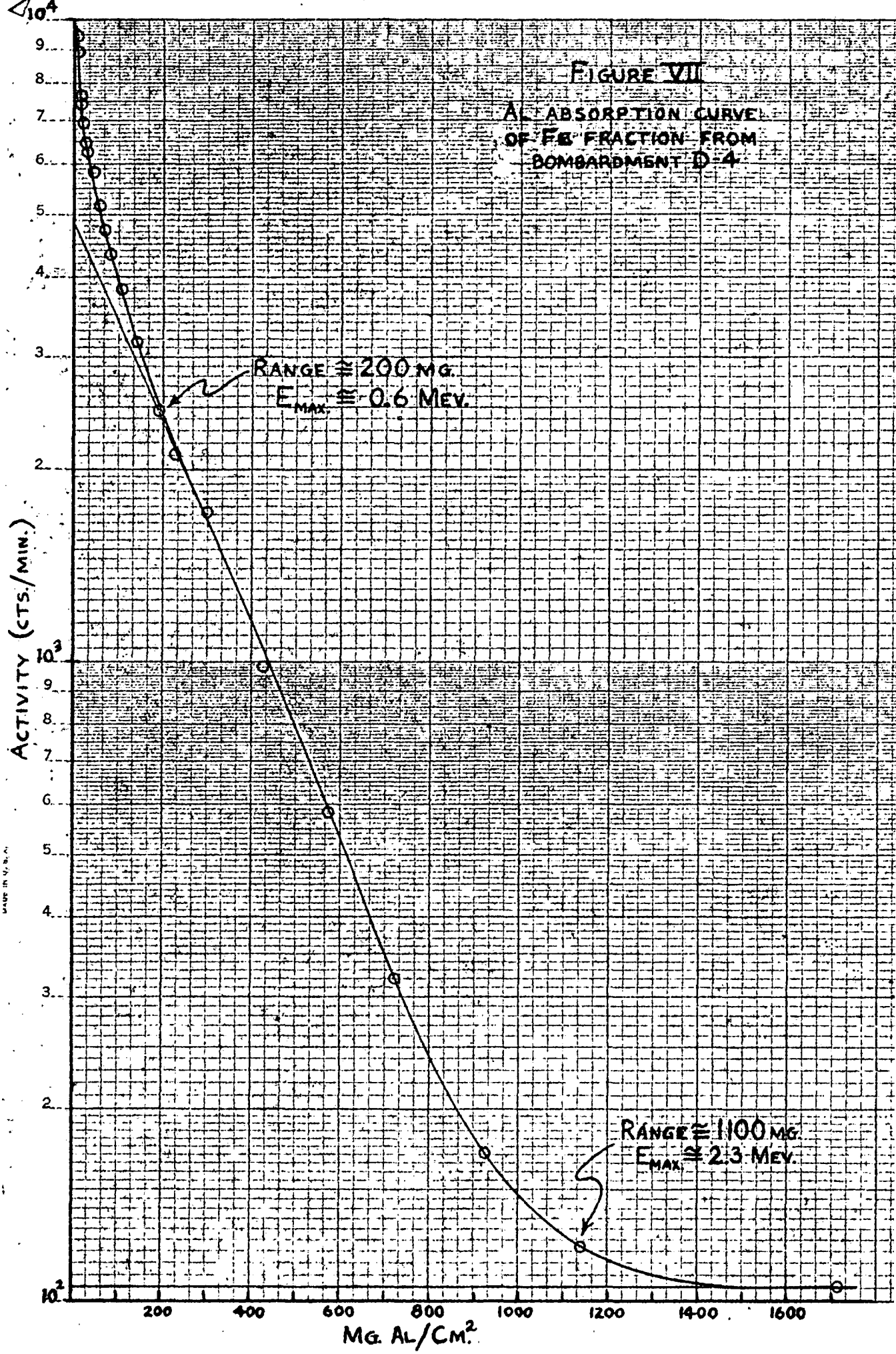


FIGURE VII
AL ABSORPTION CURVE OF Fe FRACTION FROM BOMBARDMENT D-4
RANGE ≅ 200 MG.
$E_{MAX.}$ ≅ 0.6 MEV.
RANGE ≅ 1100 MG.
$E_{MAX.}$ ≅ 2.3 MEV.
ACTIVITY (CTS./MIN.)
MG. AL/CM.²
10^4
10^3
10^2
200
400
600
800
1000
1200
1400
1600

practically all of the activity present. Visual examination of this curve indicates two components, one of energy about 0.6 Mev and the other of energy 2.3 Mev. The energy of the positron of the 21-minute Mn^{52} is reported to be 2.2 Mev; this confirms the assignment of the 7.8-hour iron to mass 52. The 0.6 Mev component is presumed to be the positron of Fe^{52}.

Mn^{52} decays by two independent paths: positron emission with a 21-minute half-life; and positron emission and orbital electron capture with a 6.5-day half-life. The 21-minute isomer has definitely been shown to be a daughter of Fe^{52}. No conclusive indications of a 6.5-day period were found in the manganese separated from the iron fractions. Because of the large difference in half-lives of the two Mn isomers, it can only be said with certainty that a minimum of 50% of the Fe^{52} atoms decay to the 21-minute Mn^{52}; this figure may be much higher in actuality.

B. Spallation Products of Copper

1. Isotopes of Gallium. An investigation of gallium isotopes was attempted following bombardment D-5; a relatively small amount of activity was found. Because of the apparent large number of components contributing to the total activity, however, no resolution of the decay curve was possible. The half-lives of the gallium isotopes of masses 64, 65, 66, 67, and 68, are 48 min., 15 min., 9.4 hrs., 83 hrs., and 68 min., respectively; it is possible that all of these periods were present in the gallium fraction. The mechanism of the production of isotopes of atomic number 31 (gallium) by deuteron bombardment of copper (atomic number 29) is not known, but there are three possible explanations. The deuteron beam might have been contaminated with a small amount of helium ions which could have caused alpha-multiple neutron reactions. Sufficiently energetic helium ions might also have been produced as products of the primary reaction of

copper plus deuterons. A third possible explanation involves emission of negative mesotrons (and possibly also neutrons) from the excited compound nuclei Zn^{65} and Zn^{67}. Since the identity and yields of the activities in the gallium fraction could not be determined, these possible mechanisms will not be discussed further. Gallium was also removed following a helium ion bombardment (A-1), but resolution of the decay curve was again impossible.

2. Isotopes of Zinc. Information concerning zinc isotopes which were identified among the spallation products of copper is listed in Table 4.

Table 4

Zinc Isotopes Formed in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min. at end of bomb.	Rel. Yield
62	(cf. Sec. III-A-1)		D-1	9.5h		4.1×10^4	0.005
			D-2	9.7h		2.0×10^6	0.036
			D-3	9.5h		5.0×10^6	
63	38m	β^+, γ	D-2	37m		3.6×10^7	0.047
			D-3	36m		1.0×10^8	
65	250d	β^+, K, γ	D-9	(250d)		$6.0 \times 10^{4*}$	0.007

* Assuming 11% counting efficiency

Since a copper fraction was not removed in D-3, the relative yields for this bombardment could not be computed. The chemical yield for zinc in D-1 was known to be low; it is felt that this explains the yield difference for Zn^{62} between D-1 and D-2. The identification of the 250-day Zn^{65} is not certain; it is based on what is presumed to be a specific isolation procedure, and on approximate 250-day half-life over a period of about 150 days.

3. Isotopes of Copper. A summary of the copper isotopes found as spallation products is given in Table 5. The half-life of Cu^{66} (5 min.) is too short to have

Table 5

Copper Isotopes Formed in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min. at end of bomb.	Rel. Yield.
60[13]	24.5m	β^+	D-2	~24m		3.1×10^8	0.28
			D-7	~26m		1.4×10^9	0.31
			A-1	~25m		2.8×10^7	0.18
61	3.4h	β^+, K	D-1	3.4h	+	2.2×10^7**	1.0
			D-2	3.2h		1.7×10^8	1.0
			D-5	3.1h		8.6×10^8	1.0
			D-6	3.2h		2.9×10^8	1.0
			D-7	3.4h		8.0×10^8	1.0
			D-10	3.2h		3.8×10^8	1.0
			A-1	3.3h		3.2×10^7	1.0
			A-2	3.3h		1.5×10^8	1.0
62	10.5m	β^+	D-2	11m		4.8×10^9	2.55
			D-7	10m		1.5×10^{10}	2.05
			A-1	11m		5.1×10^8	2.44
64	12.8h	β^+, β^-, K, γ	D-1	13.6h		3.5×10^6**	0.63
			D-2	13.0h		2.4×10^7	0.59
			D-4	12.5h		3.5×10^7	(0.58)*
			D-5	12.8h		1.2×10^8	0.55
			D-6	12.8h	+,-	4.1×10^7	0.56
			D-7	13.2h		1.1×10^8	0.55
			D-8	12.6h		5.3×10^7	(0.55)*
			D-10	12.7h		5.1×10^7	0.53
			A-1	13.3h		4.2×10^6	0.53
			A-2	12.8h		2.7×10^7	0.70
67[6]	56h	β^-	A-1				≤ 0.0006
			A-2				≤ 0.03

* Assumed values; used as basis for relative yield calculations for other products of these bombardments.

**Assuming 100% counting efficiency.

been detected. No definite evidence for formation of Cu^{67} was found; in A-1 (400 Mev helium ions), the copper activity decreased to zero with no indication of any half-life longer than 13 hrs. In A-2 (200 Mev helium ions), there was an indication of a small amount of activity with a half-life of 60-90 hrs.; the maximum yield quoted in Table 5 was calculated on the assumption that all of this longer-lived activity was due to Cu^{67}.

4. Isotopes of Nickel. The isotopes of nickel (atomic number 28) identified in this work are presented in Table 6, together with the observed yields. The yield of both isotopes in D-7 is lower than in D-1. This might be blamed

Table 6

Nickel Isotopes Formed in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min. at end of bomb.	Rel. Yield
57	36h	β^+	D-1	38h	+	9.0×10^4	0.046
			D-7	37h		1.4×10^6	0.02
			A-1	38h		1.4×10^5	0.05
65 (14)	2.6h	β^-, γ	D-1	2.5h	-	1.4×10^6	0.047
			D-7	2.55h		1.1×10^7	0.012
			A-1	2.6h		2.1×10^6	0.057

on poor chemical recovery in D-7, although the yield ratios are not the same for the two isotopes.

5. Isotopes of Cobalt. The isotopes of cobalt (atomic number 27) which were definitely identified are listed in Table 7. In addition to these isotopes, some long-lived activities (>70 days) were present; Co^{56}(72-day β^+,K),

Table 7

Cobalt Isotopes Formed in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min. at end of bomb.	Rel. Yield
55	18.2h	β^+,γ	D-1	17h	+	1.2×10^5	0.028
			D-7	17.3h		5.8×10^6	0.037
61 (15)	108m	β^-	D-1	100m	-	5.7×10^6	0.14
			D-7	106m		1.8×10^8	0.13

Co^{57}(270-day β^+,K), and Co^{58}(72-day β^+,K) are no doubt contributing. Assuming the long-lived tail to consist of one component of 72-day half-life, and assuming a 50% counting efficiency the approximate relative yield was 0.50. Co^{60} consists of two isomers: a 5.3-year negative beta emitter, and a 10.7-min state decaying by isomeric transition and negative beta emission. The 5.3-year isomer might have been a component of the long-lived activities observed, but the cobalt fraction was never isolated soon enough to have observed a 10.7-minute period.

6. Isotopes of Iron. The activities observed in the iron fractions are listed in Table 8. Fe^{55} (ca. 4-yr. K-capture) has not been observed, probably because of its long half-life and low counting efficiency; the iron fraction from D-9 has decayed with a 48 $\pm$ 1 day half-life for about 230 days without any indication of a longer component.

Table 8

Iron Isotopes Formed in
the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min. at end of bomb.	Rel. Yield
52	(cf. Sec. III-A-2)		D-1	7.6h	+	2.0×10^4	0.002
			D-4	7.8h		3.6×10^5	0.0037
			D-6	8.0h		3.0×10^5	0.0027
			D-7	~8h		9.2×10^5	0.0028
53	8.9m	β^+	D-4	8.6m		2.4×10^8	0.10
			D-6	10m		1.0×10^8	0.04
59	47d	β^-, γ	D-1	47d	-	2.8×10^3	0.042
			D-4	50d		5.8×10^4	0.092
			D-6	~50d		5.2×10^4	0.063
			D-7	50d		2.0×10^5	0.061
			D-9	48d		2.5×10^6	(0.07)*
			A-1	47d		4.8×10^3	0.051

* Assumed value; used as basis for calculating relative yields of other products from this bombardment

7. Isotopes of Manganese. Table 9 summarizes the data obtained on the isotopes of manganese (atomic number 25).

Table 9

Manganese Isotopes Formed in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min. at end of bomb.	Rel. Yield
51	46m	β^+	D-7	45m		1.3×10^8	0.043
			A-1	~50m		3.1×10^6	0.038
52	6.5d	β^+, K, γ	D-1	5.5d	+	6.7×10^4*	0.12
			D-7	5.7d	+	2.0×10^6	0.20
			D-9	6.0d		5.2×10^6	0.074
			A-1	5.8d		9.5×10^4	0.12
54	310d	K, γ	D-9	(310d)		6.3×10^5**	0.09
56	2.59h	β^-, γ	D-1	2.5h	-	3.1×10^6	0.10
			D-7	2.5h		2.1×10^8	0.20
			A-1	2.55h		2.7×10^6	0.07

* Assumed 40% counting efficiency

** Assumed 10% counting efficiency

The identification of the 310-day Mn^{54} is based only on a close adherence to a 310-day half-life for about 170 days, and is therefore uncertain. It should be pointed out that these data all indicate that the half-life of Mn^{52} is less than 6.5 days; the best value is that obtained from A-1; the decay of the Mn fraction in this bombardment followed a 5.8-day half-life for about 30 days, after which the half-period lengthened rapidly to a very high value. Assuming this long-lived activity was the 310-day Mn^{54} and subtracting it from the total activity, a straight 5.8-day half-life over a period of 50 days was obtained. The Mn fraction was never removed sufficiently early to observe definitely the 21-minute isomer of Mn^{52}.

8. Isotopes of Chromium. The data for the two observed isotopes of chromium (atomic number 24) are given in Table 10. Cr^{55} has been reported with various half-lives ranging from 1.6 to 2.3 hrs. In the present work a small

Table 10

Chromium Isotopes Formed
in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min. at end of bomb.	Rel. Yield
49	41.9m	β^+, γ	D-1	41m	+	2.8×10^5	0.003
			D-4	41m		1.2×10^7	0.013
			D-7	39m		2.5×10^7	0.008
			A-1	43m		1.0×10^6	0.009
51	26.5d	K, γ	D-7	27d		1.9×10^5*	0.044
			D-9	27.5d		9.0×10^5	0.022
			A-1	26d		4.1×10^3	0.024

* Assumed 10% counting efficiency, and corrected for amount formed by decay of Mn^{51}

amount of activity of half-life intermediate between 42 min. and 26.5 days was usually observed, but it could not be resolved into any definite periods constant among the several bombardments.

9. Isotopes of Vanadium. Because of the lack of a satisfactory chemical procedure, only one vanadium isotope, V^{48} (16 d β^+, K, γ) has been definitely identified among the spallation products of copper. It was observed in bombardment D-9 and followed a 16.5-day half-life for a period of about 90 days; the particles emitted were shown to have a positive sign. The relative yield was calculated to be 0.05, assuming a counting efficiency of 60%.

10. Isotopes of Titanium. Table 11 summarizes the results obtained on the isotopes of titanium. About two half-lives of decay of Ti^{51} have been followed in D-9, but only one-half of a half-life has been observed in D-10. In both bombardments there was an indication of the presence of a 3-4 day activity; the sign of its radiation was not determined. It is probable that this activity belongs to titanium, and may be due to the isotope at mass 44 or 52.

Table 11

Titanium Isotopes Formed in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min.at end of bomb.	Rel. Yield
45	3.08h	β^+	D-10	3.0h	+	9.5×10^5	0.003
51	72d	β^-	D-9	74d		2.5×10^4	0.001
			D-10	(70d)		2.6×10^3	0.004

11. Isotopes of Chlorine. Information obtained from the chlorine fractions is listed in Table 12. No particles of positive sign could be detected in any

Table 12

Chlorine Isotopes Formed in the Spallation of Copper

Reported in Literature			Experimental Results				
Mass No.	$T_{1/2}$	Radiation	Bomb. No.	$T_{1/2}$	Sign of Rad.	Dis./min.at end of bomb.	Rel. Yield
38	37m	β^-, γ	D-7	38m	-	1.6×10^6	0.00048
			D-8	37.5m	-	7.7×10^5	0.00052
			A-1	36.5m	-	4.0×10^5	0.0033
			A-2	38m	-	3.6×10^5	0.00060
(See text)			D-7	60m	-	2.4×10^5	0.00010
			D-8	59m	-	1.4×10^5	0.00014
			A-1	61m	-	5.7×10^4	0.00068
			A-2	62m	-	4.8×10^4	0.00012

of the chlorine fractions; the sensitivity limit of the magnetic deflector sets the maximum yield of Cl^{34} (33 min.β^+) a factor of 10 to 15 below that observed for Cl^{38}. The decay of all chlorine fractions started with about a 37-38 minute period, but the half-life slowly increased until a half-life of about 60 minutes was reached when the sample became practically inactive; a typical decay curve is reproduced in Figure VIII (Bombardment D-8)*. The ratios of the yields of the 37-minute to 60-minute activities produced in the four bombardments are

* The first part of this curve presents measurements taken on one-seventh of the total fraction. The points for the last 8-10 hours were taken on six-sevenths of the total and then corrected to the basis of the smaller sample.

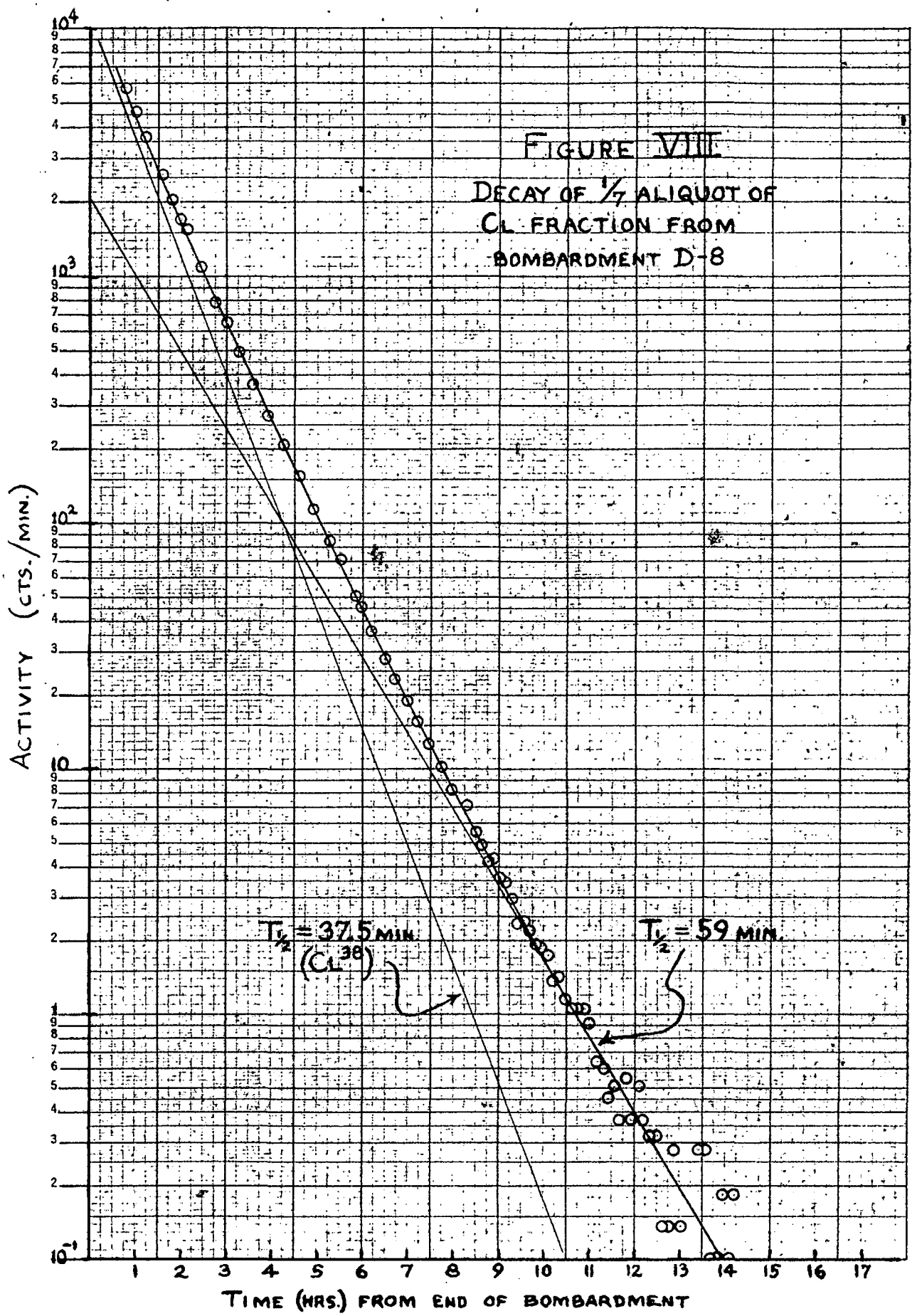


FIGURE VIII
DECAY OF 1/7 ALIQUOT OF
CL FRACTION FROM
BOMBARDMENT D-8
$T_{1/2}$ = 37.5 MIN.
(Cl^{38})
$T_{1/2}$ = 59 MIN.
ACTIVITY (CTS./MIN.)
TIME (HRS.) FROM END OF BOMBARDMENT

4.8, 3.8, 4.9, and 5.0, in the order listed in Table 12. In the same order, the numbers of AgCl precipitations used in the chemical isolation were 1, 3, 1, and 3. Because of the relative constancy of the yield ratios with varying degree of purification of the sample, and because of the high specificity of the chemical procedure, it seems likely that the 60-min. activity belongs to an isotope of chlorine. Since no positive particles were detected in these samples, the 60-minute period might be a negative beta-emitter at mass 39.

12. Isotopes of Phosphorus. A reasonably certain identification of P^{32} (14.3-day β^--emitter) as a spallation product of copper was made. Phosphorus (atomic number 15) was separated and isolated in only bombardment D-9. A half-life of 15 $\pm$ 1 days was observed in the final fraction for a period of about 90 days, after which the activity in the s ample was so low that it could no longer be accurately determined. The activity level was too low to determine the sign of the emitted particles by the magnetic deflector, but an aluminum absorption curve was taken (Figure IX). The statistical uncertainties attached to these counts are large, because of the low counting rate. However, the best curve drawn through the points (neglecting the last one) was subjected to Feather analysis and indicates a maximum energy of about 1.8 Mev. The standard for the Feather analysis was a RaD-RaE sample (range = 480 mg Al/cm^2; E_{max} = 1.17 Mev). The reported values for the maximum energy of the P^{32} beta-particle are in the range 1.69-1.75 Mev. The relative yield of this 15-day activity was 0.0005.

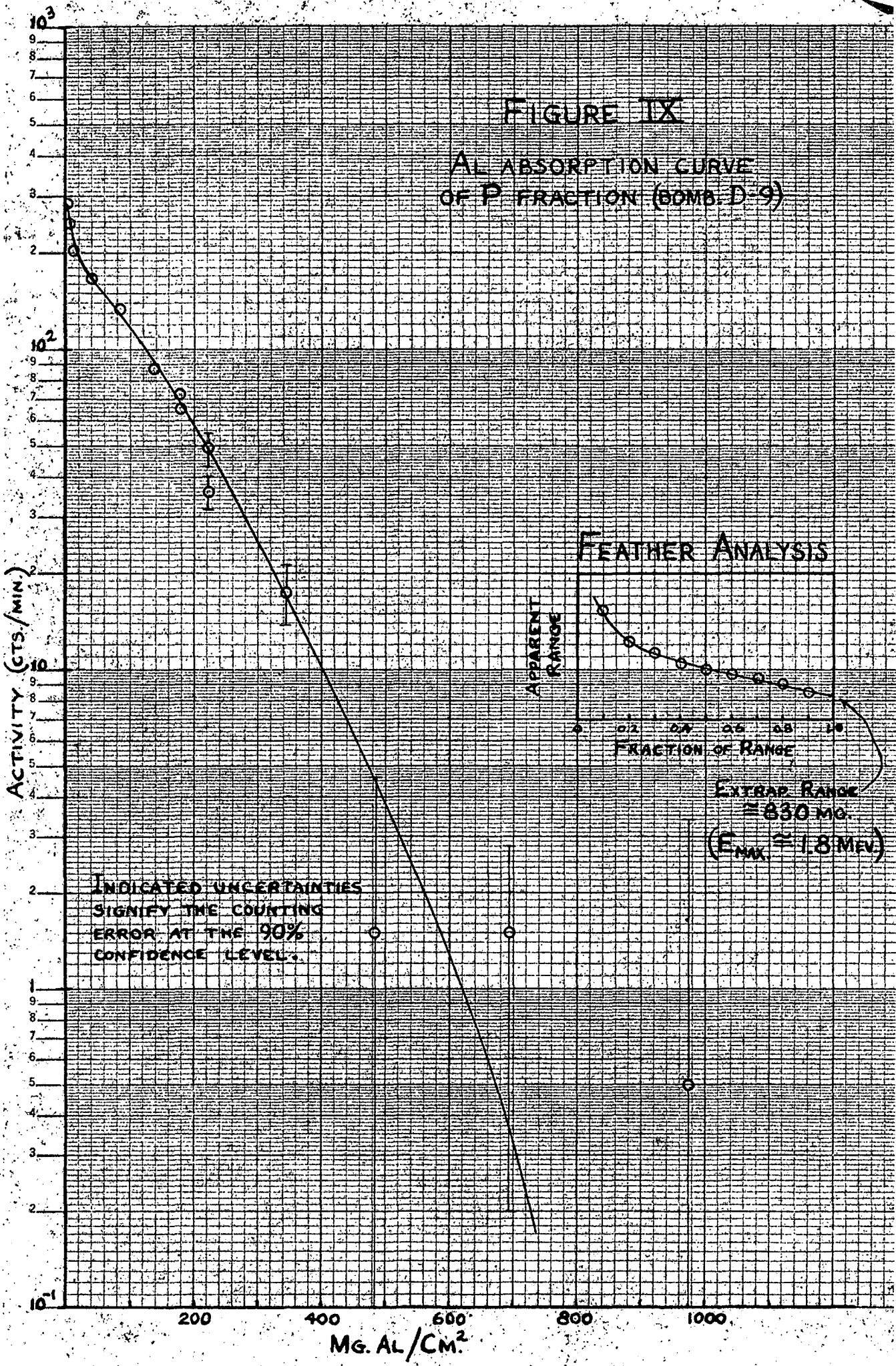


FIGURE IX
AL ABSORPTION CURVE
OF P FRACTION (BOMB. D 9)
FEATHER ANALYSIS
APPARENT RANGE
FRACTION OF RANGE
EXTRAP. RANGE
≅ 830 MG.
($E_{MAX.}$ ≅ 1.8 MEV.)
INDICATED UNCERTAINTIES
SIGNIFY THE COUNTING
ERROR AT THE 90%
CONFIDENCE LEVEL.
ACTIVITY (CTS./MIN.)
MG. AL/CM.²
200
400
600
800
1000

IV. DISCUSSION

A. General Observations

As an aid in the following discussion, a summary of the results presented in the previous section is given in Table 13. In evaluating the average yield for any one isotope, the higher values have been weighted more heavily; this was done mainly on the assumption that the lower figures are due to an abnormally low chemical recovery of the element. It should also be pointed out that while the yield figures for the reaction with 190 Mev deuterons represent averages resulting in practically all cases from two to seven bombardments, the yields for 190 Mev and 380 Mev helium ion reactions were obtained from only one bombardment each.

The wide distribution and large number of radioisotopes formed in the spallation of copper are immediately evident. In the most extensively investigated region, from titanium through zinc, practically all radioisotopes which could have been observed in view of experimental time considerations were actually observed. The exceptions to this statement are the poorly characterized isotope of chromium of mass 55, and Cu^{67} from the helium ion bombardments. In the chlorine and phosphorus fractions, Cl^{34} is the only radioisotope which might have been observed, but was not. The virtual absence of Cl^{34} among the spallation products of copper will be discussed later.

The widespread location of the active spallation products leads one to the assumption that stable isotopes are also formed in good yields as primary products of the spallation reaction. This assumption is substantiated by the probable identification of Mn^{54} among the products; this radioisotope is shielded by two stable isotopes against formation by radioactive decay, and could only be formed directly by spallation. It can be inferred that the neighboring stable isotopes

Table 13

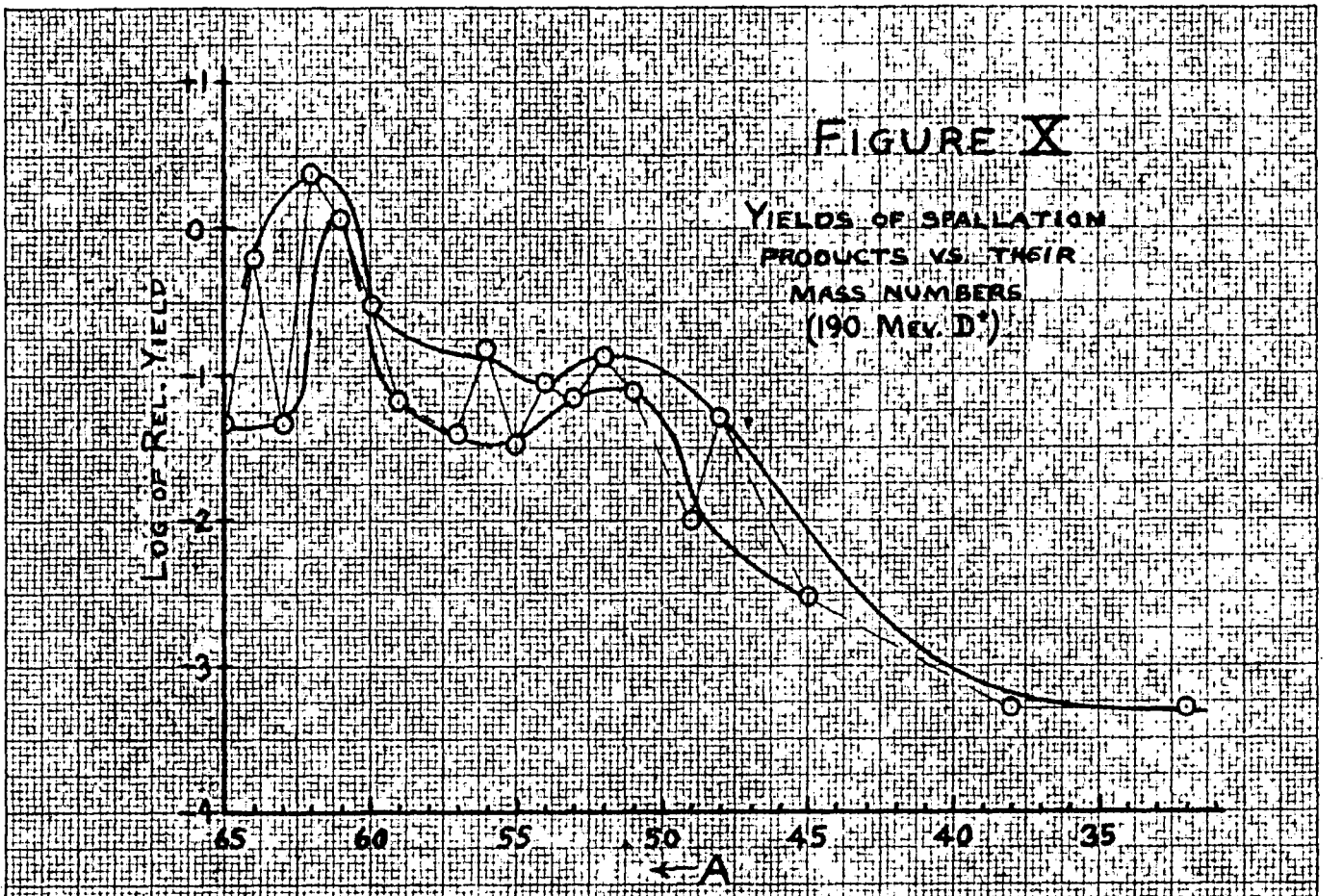
Summary of Isotopes Observed
as Spallation Products of Copper

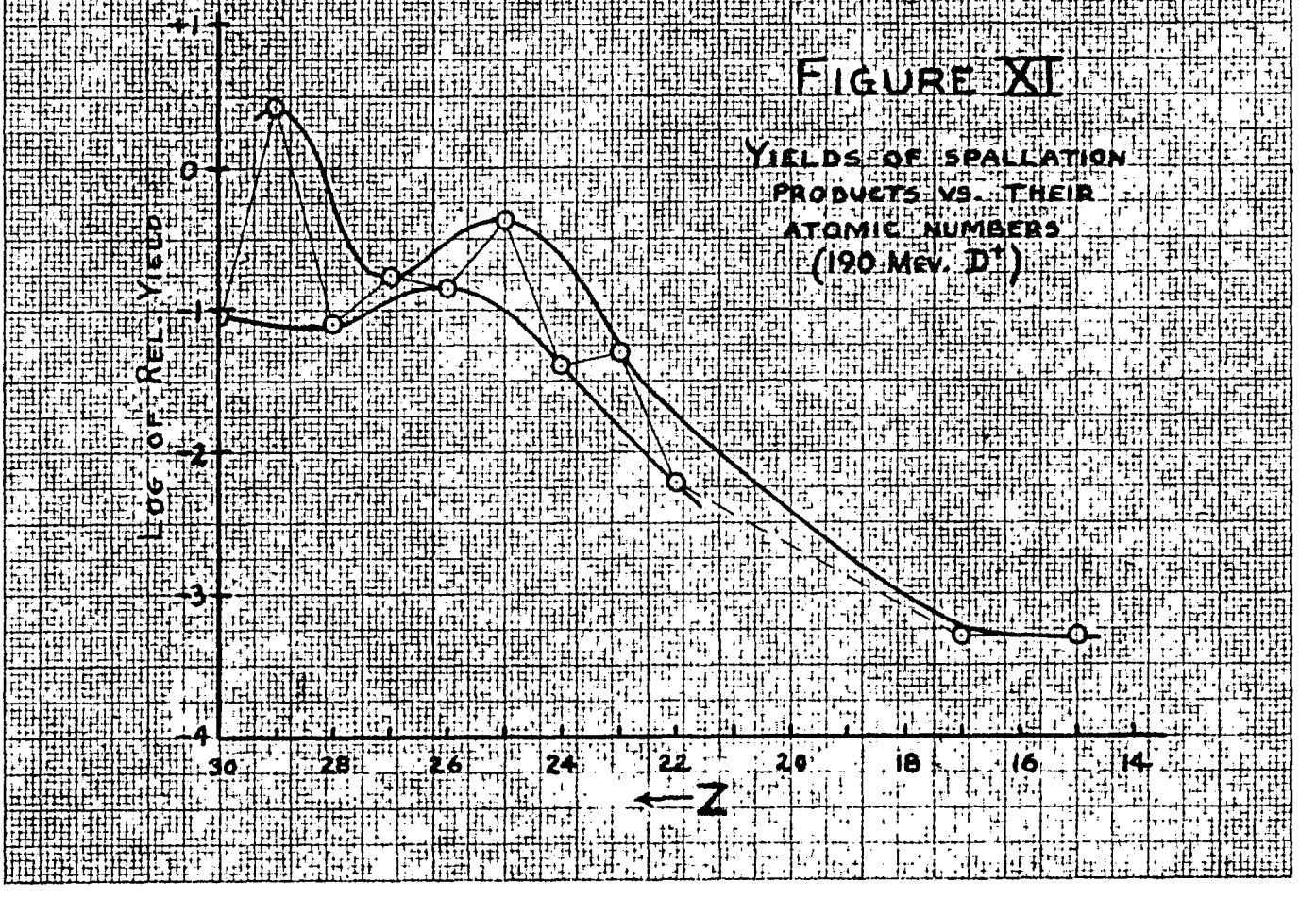
Isotope	Type of Rad'n	Obs. $T_{1/2}$	Average yield relative to Cu^{61}		
			190 Mev D^+	190 Mev He^{++}	80 Mev He^{++}
$_{30}Zn^{62}$	(K)	9.5h	0.036	—	—
Zn^{63}	β^+	37m	0.047	—	—
Zn^{65}	β^+,K	(250d)	0.007	—	—
$_{29}Cu^{60}$	β^+	25m	0.30	—	0.18
Cu^{61}	β^+,K	3.3h	1.0	1.0	1.0
Cu^{62}	β^+	11m	2.3	—	2.4
Cu^{64}	β^+,β^-,K	13h	0.57	0.70	0.53
Cu^{67}	β^-	—	—	<0.03	<0.0006
$_{28}Ni^{57}$	β^+	38h	0.04	—	0.05
Ni^{65}	β^-	2.55h	0.04	—	0.057
$_{27}Co^{55}$	β^+	17h	0.033	—	—
Co^{61}	β^-	103m	0.14	—	—
$_{26}Fe^{52}$	β^+	7.8h	0.003	—	—
Fe^{53}	β^+	9m	0.07	—	—
Fe^{59}	β^-	48d	0.067	—	0.051
$_{25}Mn^{51}$	β^+	47m	0.043	—	0.038
Mn^{52}	β^+,K	5.8d	0.13	—	0.12
Mn^{54}	K	(310d)	0.09	—	—
Mn^{56}	β^-	2.5h	0.15	—	0.07
$_{24}Cr^{49}$	β^+	41m	0.01	—	0.009
Cr^{51}	K	27d	0.03	—	0.024
$_{23}V^{48}$	β^+,K	16.5d	0.05	—	—
$_{22}Ti^{45}$	β^+	3.0h	0.003	—	—
Ti^{51}	β^-	74d	0.003	—	—
$_{17}Cl^{38}$	β^-	37.5m	0.00050	0.00060	0.0033
$_{15}P^{32}$	β^-	15d	0.0005	—	—

are formed in approximately the same yield. Moreover, inspection of the results for any one of several elements listed in Table 13 reveals that the isotope of that element having the largest neutron deficiency is always formed in the lowest yield, and that in general the yield increases as the mass of the isotope approaches the mass of the stable isotope(s). Extrapolation of this effect into the stability region again suggests that the stable isotopes are formed in good yields.

The total relative yield for all isotopes detected as products of 190 Mev deuteron bombardment is about 5.16. This is equivalent to a cross-section of about 0.31×10^{-24} cm^2. This figure represents observations on 25 radioisotopes; a total of 70 - 75 fairly well characterized isotopes (radioactive plus stable) exist for the elements in question. If all undetected isotopes were formed in about the same yield as those isotopes near them which were detected, a total cross section close to the geometrical cross-section (1.1×10^{-24} cm^2) is indicated. This is a logical result and further substantiates the assumption that stable isotopes are formed as primary spallation products in yields of the same order of magnitude as the radioisotopes.

The overall trend in yields of the various products shows a general decrease with decreasing mass and charge of the products. However, there are interesting variations in this trend which will be pointed out now and discussed more fully later. The variation of yield with the mass number of the product, i.e., with the difference in the total number of nucleons between target and product nuclei, is plotted in Figure X. Where several products of the same mass number were observed, the total yield for the several isobars was used. Figure XI shows the variation of yield with the atomic number of the product, i.e., with the difference in the number of protons between target and product nuclei,





the total yield of all the observed radioisotopes of each element is plotted against the atomic number of that element. Both of these figures were constructed from the data of Table 13 for the 190 Mev deuteron bombardments. Although these graphs represent only data for the observed products, it is felt that the general trends that they portray would not be appreciably altered if yield data for all possible product isotopes were known and included.

A striking feature of both these curves is the high yields of products whose nuclei differ from the target nuclei by only several nucleons. About 85% of the total yield is made up of products with masses 60-65, and about 80% of the total yield is represented by copper isotopes. These observations are difficult to interpret in terms of the exact number of neutrons and/or protons ejected from the target "nucleus", since there are two target nuclei, Cu^{63} and Cu^{65}.

There is no appreciable general decrease in yield for the products from mass 59 down to about 50. The yields for consecutive mass numbers alternate, with an even mass number showing higher yield than either of its neighboring odd mass numbers. Because of this alternation, the points in Figure X representing even mass numbers have been joined by one smooth curve, and those for odd mass numbers by another. Both these curves then show similar properties: maxima at $A = 61$ or 62; a plateau at $A \cong 59$ to $A \cong 50$, followed by an interpolated sharp yield decrease (through a factor of about 100) to $A \cong 40$; and perhaps a second relatively level portion around $A = 35$. On the basis of the accuracy of the present data, the apparent maxima around $A = 51$-52 are not necessarily significant, although they might be real.

The total elemental yields also show an alternation (Fig. XI) and again two curves are drawn, one for odd Z and one for even Z; the former curve lies higher than the latter. At least a part of the reason for the higher observed yields of the odd-Z elements may be the fact that they possess only one or two

unobserved stable isotopes, while the even-Z elements possess 4 or 5 stable isotopes, and thus a larger fraction of the odd-Z isotopes were actually observed. This does not seem to be the complete explanation, however; from Z = 22 to Z = 30 the upper curve represents identifications of 42% of possible known product isotopes (stable plus active), and the lower curve 33%. The two curves differ by an average factor of about 5 in yield. Moreover, in the isobaric pairs Fe^{52}-Mn^{52}, Mn^{51}-Cr^{51}, and Zn^{62}-Cu^{62}, the member of the pair with odd atomic number is independently formed in the higher yield. Figure XI also shows, after the initial maxima, a plateau region from Z = 28 to Z ≅ 25; the apparent maxima at Z = 26 and 25 again may or may not be significant. The yields then decrease with decreasing atomic number.

The following general summary can be presented. The ejection of only several nucleons from the compound and/or excited nucleus is a very probable event, leading to very high yields of isotopes with mass numbers 60-65, and especially those which are copper isotopes. Products with masses about 5-15 units less than the target mass (and charges of about 1-4 units less) are formed in lower yields, but there is no consistent decrease in yield with decreasing mass or charge in this region. There appears to be a relatively rapid decrease in yield with increasing number of nucleons ejected from the excited nucleus, starting at the emission of about 15 nucleons. In general, an isotope of mass about 30 units less than that of the target is formed in a yield of about one-hundredth of an isotope whose mass is about 5-15 units less; an isotope in this latter region is produced in a yield of about one-tenth of the average yield of an isotope with a mass within 5 units of the target isotope.

These results have been compared with those obtained by the bombardment of $_{33}As^{75}$ with 190 Mev deuterons[3]. In general, the same overall trends were obtained with arsenic as with copper. To some extent, the same high-low alterna-

tion of yields with consecutive mass or atomic numbers was observed with arsenic, but this effect was not as consistent as with copper.

D. Comparison of Results of Deuteron Bombardments with those of Helium Ion Bombardments.

Since fewer helium ion bombardments were conducted, any comparison of the effects of deuterons and helium ions cannot be extensive. In general, the yields of an isotope formed by the two types of bombardments would have to differ by at least a factor of two before they could be termed significantly different. With this limitation, the results of the 380 Mev helium ion bombardment were the same as those of the 190 Mev helium ion bombardments, with one exception: six times as much Cl^{38} was formed by 380 Mev helium ions as by 190 Mev deuterons. With 190 Mev helium ions, however, the yield of Cl^{38} was about the same as with 190 Mev deuterons. The significance of these differences will be discussed in the following section.

C. Mechanisms of High Energy Nuclear Reactions

The accepted and experimentally supported theory of nuclear reactions at low energies ($<$40 Mev) involves the formation of a compound, excited nucleus; the incident particle is captured by the target nucleus to form a different compound nucleus possessing an excitation energy approximately equal to the kinetic energy of the incident particle. This excited nucleus then rapidly boils off particles and energy and becomes the product nucleus. It will be interesting to consider on this basis the excitation function of a particular reaction leading to a particular product, i.e., the effect of incident particle energy on the yield of a particular nucleus. As the excitation energy is increased, the yield will go through a maximum at which the excitation is most appropriate for emitting the required number of particles. The yield should then decrease rapidly as the energy becomes higher, since the evaporation of more particles from the excited nucleus will be much more probable.

Using only the picture of the compound nucleus presented above, one would therefore expect that bombardment with particles of several hundred Mev of energy would lead to very low yields of isotopes which are within several mass units of the target isotope. On the contrary, however, the data presented in the preceding sections show that a large majority of the reactions of copper with 190 Mev deuterons and 380 Mev helium ions lead to product isotopes which differ from the target isotopes by a loss of only several nucleons. Clearly, then, a different picture of nuclear reactions at high energies is needed.

Serber[16] has recently suggested a mechanism which very satisfactorily explains the observed facts. According to this theory, bombardment with 200 Mev deuterons or with 400 Mev helium ions can be regarded as a simultaneous bombardment by the several individual nucleons (neutrons and protons) making up the incident particle; the binding of the nucleons in the incident particle is important mainly in causing a spatial correlation between them. Serber points out that the collision time between a high-energy incident particle and a particle in the nucleus is short compared to the time of collision of the particles in the nucleus, suggesting that collisions between incident particles and the individual particles in the nucleus must be of primary importance. This leads to two points: (1) at sufficiently high energies the nucleus begins to become transparent to the incident particles; (2) the incident particle loses only a small fraction of its energy to the s truck particle. The kinetic energy transfer to the struck particles is estimated to be about 25 Mev, and the struck particle may either escape from the nucleus without further collision, if it was located near the edge, or it may collide with other nuclear particles and distribute its energy among them. The nuclear excitation energy could then be dissipated by evaporating off several particles with several Mev kinetic energy each.

Serber[17] has also proposed and demonstrated a second mechanism applicable to high energy nuclear reactions. In this process, the proton (or neutron) in the high energy deuteron strikes the edge of a nucleus and is stripped off, while the neutron (or proton) misses and continues on its way. The target nucleus is therefore excited to the extent of an average of about 100 Mev, and this excited nucleus differs from the target nucleus in that it contains an additional proton or neutron. This compound nucleus could then dispel its excitation energy by an evaporation process. The cross section for this "stripping" process is $\sigma = \frac{1}{2}\pi R R_d = 5A^{1/3} \times 10^{-26}\ cm^2$, where R is the nuclear radius, R_d is the deuteron radius, and A is the mass number of the target.

In addition to the preceding two paths by which high energy nuclear reactions may proceed, it is also conceivable that a very small fraction of the reactions might proceed by capture of the entire incident particle, producing a highly excited (200 or 400 Mev) compound nucleus which would dissipate its energy by boiling off a large number of particles. To summarize, the reaction of 200 Mev D^+ or 400 Mev He^{++} on a target Z^A might proceed via three different paths:

(1) inelastic collisions, exciting Z^A to the extent of about 25 Mev;

(2) the stripping process, producing a compound nucleus $(Z + 1)^{A + 1}$ or $Z^{(A + 1)}$ with about 100 Mev excitation energy;

(3) amalgamation of the entire incident particle with the target nucleus to produce a compound nucleus $(A + 1)^{A + 2}$ (for deuterons) or $(Z + 2)^{A + 4}$ (for helium ions), possessing excitation energy of 200 Mev or 400 Mev, respectively.

It is also possible that a small fraction of the nuclear reactions proceed by mechanisms which could not be classed exclusively as any one of the above, but which might be described as something in between two of them.

The results obtained in the investigation of the spallation of copper will now be examined in the light of the above three mechanisms. The excitation energy of 25 Mev obtained by mechanism (1) is sufficient to boil off about three nucleons; the maximum yield due to this effect should therefore be observed at masses 60-62 (from Cu^{63} and Cu^{65}). It was shown in Figure X that a high maximum was actually observed at masses 61-62, suggesting that these products arose from the inelastic collision mechanism. The stripping process, mechanism (2), should lead to product nuclei containing of the order of 10-15 nucleons less, on the average, than the target nuclei. Figure X shows a possible maximum at mass numbers 51-53, representing a loss of about 12 nucleons from the target nuclei. It therefore seems logical to identify the products in the mass region of about 50 to 58 as those which arose mainly by the stripping process. The product nuclei far removed from the target nuclei, i.e., representing losses of 20 or more nucleons, probably arise from mechanism (3). This contention is supported by the observed yields of Cl^{38} following bombardment with 190 Mev deuterons (95 Mev per nucleon) and 190 Mev (47.5 Mev per nucleon) and 380 Mev (95 Mev per nucleon) helium ions. The yields from 190 Mev deuterons and 190 Mev helium ions are practically the same, while the yield from 380 Mev helium ions is a factor of six higher. This indicates that Cl^{38} is formed from a compound nucleus which has absorbed the entire energy of the incident particle. If it arose from a compound nucleus formed from just one of the nucleons of the incident particle, as in the stripping process, the yield would have been the same with 190 Mev deuterons and 380 Mev helium ions, and lower with 190 Mev helium ions. That mechanism (3) does not contribute appreciably to the yield of products around the target nuclei is indicated by the failure to obtain evidence for the formation of Cu^{67} following helium ion bombardment. This isotope can be formed only by an $(\alpha,2p)$ reaction

on Cu^{65}. It is interesting to note that although no definite evidence for the presence of Cu^{67} was obtained from either helium ion bombardment, the maximum yield which could be set for the 190 Mev bombardment was 50-fold higher than that for the 380 Mev bombardment.

The yield curve (Fig. X) actually obtained, then, could be formed by the addition of three separate portions, one for each of the three mechanisms just discussed: (1) a relatively sharp maximum at A = 61-62; (2) a somewhat broader maximum at A = 51-53; and (3) a low, broad portion detectable by itself only at $A \lesssim 45$. Corresponding characteristics can also be discerned in Figure XI. The present data indicate that about 80% of the nuclear events occur by the inelastic collision mechanism, about 20% by the stripping process, and less than 1% by amalgamation of the entire incident particle with the target nuclei. It is interesting to note that the stripping cross section for copper, calculated according to Serber[17], is about 0.2×10^{-24} cm^2, which is about 20% of the geometrical cross section.

Regarding the types of nucleons emitted from the nuclei excited in the three different ways, the data indicate that a nucleus excited by inelastic collision preferentially emits neutrons rather than protons, as illustrated by the high yields of copper isotopes. Neutron emission also seems to be more probable following excitation by the stripping process; the second maxima in Figures X and XI indicate the most probable combination of emitted particles is about 4 protons and 8 neutrons. These may more likely be ejected as 2 alpha-particles plus 4 neutrons. A nucleus excited by mechanism (3) would probably emit as many of its particles as possible in the form of alpha-particles, since this is energetically more economical than emission of only neutrons and protons. Apparently very few excited nuclei possess sufficient energy to emit four neutrons in addition to the number of particles required to form Cl^{38}, since Cl^{34} could not be detected.

The data also hint that the emission of almost exclusively neutrons (or protons) from a nucleus excited by the stripping process, followed by a series of rapid positron (or negative beta-particle) decays is a relatively rare event. If this did occur to an appreciable extent, the daughter member of isobaric pairs would be formed almost exclusively by decay of the parent member; the opposite effect was observed in two or three cases. The appreciable yield of the shielded Mn^{54} contributes to the argument. The conclusion is that most of the primary products formed following excitation by stripping are those which are close to the region of stable nuclei.

The reasons for the "saw-tooth" character of the yield curves (Figures X and XI) have not been discussed and are not immediately evident. To explain the fact that the total yield for elements of odd Z is higher than the total yield for either neighboring element of even Z, let us consider first only those products formed mainly as a result of the stripping process. One might first assume that mainly alpha-particles and neutrons are emitted by the excited nucleus. Then if one assumes that capture of a stripped neutron to form this excited nucleus is more probable than capture of a stripped proton, the products with odd Z would be more abundant than those with even Z. Considering the products differing from the target nuclei by only a few nucleons, zinc isotopes would be expected in lower yields than copper isotopes, since the former can arise not from the relatively probable inelastic collision mechanism, as do the copper isotopes, but only through the formation of a compound nucleus (mainly by the stripping process). Since the excitation energy induced by stripping is sufficiently high so that emission of less than 5 neutrons following proton capture would be a relatively improbable event, one would expect relatively low yields of zinc. The excitation functions of several zinc isotopes in the deuteron bombardment of copper have

been investigated by Bockhop, Helmholz and Peterson[18]; their results confirm this picture, and also indicate that nickel and cobalt isotopes arise mainly from excitation by inelastic collision.

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